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X-ray imaging of transplanar liquid transport mechanisms in single layer textiles

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ABSTRACT: Understanding the penetration of liquids within textile fibers is critical for the development of next-generation smart textiles. Despite substantial research on liquid penetration in the plane of textile, little is known about how the liquid penetrates in the thickness direction. Here we report a time-resolved high-resolution X-ray measurement of the motion of the liquid-air interface within a single layer textile, as the liquid is transported across the textile thickness following the deposition of a droplet. The measurement of the time-dependent position of the liquid meniscus is made possible by the use of ultra-high viscosity liquids (dynamic viscosity from 10^5 to 2.5×10^6 larger than that of water). This approach enables imaging due to the slow penetration kinetics. Imaging results suggest a three-stage penetration process with each stage being associated with one of the three types of capillary channels existing in the textile geometry, providing insights into the effect of the textile structure on the path of the three-dimensional liquid meniscus. One dimensional kinetics studies show that our data for the transverse penetration depth Δx_L vs. time do not conform to a power law, and that the measured rate of penetration for long times is smaller than that predicted by Lucas-Washburn kinetics, challenging commonly held assumptions regarding the validity of power laws when applied to relatively thin textiles.

INTRODUCTION

The prediction of liquid transport in textiles plays an important role in many applications, from the design of liquid repellent textiles for skin protection ^{1, 2}, to the development of resin impregnation methodologies ^{3, 4}, and the manufacturing of textiles with improved wearability ^{5, 6}. Classical problems in this field are the prediction of wicking rates in experiments where the textiles are put in contact with large liquid volumes ⁷, the analysis of the liquid distribution due to moisture condensation ⁸, and the prediction of in-plane and transplanar liquid transport following drop deposition ^{9, 10}. In this paper we focus on the last problem.

The process of liquid wicking in a textile when a drop is deposited on its surface can be conceptually described as composed of two stages, namely transplanar penetration (also called transplanar uptake ¹¹ or transplanar wicking ^{5, 12}) and in-plane spreading ¹³. The first stage refers to liquid transport normal to the plane of the textile, and occurs temporally before in-plane spreading (although the two mechanisms can be simultaneous in the early stages of spreading). While substantial knowledge has been accumulated in understanding issues of in-plane transport ^{7, 9, 14, 15, 16, 17, 18, 19, 20}, investigations on the mechanisms of transplanar transport are scarce ^{5, 11, 12}. Transplanar penetration is a relatively fast process ²¹, and is there-

fore often neglected when examining the long-time behavior of the liquid.

In the case of single layer textiles, spontaneous transplanar liquid penetration (i.e., with no imposed pressure on the liquid) has so far been limited to approximate measurements with the so-called “demand wettability” method ¹¹. In this method, the fabrics are placed horizontally directly in contact with either a large tank of liquid, or a thin paper fully saturated by liquid. The liquid uptake is recorded by measuring the weight loss of the liquid source with a microbalance. In refs. [11] and [12] the mass of absorbed liquid was reported to vary approximately linearly with time, and then saturate to a constant value. The demand wettability method does not provide data that can aid the analysis of microscopic transport mechanisms.

For multilayer textiles, Bencsik et al. ²² employed Nuclear Magnetic Resonance (NMR) to visualize the oil distribution in a three-layer textile as a function of an applied compressive load. The volume of oil in the textile was analyzed as a function of the coordinate normal to the plane of the textile for different compressive loads. The three-dimensional distribution of the liquid within the textile was not considered. It should be emphasized that liquid penetration driven by a pressure gradient follows a different dynamic from the liquid penetration driven by a capillary pres-

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sure. Transplanar wicking in a two-layer knitted textile from a reservoir of liquid being continuously supplied to the textile via a capillary tube was studied by Birrfelder et al.⁵ using X-ray radiography. Two-dimensional cross sections of the textile were presented showing the liquid front in both layers. A mechanism of liquid transport was proposed whereby intra-yarn pores were responsible for transplanar penetration of liquid within one layer while large inter-yarn pores were associated with layer-to-layer transport. However the paper did not present quantitative information on the motion of the liquid front. Transplanar wicking in a three-layer textile from a reservoir of liquid was studied using X-ray radiography by Rossi et al.²³ The water distribution in the textile was characterized by the attenuation intensity of the X-rays. The work focused on the amount of water being absorbed by individual layers. No description of microscopic transport was attempted. X-ray tomography was used by Weder et al.²⁴ to investigate transplanar wicking in a four-layer woven textile. Water distribution as a function of the thickness of the material was presented. This paper focused primarily on monitoring the amount of liquid in each layer over time rather than observing the liquid front movement. The spatial resolution was relatively coarse, with pixel sizes of about $70\mu\text{m}$.

Data for transplanar liquid penetration is available for relatively thin porous media other than textiles. Bayramli and Powell studied transplanar penetration in carbon fiber tows²⁵. A carbon fiber tow is geometrically similar to a textile yarn, as it is formed by concentric layers of packed carbon fibers oriented along the tow axis. The liquid penetration depth normal to the fibers was measured by examining the weight of the wet tow, and found to be proportional to the square root of time, as expected from the Lucas-Washburn kinetics¹⁶. Data on transplanar liquid penetration across paper is available in the publication from Karppinen et al.²⁶, where for the transplanar transport due to the deposition of a droplet was analyzed. The authors measured the intensity of diffuse light reflection from the paper. The light reflection curve showed a monotonic decrease with time and an asymptotic plateau. The penetration depth was not characterized and no attempt to model the data was made. A simulation of drop penetration in a paper layer of thickness larger than the drop diameter was carried out by Hyväluoma et al.²⁷. The transplanar liquid penetration depth was found to be much smaller than the prediction by the Lucas-Washburn law. Although the structure of paper has some similarity to that of a textile, paper lacks the hierarchy of pore scales that characterizes the structure of textiles. In particular, paper is characterized by a marked scale separation between the size of the largest pores in the porous structure and the thickness of the paper layer. Such separation of scales is not ensured in textiles, which have relatively large pores.

The issue of scale separation also relates to the distinction between single and multilayer textiles. A sufficiently thick multilayer textile can be considered a bulk, three-dimensional porous medium sample. It is therefore expected that the long time capillary wicking behavior for liquid penetration in a thick textile follows continuum theories, in contrast for a single layer textile, the time taken for the fluid to cross the textile thickness may not be enough time for an asymptotic behavior to be established.

From the point of view of the development of theoretical models, there is large uncertainty regarding the three-dimensional distribution of liquid within porous layers when a drop is placed on them. The theoretical model of Patnaik et al.¹⁷ for liquid penetration in a single layer textile assumes that transplanar penetration occurs uniformly under the droplet, and that the liquid front is planar. In contrast, the model of Starov et al.²⁸, developed to analyze liquid penetration in millimetric glass and metal filters, assumes that the advancing liquid front inside the material is approximately hemispherical. In these studies the microscopic, three-dimensional distribution of the liquid is not analyzed, and a continuum approach is taken. The model of Gillespie for spreading in paper¹⁴ neglects the initial stage of transplanar penetration. The lack of experimental data that could aid the development of theoretical models is one of the main motivations for the current work.

Experimental challenges in measuring transport by Nuclear Magnetic Resonance, Neutron Radiography and X-ray Computed Tomography (XCT) include loss of signal over long experimental duration and insufficient spatial and temporal resolution. Measuring transplanar liquid transport is particularly challenging, as for low viscosity liquids the time scale of liquid penetration across the textile layer is small (the required time resolution for water is typically below 0.1s ²¹)

In this paper, to overcome time resolution limitations, we use silicone fluids with ultra-high viscosities to slow down the penetration kinetics and enable time-resolved XCT imaging. In contrast to all previous work on single-layer textiles, and most work on multilayer textiles, we report the three-dimensional, microscopic distribution of the liquid within the textile, and discuss potential transport mechanisms that account for the multiscale structure of the textile.

MATERIAL AND METHODS

High viscosity silicone oils (100% polydimethylsiloxane) were used to achieve direct imaging of the transplanar penetration. The viscosities are $\eta = 97\text{Pa}\cdot\text{s}$ (Sigma Aldrich) and $\eta = 2.4\cdot 10^3\text{Pa}\cdot\text{s}$ (Clearco Products). Both oils are non-volatile at room temperature. The oil-air interfacial tension is reported by the companies to be in the range of $19 - 21\text{mN}\cdot\text{m}^{-1}$.

1 PDMS displays shear thinning behavior at sufficiently
2 large values of the shear rate²⁹. In our case, because
3 of the relatively small shear rate values characteristic
4 of flow within the textile (we estimate $\dot{\gamma} \sim \frac{Y}{\mu d} \cong 1 -$
5 10 s^{-1}), non-Newtonian effects are neglected in the
6 analysis.

7 The plain woven textiles are produced by yarns in two
8 orientations aligned at 90° to one another. Fibers in
9 the yarns are cotton and polyester mixed at a ratio of
10 19:1³ (95% cotton). The typical dimensions of the tex-
11 tile are: thickness $h \sim 300 \mu\text{m}$; inter-yarn pore size $150 \cdot$
12 $20 \mu\text{m}^2$, inter-fiber pore size $d_i \sim 3 \mu\text{m}$, fiber diameter
13 $d \sim 10 \mu\text{m}$.

14 For the sample preparation, textile is cut into square
15 sections with approximate side length of 5mm and
16 mounted vertically on a standard XCT bin using ad-
17 hesive. Due to the extremely high viscosity, small
18 amount of both oils are manipulated with needles to
19 form droplets and carefully transferred to the sample
20 surface. The droplet height is on the same order of
21 magnitude as the textile thickness.

22 XCT was carried out only with the highest viscosity
23 liquid ($\eta = 2.4 \cdot 10^3 \text{ Pa} \cdot \text{s}$) using an X-ray microscope
24 (520 Versa, Carl Zeiss, USA), illustrated in Fig. 1. A
25 $2.6 \mu\text{m}$ isotropic voxel size was achieved from imaging
26 samples using a total of 601 projections across 360
27 degrees of sample rotation. Each projection was col-
28 lected using a 0.5s exposure time. The obtained 2D
29 projections were reconstructed to a 3D volume using
30 a filtered back projection algorithm implemented in the
31 manufacturer's software. A standard Shepp-Logan
32 filter, Gaussian filter (strength of 0.5) and beam hard-
33 ening correction (strength of 0.05) was applied to the
34 projections. The imaging conditions allowed the
35 shortest imaging time possible while maintaining a
36 relatively high spatial resolution. The 70-minute ex-
37 perimental duration allowed the collection of three
38 tomography scans.

39 Radiography images were obtained by X-ray beam
40 transmission along the y axis and through the sample
41 to the detector as indicated in Fig. 1. The radiography
42 projections offer a superimposed view of all the xz
43 planes along the y axis thus enable real-time monitor-
44 ing of the liquid front within the textile. Pixel size,
45 number of projections, exposure and waiting time be-
46 tween projections were set to $1.296 \mu\text{m}$, 1200, 4s and
47 1s respectively.

48 The shift of the X-ray intensity profiles at successive
49 times shows the displacement of the spatially aver-
50 aged advancing liquid front in the textile in the x direc-
51 tion. To compute the displacement, intensity threshold
52 values 115, 120, 122, 125 and 126 were selected. For
53 each threshold value, the displacement Δx_L is calcu-
54 lated using the equation: $\Delta x_L = x_t - x_0$, where x_t is

the coordinate of the advancing liquid front at t and x_0
is the initial coordinate of the liquid front at $t = 0\text{s}$.

The obtained XCT slices from the resulting three di-
mensional reconstruction is segmented into liquid and
textile (including background) phases by machine
learning-based segmentation (Weka, 3.9.0, ImageJ,
U. S. National Institute of Health, Bethesda, Maryland,
USA) to render the liquid distribution in the textile
more evident. By coloring the liquid, air and textile
phase red, green and purple respectively in the seg-
mentation results, we are able to clearly visualize the
time-evolved liquid distribution in the textile. In addi-
tion, when only the liquid phase needs to be present-
ed, the liquid phase is shown in black while the textile
and air phases are shown in white for a good con-
trast.

To classify image points as belonging to textile, air, or
liquid we use a Weka learning scheme³⁰, as imple-
mented in ImageJ. Briefly, the Weka learning scheme
is based on applying a series of filters, such as edge
detectors (Laplacian and Sobel, Gabor etc.) texture
filters (minimum, maximum, median, entropy etc.) and
noise reduction filters (Gaussian, Kuwahara, Lipschitz
etc.) to an original image. The filtered images form an
"image pipeline", representing different image fea-
tures. Pixel samples are manually labelled in the orig-
inal image and subsequently the labels will be applied
to the corresponding pixels from the images in the
pipeline. The pixel values will then give a range for
the liquid, air and textile phase which are then used
as training sets to make a classifier. Applying the
classifier to images to be segmented will classify the
pixels automatically and then present the liquid, air
and textile phase in different colors.

RESULTS AND DISCUSSION

Our sample is characterized by an ordered multiscale
structure whose main geometric features are
illustrated in Fig. 2. The inter-fiber pores are the
smallest pores within the textile. The textile is formed
by relatively thick (diameter $\sim 100 \mu\text{m}$) yarns disposed
in a mesh-like arrangement. Each yarn is composed
of small diameter fibres (diameter $\sim 10 \mu\text{m}$) twisted
around the yarn's central axis. In the following text we
will denote the relatively large (linear dimension \sim
 $100 \mu\text{m}$) pores of the textile as inter-yarn pores.

The shape of the center line of each yarn in the out-
of-plane direction is sinusoidal. Each yarn first passes
above a perpendicular row of yarns and then below
the successive one. This arrangement yields a sad-
dle-shaped contact region, whose projection on the y -
 z plane is indicated in Fig. 2 by a dark, dashed region.
Because of the irregular surface of each yarn, the
contact region has a finite maximum thickness of
roughly $10 \mu\text{m}$ (which in turn is of the order of the fiber
size). The inter-fiber pores, *i.e.* the small interstices
within the yarns, have a typical diameter of

1 about $3\mu\text{m}$. This is slightly smaller than each fiber di-
2 ameter. We have examined the diameters of the ca-
3 pillary pores and of the fibers in the XCT images by
4 averaging over 15 fibers for each diameter value re-
5 ported.

6 Fig. 3 shows front (yz) and side (xz) views of the
7 droplet outside of the textile. The droplet spreads lat-
8 erally while simultaneously penetrating in the normal
9 direction. The motion of the air-liquid-solid contact line
10 is relatively slow, and is hindered by the large out-of-
11 plane roughness of the textile (which results from the
12 wavy shape of the yarns) as well as by the presence
13 of fibers that “stick” out of plane, piercing the droplet.
14 Out-of-plane fibers constitute unavoidable structural
15 features that are due to the rupture of fiber bundles.
16 This rupture is the result of the weaving process or
17 the repeated manual handling of the textile sample.
18 The front view in Fig. 3 shows that the shape of the
19 droplet projected onto the yz plane is approximately
20 circular for $t = 25$ mins. At later times the projection of
21 the contact line onto the yz plane tends to assume a
22 polygonal shape. This feature is likely to result from
23 the triple line conforming to the regular microstructure
24 of the textile, as suggested by studies of droplet
25 spreading on microfabricated surfaces formed by reg-
26 ular arrays of pillars³¹.

27 Fig. 4 shows side (xz) views of the liquid within the
28 textile for $\eta = 2.4 \cdot 10^3 \text{Pa} \cdot \text{s}$, focusing on the inter-yarn
29 pore region. To render the distribution of the liquid
30 more evident, Weka segmentation (see Material and
31 Methods section) was used to associate different col-
32 ors to the liquid, air and solid phases. For $t = 25$ mins,
33 the inter-yarn pore is filled with liquid for 3/4 of its
34 depth. For the remaining 45 minutes, the motion of
35 the liquid-air interface in the inter-yarn pore region is
36 almost negligible. The dynamics for $t < 25$ min is not
37 accessible due to the experimental requirement of 25
38 minutes to complete one tomography scan. If the
39 largest pores were modeled as circular channels of
40 uniform cross-sectional radius R and depth h , capillary
41 penetration would require a time $T \cong \frac{2h^2}{R} \frac{\eta}{\gamma \cos\theta}$ of the
42 order of few minutes. For example, $T \cong 5$ mins assum-
43 ing $h \cong 250 \mu\text{m}$, $R \cong 50 \mu\text{m}$, $\eta = 2.4 \cdot 10^3 \text{Pa} \cdot \text{s}$, $\cos\theta \cong$
44 1 , $\gamma \cong 20 \text{mN/m}$). The slow motion of the air-liquid
45 interface in Fig. 4 could be due therefore to the pres-
46 ence of a capillary channel having a complex geome-
47 try.
48

49 Fig. 5 shows the detail of a contact region between
50 two different yarns. After the inter-yarn pores have
51 been filled, the liquid is seen to slowly creep in the
52 relatively wide channel offered by the contact region.
53 Because each contact region is shared by several
54 inter-yarn pores, the liquid is transported from several
55 inter-yarn pores simultaneously. Examination of other
56 images show that the filling behavior illustrated in Fig.
57 5 occurs frequently.
58

The contact region is a relatively low-permeability
path through which the liquid in the pores reaches the
interior of the yarns. The side (xz) view in Fig. 6
shows two yarns, one parallel and one perpendicular
to the page. The top portion of the yarn parallel to the
page is in contact with the droplet. Most of the yarn
perpendicular to the page appears to be dry. For
 $t = 25$ mins, the contact region is only partially filled
with liquid. As time progresses, the contact region is
progressively being replenished with liquid. From the
contact region, the liquid is then transported into the
interior of the yarns. Liquid is also transported directly
from the drop to the interior of the yarns. The reduc-
tion in the extent of the dry regions within each yarn is
particularly evident in the segmented images of Fig. 6
(right column).

Considered collectively, Figures 4, 5 and 6 suggest a
multi-stage transport mechanism (Fig. 7). In the first
stage of liquid penetration, which we term
pore penetration, the liquid is transported through the
relatively large inter-yarn pores. During the pore
penetration stage, some liquid also penetrates in the
yarns that are directly in contact with the droplet. In
the second stage (*contact region filling*) the liquid is
transported mostly in the direction parallel to the
plane of the textile through the channels correspond-
ing to the regions of contact between the yarns. In the
third stage (*yarn saturation*) the liquid slowly wicks
towards the interior of the yarns flowing from the con-
tact region and from the interior surfaces of the inter-
yarn pores. In the case of the yarns that are in contact
with the drop, the liquid originating from the contact
region meets the liquid from the droplet until the yarn
becomes fully saturated.

While the liquid distribution is three-dimensional, in-
formation about the volume-averaged saturation dis-
tribution is often useful. Theoretical analyses typically
describe continuum volume-averaged fields. The the-
oretical model of Patnaik et al¹⁷, for example, as-
sumes that transverse penetration occurs uniformly
under the droplet, and that the liquid front is planar. In
contrast, the model of Starov and coworkers²⁸ as-
sumes that the advancing liquid front in the material
has a hemispherical shape with an increasing radius.
Fig. 8 shows the shape of the region saturated with
liquid as an $r - x$ curve. For any given value of x , r is
calculated as $r = \sqrt{A(x)/\pi}$, where $A(x)$ is the area of
the wet region at x . The shape reported in Fig. 8 is
thus the high-saturation distribution averaged over the
azimuthal angle. To calculate $A(x)$ we applied traina-
ble Weka Segmentation to every yz slice and com-
puted the area of the liquid region.

The shape of the azimuthally-averaged high-
saturation region in Fig. 8 resembles half an hour-
glass. The high-saturation region below the drop is
approximately hemispherical. The back of the textile
is connected with the hemispherical region through a

neck of initial radius $\cong 100\mu\text{m}$. As time proceeds, the radius of the neck increases, and the boundary of the hemispherical region shifts towards the back surface. In the region of minimal slope of the $x - r$ curve (for $r \cong 350\mu\text{m}$), the displacement of the boundary between $t = 25$ mins and $t = 45$ mins is larger than between $t = 25$ mins and $t = 45$ mins, suggesting a slowing down of the average rate of liquid penetration.

We further characterize the average motion of the liquid front by measuring absorption due to X-ray propagation in the yz plane, using the X-ray setup in radiography imaging mode. Fig. 9A shows the X-ray intensity leaving the sample vs. the co-ordinate x for selected times (the corresponding video is available in the Supporting Information). Assuming an inverse correlation between X-ray intensity and liquid saturation concentration, for any given value of x the corresponding X-ray intensity characterizes the area-averaged liquid saturation distribution in the corresponding yz plane. The high liquid saturation region within the textile corresponds to the range from $x \cong 400\mu\text{m}$ to $700\mu\text{m}$ roughly. The range $150\mu\text{m} < x < 400\mu\text{m}$, approximately, corresponds to the portion of the drop outside the textile. The area-averaged position of the liquid-air interface corresponds to the region of largest intensity gradient near the back of the textiles, of which Fig. 9B provides an enlarged view.

We measure the motion of the high-gradient region corresponding to the liquid meniscus by calculating the shift in the x direction of the intensity profiles (see Materials and Methods section). The shift Δx_L of the average x co-ordinate of the liquid-air interface with respect to the front initial position is plotted in log-log scale against time for $\eta = 97\text{Pa} \cdot \text{s}$ in Fig. 9C, and for $\eta = 2.4 \cdot 10^3\text{Pa} \cdot \text{s}$ in Fig. 9D, respectively. The data in Fig. 9C and 9D characterize the one-dimensional kinetics when the liquid-air interface is close to the back of the textile. Evaluation immediately following the deposition of the drop were inaccessible owing to the time needed to place the sample in the X-ray chamber and to reach a stable X-ray source intensity.

For both the smaller and larger viscosity liquids, a single power law does not fit the experimental data. For example, where $\eta = 97\text{Pa} \cdot \text{s}$ the slope of the $\Delta x_L - t$ curve is smaller than 0.5 (the value predicted by the Lucas-Washburn law) for $t > 1000\text{s}$ roughly. Such a poor fit to the data qualitatively highlights the limitation of the single power law description. An observable variation in the liquid penetration behavior shown in Fig. 9 might be due to differences in observation window. Assuming that the penetration rate is inversely proportional to the viscosity, the features corresponding to $t = 1000\text{s}$ in Fig. 9C would correspond to $t = 24700\text{s}$ in Fig. 9D, while our observation window for $\eta = 2.4 \cdot 10^3\text{Pa} \cdot \text{s}$ extends only up to $t = 6000\text{s}$.

A single power-law would be expected if: i) entrance effects associated with the small length of the capillary channels did not alter the dynamics of the liquid penetration process and ii) the liquid transport was governed by a single transport law for any value of t . Continuum theories for liquid transport in porous media, such as the Lucas-Washburn theory¹⁶ or variations of it^{32, 33}, are derived under the assumption that the porous medium can be modelled as a collection of parallel channels having length $L \gg R$, where R is the average pore radius¹⁶. By contrast, in our case, even the largest pores that control the initial motion of the liquid have an aspect ratio close to 1.

Staples and Shaffer³⁴ examined the law of propagation of a liquid meniscus in a capillary tube of sinusoidal cross section. They proved that if the length of the capillary tube is much larger than the wavelength of the sinusoid, the predicted liquid meniscus position fluctuates around the curve predicted by the Lucas-Washburn law. This behavior can be understood by considering the speed of propagation $\frac{dL}{dt}$ of a meniscus in a circular channel of non-uniform cross sectional radius $R(z)$ ³⁴:

$$\frac{dL}{dt} = \frac{1}{4} \frac{\gamma \cos\theta}{\eta [R(L)]^3} \frac{1}{I(L)} \quad \text{Eq. (1)}$$

where

$$I(L) = \int_0^L \frac{dz}{[R(z)]^4} \quad \text{Eq. (2)}$$

For $R = \text{const.}$, integration of Eq. (1) gives $L^2 = \frac{1}{2} \frac{\gamma \cos\theta R t}{\eta}$, *i.e.*, Lucas-Washburn's law. For a small sinusoidal perturbation to a constant radius channel, $R = R_0[1 - \varepsilon \sin(\lambda z)]$ with $\varepsilon \ll 1$. In this case, $I \cong \frac{L}{R_0^4} + 4 \frac{\varepsilon}{R_0^4} \int_0^L \sin\lambda z dz$. For $L \gg \lambda$, the integral of the sine function has zero average, and the Lucas-Washburn law is recovered in a time-averaged sense. However, for $\lambda \sim L$, the solution of Eq. (1) gives a marked non-linear variation from the Lucas-Washburn law, whose features depend on the actual geometry of the pore³⁴. This simple model illustrates that applying results for thick multi-layer textiles to single-layer textiles can give qualitatively and quantitatively incorrect results, and could explain the non-linear behavior seen in Fig. 9C and 9D.

1 An important aspect that we believe could explain the
2 slow down observed in Fig. 9C and 9D is the change
3 in dominant direction of liquid propagation as time
4 progresses. Using the terminology introduced in the
5 discussion of Fig. 7, in the pore filling stage the liquid
6 is transported mainly in the x direction. However, in
7 the contact region filling and yarn saturation stages
8 the fluid is mostly transported in the lateral (in-plane)
9 direction, contributing little to the x component of the
10 average fluid velocity.

11 CONCLUSIONS

12 We have examined by X-ray tomography and radiog-
13 raphy imaging the time-dependent transport of ultra-
14 high viscosity liquids in the transplanar (normal) direc-
15 tion following the deposition of a small droplet on the
16 surface of a single-layer textile.

17 A key insight of our investigation is that the transverse
18 liquid transport appears to proceed in stages, each
19 stage being associated to a geometric feature of the
20 fiber arrangement. The fast initial transport appears to
21 be mainly associated to the fluid filling the large, inter-
22 yarn pores. The late stages of penetration are associ-
23 ated to the slow impregnation of the interior of the
24 yarns. In the intermediate stage where the liquid flows
25 mostly through the region of contact between the
26 yarns.

27 We have quantified the one dimensional kinetics of
28 liquid penetration by using X-ray radiography. The
29 results show that a single power law does not capture
30 all the data. The asymptotic transport rate is much
31 smaller than that predicted by the Lucas-Washburn
32 equation.

33 Models that consider single layer textiles as homoge-
34 neous and isotropic porous media have been applied
35 to lateral, in plane transport with some success^{9, 35}.
36 This success is likely due to the fact that in the lateral
37 direction the asymptotic flow is to a first approximation
38 independent of the microscopic geometry of the tex-
39 tile. In our case, the small thickness of the single layer
40 textile can prevent an asymptotic flow behavior being
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49 REFERENCES

- 50 1. Brewer, S. A., & Willis, C. R. Structure and oil repellency: textiles with liquid repellency to hexane. *Appl. Surf. Sci.* **2008**, 254 (20), 6450-6454.
- 51 2. Leng, B., Shao, Z., de With, G., & Ming, W. . Superoleophobic cotton textiles. *Langmuir* **2009**, 25 (4), 2456-2460.
- 52 3. Advani, S. G., Brusckhe, M. V., & Parnas, R. S. . Resin transfer molding flow phenomena in polymeric composites. *Compos. Mater. Ser.* **1994**, 465-465.
- 53 4. Abrate, S. Resin flow in fiber preforms. *Appl. Mech. Rev.* **2002**, 55 (6), 579-599.
- 54 5. Birrfelder, P., Dorrestijn, M., Roth, C., & Rossi, R. M. . Effect of fiber count and knit structure on intra-and inter-yarn transport of liquid water. *Text. Res. J.* **2013**, 83 (14), 1477-1488.
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established before the liquid reaches the back surface of the textile. This feature could explain the strong deviations from a power law behavior seen in Fig. 9C and 9D.

In addition, to aid the development of theoretical models, our results may have implications for the development of strategies to manipulate liquid transport in textiles. For example, the XCT images suggest that the initial liquid mass flux is governed by transport in the relatively large inter-yarn pores. Altering these features, for instance by utilizing nanoparticles to partially obstruct the pores, could represent an approach to tailor the liquid transport rates in the transplanar direction.

ASSOCIATED CONTENT

Supporting information

Radiography video of transverse transport of a droplet for $\eta = 97\text{Pa} \cdot \text{s}$. This supporting information is available free of charge on the ACS Publication Website at DOI:

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Notes

The authors declare no competing financial interest

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6. Bartels, V. T. Physiological comfort of sportswear. *Textiles in sport* **2005**, 177-203.
7. Pezron, I., Bourgain, G., & Quéré, D. Imbibition of a fabric. *J. Colloid Interface Sci.* **1995**, 173 (2), 319-327.
8. Adler, M. M., & Walsh, W. K. . Mechanisms of transient moisture transport between fabrics. *Text. Res. J.* **1984**, 54 (5), 334-343.
9. Kissa, E. Capillary sorption in fibrous assemblies. *J. Colloid Interface Sci.* **1981**, 83 (1), 265-272.
10. Kissa, E. Wetting and wicking. *Text. Res. J.* **1996**, 66 (10), 660-668.
11. Miller, B., & Tyomkin, I. Spontaneous transplanar uptake of liquids by fabrics. *Text. Res. J.* **1984**, 54 (11), 706-712.
12. Sarkar, M., Fan, J., & Qian, X. Transplanar water transport tester for fabrics. *Meas. Sci. Technol.* **2007**, 18 (5), 1465.
13. Tang, K. P. M., Kan, C. W., & Fan, J. T. . Evaluation of water absorption and transport property of fabrics. *Text. Prog.* **2014**, 46 (1), 1-132.
14. Gillespie, T. The spreading of low vapor pressure liquids in paper. *J. Colloid Sci.* **1958**, 13 (1), 32-50.
15. Adams, K. L., & Rebenfeld, L. In-plane flow of fluids in fabrics: structure/flow characterization. *Text. Res. J.* **1987**, 57 (11), 647-654.
16. Washburn, E. W. The dynamics of capillary flow. *Phys. Rev.* **1921**, 17 (3), 273.
17. Patnaik, A., Rengasamy, R. S., Kothari, V. K., & Ghosh, A. Wetting and wicking in fibrous materials. *Text. Prog.* **2006**, 38 (1), 1-105.
18. Kawase, T., Sekoguchi, S., Fuj, T., & Minagawa, M. Spreading of liquids in textile assemblies: Part I: Capillary spreading of liquids. *Text. Res. J.* **1986**, 56 (7), 409-414.
19. Perwuelz, A., Mondon, P., & Caze, C. . Experimental study of capillary flow in yarns. *Text. Res. J.* **2000**, 70 (4), 333-339.
20. Danino, D., & Marmur, A. Radial capillary penetration into paper: limited and unlimited liquid reservoirs. *J. Colloid Interface Sci.* **1994**, 166 (1), 245-250.
21. Salminen, P. Studies of water transport in paper during short contact times. PhD, Åbo Akademi 1988.
22. Bencsik, M., Adriaensen, H., Brewer, S. A., & McHale, G. . Quantitative NMR monitoring of liquid ingress into repellent heterogeneous layered fabrics. *J. Magn. Reson.* **2008**, 193 (1), 32-36.
23. Rossi, R. M., Stämpfli, R., Psikuta, A., Rechsteiner, I., & Brühwiler, P. A. Transplanar and in-plane wicking effects in sock materials under pressure. *Text. Res. J.* **2011**, 0040517511413317.
24. Weder, M., Brühwiler, P. A., & Laib, A. . X-ray tomography measurements of the moisture distribution in multilayered clothing systems. *Text. Res. J.* **2006**, 76 (1), 18-26.
25. Bayramli, E., & Powell, R. L. . The normal (transverse) impregnation of liquids into axially oriented fiber bundles. *J. Colloid Interface Sci.* **1990**, 138 (2), 346-353.
26. Karppinen, T., Kassamakov, I., Aaltonen, J., Pajari, H., & Hæggström, E. . Measuring liquid penetration in the thickness direction of paper. *The European Physical Journal Applied Physics* **2005**, 32 (1), 65-71.
27. Hyväluoma, J., Raiskinmäki, P., Jäsberg, A., Koponen, A., Kataja, M., & Timonen, J. Simulation of liquid penetration in paper. *Phys. Rev. E* **2006**, 73 (3), 036705.
28. Starov, V. M., Zhdanov, S. A., & Velarde, M. G. Spreading of liquid drops over thick porous layers: complete wetting case. *Langmuir* **2002**, 18 (25), 9744-9750.
29. Rudolf, M., Boutelier, D., Rosenau, M., Schreurs, G., & Oncken, O. Rheological benchmark of silicone oils used for analog modeling of short-and long-term lithospheric deformation. *Tectonophysics* **2016**, 684, 12-22.
30. Arganda-Carreras I, K. V., Rueden C, Eliceiri KW, Schindelin J, Cardona A, Seung HS. Trainable Weka Segmentation: a machine learning tool for microscopy pixel classification. *Bioinformatics* **2017**.
31. Courbin, L., Denieul, E., Dressaire, E., Roper, M., Ajdari, A., & Stone, H. A. . Imbibition by polygonal spreading on microdecorated surfaces. *Nat. Mater.* **2007**, 6 (9), 661-664.
32. Marmur, A. The radial capillary. *J. Colloid Interface Sci.* **1988**, 124 (1), 301-308.
33. Marmur, A., & Cohen, R. D. Characterization of porous media by the kinetics of liquid penetration: the vertical capillaries model. *J. Colloid Interface Sci.* **1997**, 189 (2), 299-304.
34. Staples, T. L., & Shaffer, D. G. Wicking flow in irregular capillaries. *Colloids and Surfaces A: Physicochemical and Engineering Aspects* **2002**, 204 (1), 239-250.
35. Marmur, A. Drop penetration into a thin porous medium. *J. Colloid Interface Sci.* **1988**, 123 (1), 161-169.

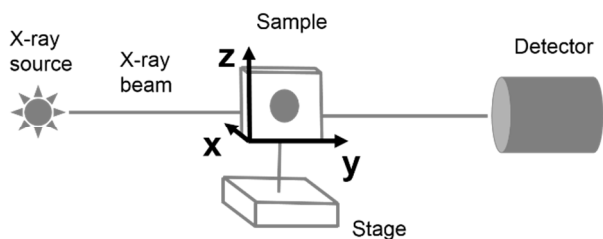


Figure 1. Schematic of the experimental setup. The dark circle in the sample represents the base of the droplet (plane $x=0$). The interior of the textile is in the positive x direction.

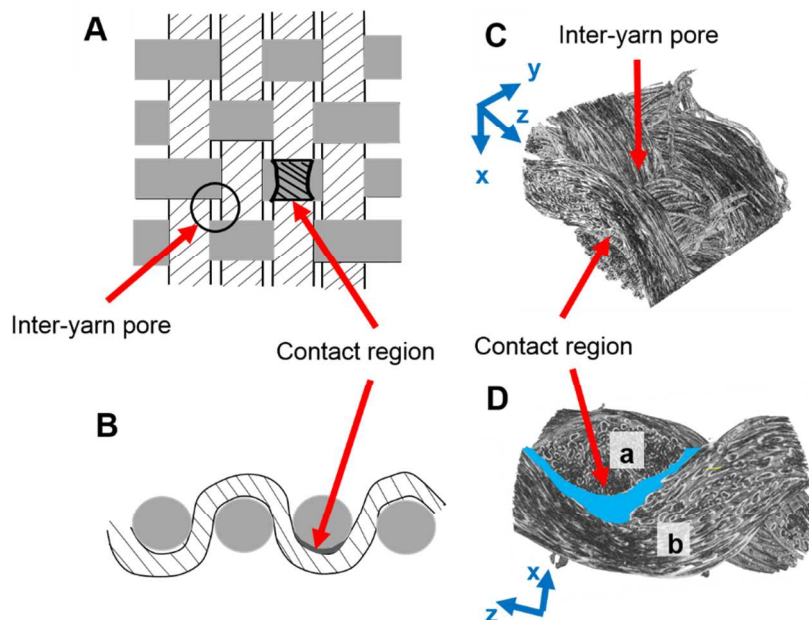


Figure 2. Schematics (A, B) and XCT images (C, D) illustrating the structure of the textile.

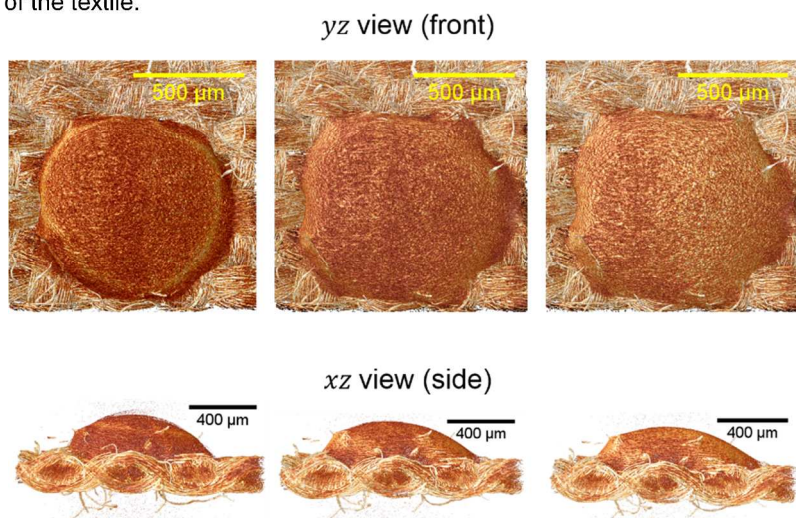


Figure 3. Tomography images showing the shape of the drop in the outside of the textile (online version in color). From left to right: $t=25$ mins, $t=45$ mins and $t=70$ mins, respectively. The liquid viscosity is $\eta = 2.4 \cdot 10^3 \text{ Pa} \cdot \text{s}$.

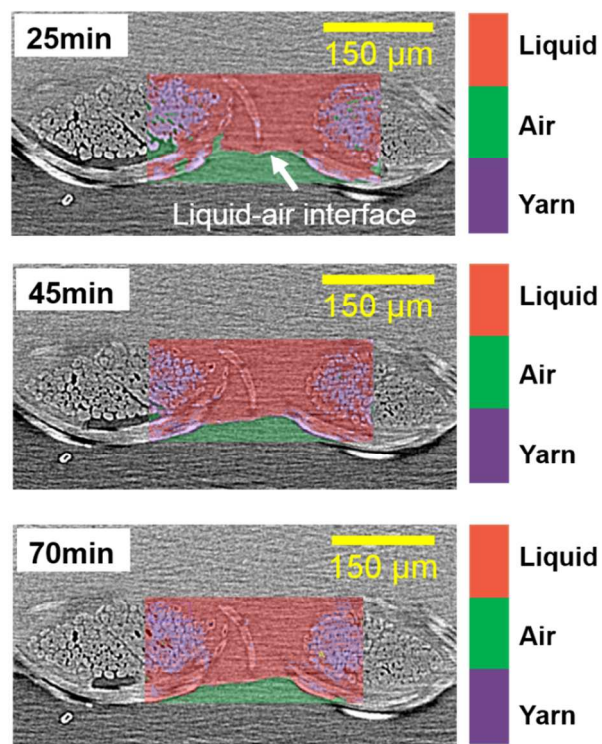


Figure 4. xz slice of liquid distribution in an inter-yarn pore at successive times for $\eta = 2.4 \cdot 10^3 Pa \cdot s$. (Online version in color)

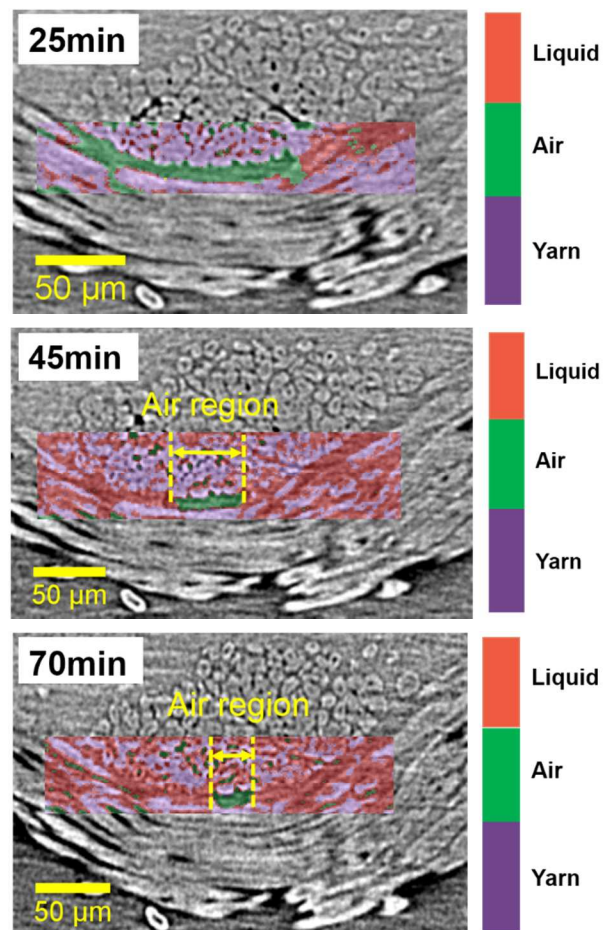


Figure 5. xz slice showing the progressive filling of the contact region by the liquid for $\eta = 2.4 \cdot 10^3 Pa \cdot s$. (Online version in color).

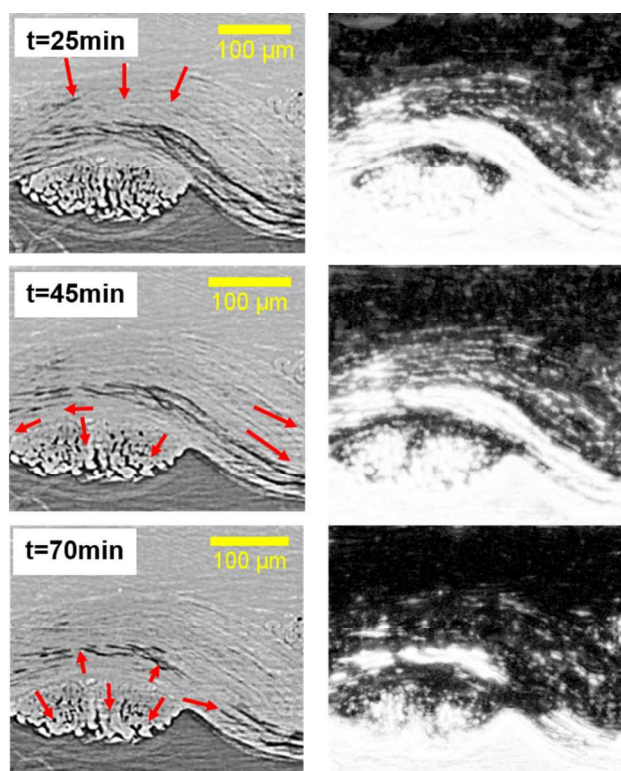


Figure 6. Left: xz slice of liquid distribution in the neighborhood of the contact region between yarns for $\eta = 2.4 \cdot 10^3 Pa \cdot s$. Arrows indicate the approximate direction of the liquid flow. Right: corresponding segmented images, showing the liquid in black.

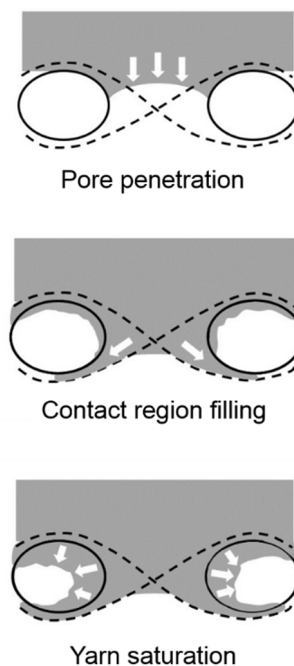


Figure 7. Three stages of transplanar penetration suggested by the XCT images.

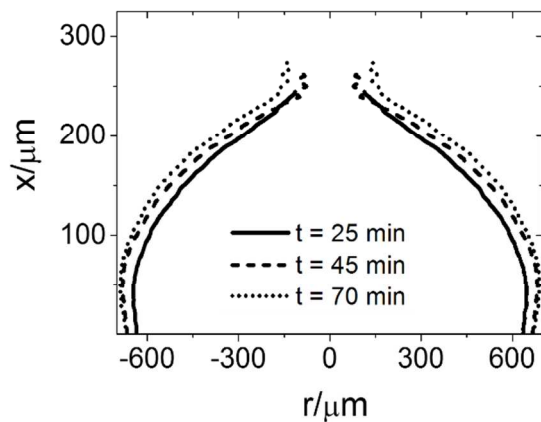


Figure 8. Radius of wetted region inside the textile vs x coordinates for $\eta = 2.4 \cdot 10^3 Pa \cdot s$.

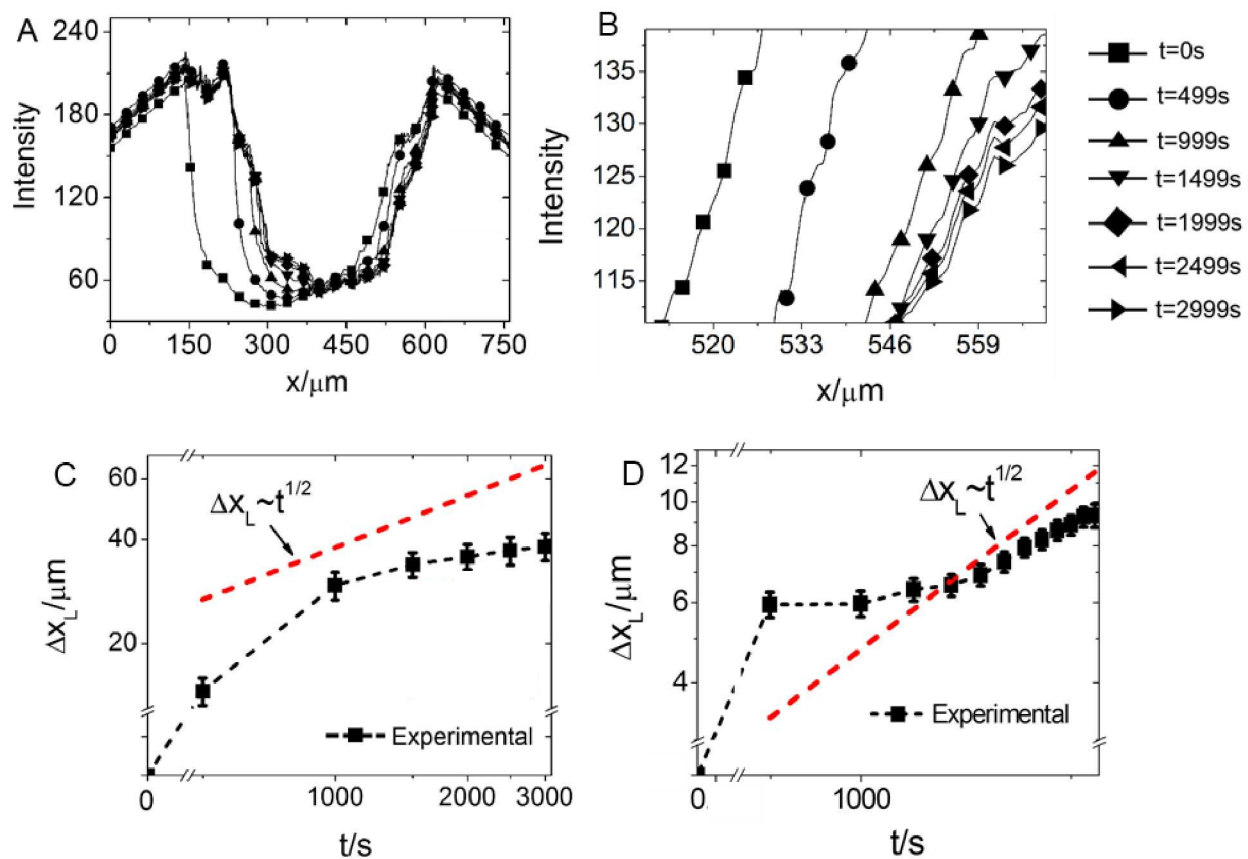


Figure 9. A: profile of X-ray intensity vs x coordinates for successive times and $\eta = 97 Pa \cdot s$. B: zoom in the region $x \in [511, 567]$. C: log-log plot of the average displacement of the liquid front as a function of time for $\eta = 97 Pa \cdot s$. D: the same as C for $\eta = 2.4 \cdot 10^3 Pa \cdot s$.

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