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CAPILLARY PRESSURE ESTIMATION DEPENDING ON FABRIC, LIQUID AND FIBRE VOLUME FRACTION FOR LIQUID COMPOSITE MOULDING (LCM) PROCESSES

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Abstract: *Liquid Composite Moulding processes are considered as promising and effective to manufacture structure composite parts reinforced with synthetic and natural fabrics. The main novelty of this work is the estimation of capillary pressure (P_{cap}) for both fabrics at different fibre volume fractions (V_f) and with different liquids. From the previous works, the P_{cap} was defined as the equivalence between Washburn's equation and Darcy's law while our novel model for the capillary wicking could predict very well the swelling behaviour of natural fabrics. The combination of our P_{cap} definition and our novel model was explored in this study. Linear trends, thresholds and extremums of P_{cap} at different conditions were found for the first time. These results are relevant to estimate the importance of capillary effects during Liquid Composite Moulding processes and extremely valuable for numerical models at the fibrous scale to predict voids formation.*

Keywords: Capillary pressure, Permeability, LCM processes

1. Introduction

In LCM processes, the impregnation of fabric by resin is commonly driven by a pressure gradient. More precisely, low pressure gradients are used to avoid fibre misalignment, meaning that all other forms of pressure such as capillary pressure (P_{cap}) cannot be neglected. This is because such pressure can contribute to the overall pressure gradient to drive flow front movement and an error in calculation of the overall pressure gradient can occur if the capillary pressure is not considered. P_{cap} is, here, a homogenized representation of local wetting effects that depends on the morphology of the medium and its interaction with the liquid. The P_{cap} was fundamentally estimated using the Laplace's equation where some shape factors appear to consider the flow. The Laplace's equation was used coupled with the Darcy's law, measuring simultaneously the P_{cap} with the permeability. Authors have estimated the P_{cap} under a constant applied pressure gradient and at a constant flow rate, but shape factors are difficult to measure, and results strongly dependent on such parameters [1].

Other works focused on the spontaneous capillary wicking to determine the P_{cap} , where no external driving pressure was applied during impregnation. The test velocity was thus non-constant allowing to define a homogenized value of P_{cap} , independent from the flow. In this case the P_{cap} is defined thanks to an equivalence between Washburn's equation and Darcy's

law. The P_{cap} thus depends on parameters accounting for the geometry of the porous medium, the fluid-solid interactions and the medium permeability. All parameters can be evaluated independently by experimental methods [2].

On the other hand, natural fibres have not a stable morphology during wicking that makes difficult the determination of P_{cap} . In this work, a model was developed to predict the capillary wicking in flax fabrics at different fibre volume fractions (V_f), considering the multi-scale swelling effects of the elementary fibres and the individual yarns [3]. Then the P_{cap} was estimated in carbon fabrics and in flax fabrics. The main novelty of this approach resides in two aspects: the study of P_{cap} in natural fabrics and the correlation between the P_{cap} and the V_f , for both types of fabrics (carbon vs flax) and with different liquids (n-hexane vs water).

2. Materials

Carbon and flax fabrics used in this study are quasi-unidirectional woven fabrics. Carbon fabrics were supplied by Hexcel with material code 48580. Such fabrics contain 3% in volume of glass tows in weft direction. They have an areal weight of 541 g/m² and a density of 1.8 g/m³. Flax fabrics were provided by Libeco under the commercial designation FLAXDRY UD 180. Their areal weight is 180 g/m² and density is 1.45 g/m³. Both types of fabrics were used in our previous works dealing with wicking [4,5,6].

3. Theory and experiments

2.1 Equivalence law for capillary pressure (P_{cap})

Assuming that fibre bundles inserted into a hollow cylinder can be approximated as a pack of capillary tubes [4] and according to the conventional Washburn's equation, the linear evolution of squared mass gain as a function of time should be obtained in constant porous medium. This can be expressed as follows neglecting gravity effects [7]:

$$m^2(t) = \left[\frac{(c\bar{r})\varepsilon^2(\pi R^2)^2}{2} \right] \frac{\rho^2 \gamma_L \cos\theta_a}{\eta} t \quad (1)$$

where c is a parameter taking into account tortuosity, \bar{r} is the mean porous radius, ε is the porosity, which is considered as $1 - V_f$ (V_f : the fibre volume fraction), and R is the radius of a hollow cylinder. ρ , γ_L and η are the liquid density, surface tension and viscosity respectively and θ_a is the apparent advancing contact angle. For constant medium porosity (related to geometric product $c\bar{r}$), Eq. 1 fits very accurately the experimental data for synthetic fabrics over time [4,6].

The flow of liquid in a porous medium can also be described by the Darcy's law [8]. This equation relates an equivalent fluid velocity v_D to a pressure drop with the help of permeability K , characterizing the ability of a porous medium to be crossed by a liquid of viscosity η :

$$v_D = -\frac{K}{\eta} \frac{dP}{dh} \quad (2)$$

In the case of wicking of fibre bundles into a hollow cylinder, Eq. 2 was rewritten considering the porosity ε and introducing a negative P_{cap} in the pressure gradient. Integrating linearly the Darcy's law and rewriting this expression as a function of the squared mass gain, a linear relation over time was obtained [4]:

$$m^2(t) = \frac{2K\varepsilon\rho^2(\pi R^2)^2 P_{cap}}{\eta} t \quad (3)$$

where R is the radius of the cylindrical sample holder and ρ is the liquid density.

In order to give a physical meaning to the P_{cap} , it is thus possible to state the equivalence between the Washburn (Eq. 1) and the Darcy (Eq. 3) equations describing the wicking of porous medium. The resulting expression is:

$$P_{cap} = (c\bar{r})\varepsilon \frac{\gamma_L \cos\theta_a}{4K} \quad (4)$$

The P_{cap} thus depends both on the porous medium morphology (a parameter related to tortuosity c , mean capillary radius \bar{r} , porosity ε and the medium permeability K) and parameters taking into account the interactions between the fluid and the porous medium (surface tension of the fluid γ_L and the apparent advancing contact angle θ_a).

2.2 The interpretation of capillary pressure calculation

In this work, the P_{cap} values of carbon and flax fabrics are calculated using Eq. 4. The porosity ε is considered as $1 - V_f$ and the surface tension of the fluid γ_L is taken from [6]. This means that the medium permeability K , the geometric product $c\bar{r}$ and the apparent advancing contact angle θ_a need to be determined in order to determine P_{cap} .

Based on regularly ordered and aligned filaments, equation of permeability along the fibres $K_{//}$ was derived by Gebart [9]:

$$K_{//} = \frac{8R_f^2 (1 - V_f)^3}{\zeta v_f^2} \quad (5)$$

where R_f is the fibre radius (3.5 μm for carbon fibre and 10 μm for flax fibre are the mean values that are considered in this study) and ζ is the shape factor with the value of 53 for a hexagonal arrangement [9].

The geometric product $c\bar{r}$ and the apparent advancing contact angle θ_a are determined by capillary wicking test using Eq. 1. In the constant porous medium, e.g. carbon fabrics, the evolution of squared mass gain as a function of time was linearly observed with both n-hexane and water as described in conventional Washburn's equation. The geometric product $c\bar{r}$ is first determined with n-hexane. Such liquid is considered as a totally wetting liquid which has an apparent equivalent advancing contact angle of 0° . Once $c\bar{r}$ is known, it can be substituted into Eq. 1 again. Hence, the apparent advancing contact angle θ_a can be calculated through a wicking test with water.

2.3 Adaptation to dual scale swelling of natural fibres

However, for natural fibres, the porous medium changes over time during wicking of water, due to fibre swelling. In this case, the wicking test with n-hexane can only be used to determine the initial non-swollen geometrical parameter of the fibrous medium ($c\bar{r}$). In a previous study of the authors [3], Washburn's model was modified, taking into account the fibre swelling at two scales, i.e. elementary fibres and individual yarns. The ($c\bar{r}$) can now be expressed as a function of the surface ratio (ϕ_{yarn}) of yarn in a section of a fabric made of natural fibres:

$$c\bar{r} = c \left((1 - \phi_{yarn})\bar{R} + \phi_{yarn}\bar{r}' \right) \quad (6)$$

where \bar{R} is the inter yarn mean pore radius and \bar{r}' the intra yarn mean pore radius [6].

During swelling of fibres and the associated swelling of yarns, the mean pore radiuses decrease along with the swelling rate of both fibres and yarns. Those two parameters have been identified in authors previous work as linear[3]. Eq. 7 and Eq. 8 can thus be written:

$$(c\bar{r})(t) = c \left((1 - \phi_{yarn})(\bar{R}_0 - bt) + \phi_{yarn}(\bar{r}'_0 - at) \right) \quad (7)$$

$$(c\bar{r})(t) = (c\bar{r})_{ini} - ct \left((1 - \phi_{yarn})b + \phi_{yarn}a \right) \quad (8)$$

where \bar{R}_0 and \bar{r}'_0 are the initial pore radiuses, and a and b are the respective apparent swelling rates of fibres and yarns. Then it is possible to insert those assumptions in the modified Washburn equation of wicking in reinforcements undergoing swelling [5] to be able to describe the deviation to Washburn's original law (Eq. 3):

if $(c\bar{r})(t) > (c\bar{r})_{fin}$

$$(c\bar{r})(t) = (c\bar{r})_{ini} - ct \left((1 - \phi_{yarn})b + \phi_{yarn}a \right) \quad (9)$$

else

$$(c\bar{r})(t) = (c\bar{r})_{fin} \quad (10)$$

The proposed model satisfactorily fits experimental results from capillary tests at different Vf, allowing to obtain the apparent advancing contact angles θ_a for flax fabrics with water. Figure 1 shows a wicking test for the flax fabrics with water and the fit of this non-linear trend with model proposed in Vo et al. 2020 [3, 6][40].

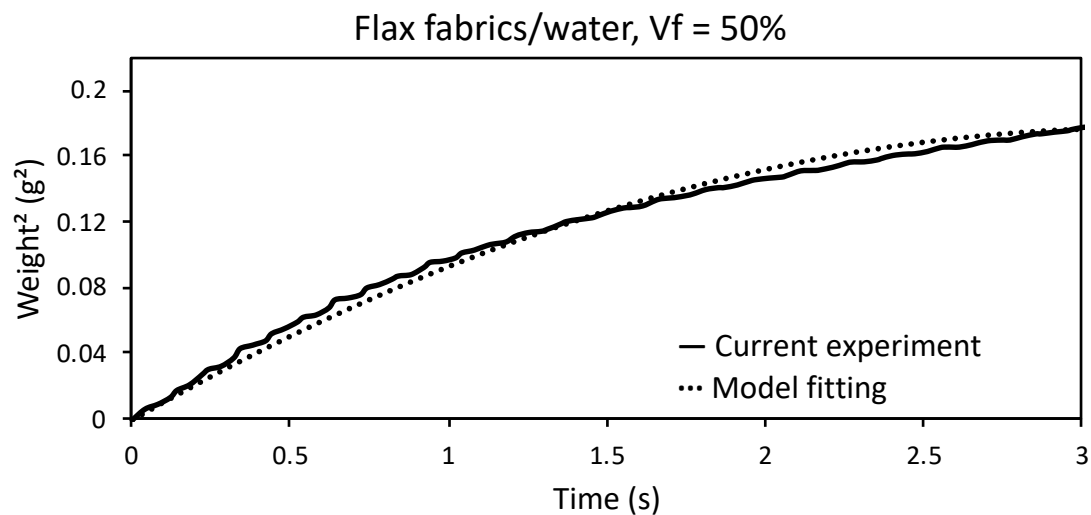


Figure 1. Fitting of experimental wicking data with the model proposed in Vo et al. 2020 [3,6] at 50% Vf.

4. Results and discussions

4.1 Permeability calculation

An important parameter to determine the Pcap is the magnitude of permeability K. According to Gebart [9], this parameter depends on the fibre radius and fibre volume fraction, as shown in Eq. 5. Table 1 presents the values of permeability of carbon and flax fabrics at different Vf. K decreases as Vf increases and the magnitude of permeability of carbon fabrics is much lower than that of flax fabrics. This is logical because the radius of carbon fibre is much smaller than the one of flax fibre at the same Vf.

4.2 Wicking in fabrics

Examples of mass gain during wicking are given in Fig. 2 and 3. Fig. 2 presents data acquired with the carbon/n-hexane couple which is the one that has the best correlation with Washburn law and Fig. 3 depict the different that appears with the couple flax/water due to swelling and modification of the capillary radius in the Washburn equation as modelled in previous works [3,6]

Table 1 : Magnitude of permeability of carbon and flax fabrics with hexagonal packing at different V_f , according to Gebart [9]

V_f (%)	$K_{//}$ of carbon fabrics (μm^2)	$K_{//}$ of flax fabrics (μm^2)
30	7.0	57.5
35	4.1	33.8
40	2.5	20.4
50	0.9	7.5
55	0.6	4.5
60	0.3	2.7
65	0.2	1.5

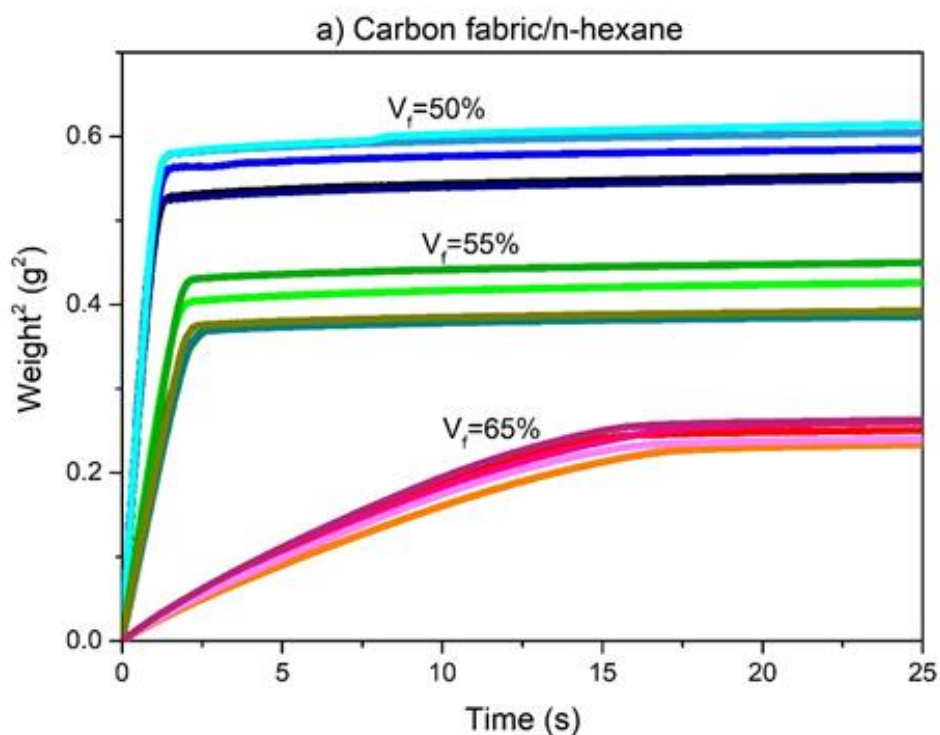


Figure 2. Wicking curves in warp direction of carbon fabrics with (a) n-hexane at 50, 55 and 65% V_f .

The table 2 summarize data obtained from three published papers [3,5,6] and shows that the model predicting the swelling of flax during wicking of water allows a reliable determination of Washburn parameters from experimental data. Using the definition of capillary pressure described by Eq. 4 [5], it is thus possible to express this parameter using.

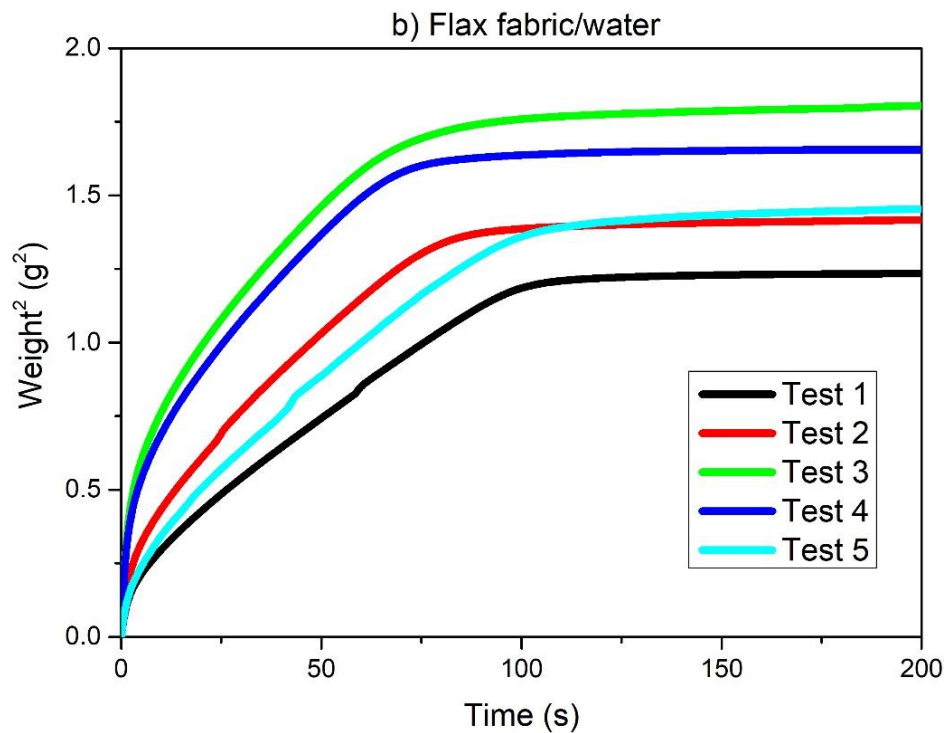


Figure 3. Wicking curves in warp direction of flax fabrics with distilled water at 50% V_f .

Table 2. Average wicking parameters of carbon and flax fabrics at different V_f [6].

Fabrics	V_f (%)	$c\bar{r}$ (μm)	θ_a ($^\circ$)
Carbon	40	28.3 ± 2.4	82.7 ± 0.9
	50	13.0 ± 0.7	75.9 ± 1.5
	55	6.4 ± 0.8	68.7 ± 1.4
	60	$2.3 \pm 0.4^{(1)}$	$55.4 \pm 3.8^{(1)}$
	65	0.9 ± 0.1	56.5 ± 4.9
Flax	30	$14.2 \pm 0.8^{(2)}$	$87.0 \pm 0.3^{(3)}$
	35	$13.4 \pm 1.3^{(2)}$	$76.5 \pm 0.4^{(3)}$
	40	$12.2 \pm 1.4^{(2)}$	$76.5 \pm 1.0^{(3)}$
	50	5.7 ± 0.8	$74.5 \pm 7.7^{(3)}$
	60	$3.8 \pm 0.4^{(1)}$	$68.3 \pm 4.2^{(3)}$

(1) Data taken from Vo et al. 2020 [3].

(2) Data taken from Pucci et al. 2016 [5].

(3) Data obtained through the previous model [3].

4.3 Determination of the equivalent capillary pressure

Using parameters from Table 2, capillary pressures at the mesoscale of the fabrics have been calculated as a function of liquids and fiber volume fraction. Results are summarised in Fig. 4. It can be noted that an extremum appears in some cases but not for all the fabric/liquid couples.

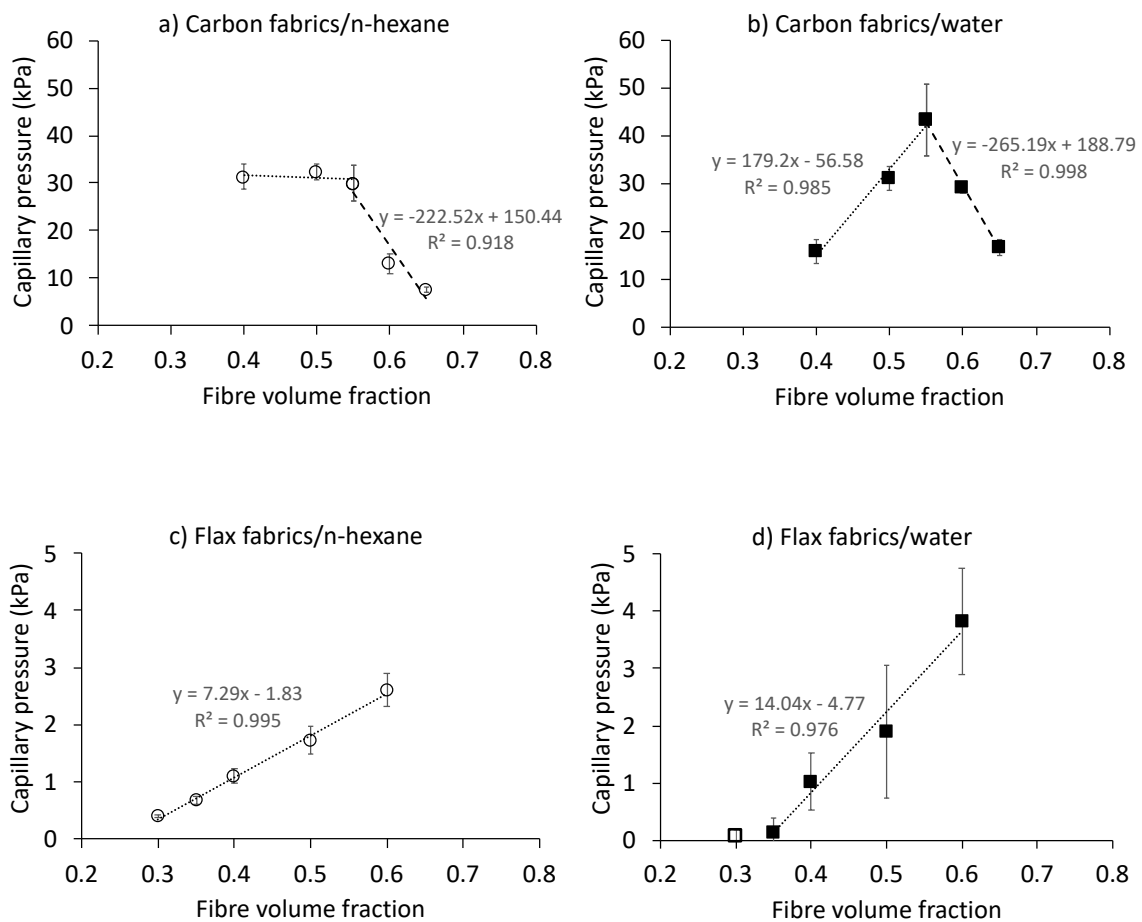


Figure 4. Capillary pressure in carbon fabrics with n-hexane a) and water b); in flax fabrics with n-hexane c) and water d) at different Vf.

5. Conclusions

The presented work proposes a significant extension of methods previously published [5,6,10] confirming the large range of validity of the P_{cap} defined through wicking. The main novelty can be summarised in three principal conclusions.

The first one is that the P_{cap} depends on the tested liquid. It is the first time that the P_{cap} is also calculated for n-hexane. The second one is linked to the reliability of the model developed to consider the swelling of natural fibres. It was proved with the present results that the model is able to provide Washburn's equivalent advancing contact angles that can be used to calculate relevant P_{cap} with water in flax fabrics. The third and main novelty of this work resides in the study of the influence of Vf on P_{cap} for different liquids and fabrics. The principal conclusions obtained here are the linear trend of the P_{cap} as a function of Vf and that it can reach a maximum. Indeed, it was demonstrated that the P_{cap} could either be constant or vary linearly, depending on the type of fabrics (synthetic or natural) and liquids. A threshold value at which the trend is modified has been shown at 55% in carbon fabrics, while such a threshold could not be reached in flax fabrics. This might be due to the values of P_{cap} that are significantly lower in flax (ten times lower).

The results obtained in the present study are of first interest for composite manufacturing simulation [10,11] and a major concern for complex part and 3D reinforcements [12].

Formulating an analytical description of the change of P_{cap} with V_f is thus the next step to be included in further numerical studies along with the understanding and predicting the occurrence of the threshold in further experimental studies.

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