Capillary Driven Saturation of Textile Reinforcing Structures Proposal for an Extension of Lucas-Washburn Equation

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Keywords: Liquid Composite Molding, Textile reinforcements, Capillarity

Abstract. Liquid composite molding (LCM) is a widely used group of various different processing techniques allowing to produce small, medium or even very big sized components from prototype level up to series production. During the infiltration it is necessary to run the process in a way preventing void formation. The typically used textile reinforcing structure results in a dual-scale impregnation consisting of micro impregnation within the constituent yarns of the textile structure and a macro impregnation between the yarns. Capillary rise experiments on flat textile samples are used and the well-known Lucas-Washburn equation has been extended to cover the special configuration. A porous capillary wall is assumed to better represent the three-dimensional nature of capillary networks within reinforcing textiles. An according test rig is presented. Accurate experimental results are gained and capillary radii are computed simple and fast via curve regression.

Introduction

Liquid composite molding (LCM) is a widely used group of various different processing techniques allowing to produce small, medium or even very big sized components from prototype level up to series production. LCM is characterized by preparing the dry reinforcing structure in the final component shape first and infiltrating this so called preform afterwards with the liquid thermoset resin. After infiltration the composite material is cured and, depending on the process setup, net-shaped components are reached.

The infiltration stage requires a special attention. It is necessary to run the process in a way resulting in a complete wetting of the reinforcing structure and preventing any void formation. Especially if higher loaded, structural components are aimed, the very small sized individual filaments (typical diameter range: 7-25 µm) are typically used in the form of yarns (consisting of several thousands offilaments). Within the yarns an arrangement of a dense package of, at least locally, parallel aligned filaments are given. Depending on the filament type, i.e. glass, carbon, bio-based, etc., the intra-yarn interstices tend to be quite constant, e.g. glass filament yarns, show more pronounced variation, e.g. carbon filament yarns, or vary significantly, e.g. natural fibers (typically natural fiber yarns are twisted to gain a stable arrangement of the discontinuous fibers). As a result of slow and unequal capillary penetration of resin into spaces between the filaments, wetting of yarns typically results in microvoids trapped in the resin between the individual filaments [1]. Capillary impregnation of aligned fibrous beds exhibits a high degree of anisotropy depending on whether the fluid flow is parallel or perpendicular [2]. During LCM-processing a low bulk flow front velocity will result in pronounced capillary flow within the yarn. In such a case the capillary flow front may lead the bulk flow front. Increasing the bulk flow front velocity will change the saturation mode. The flow front surrounding the yarn will lead and transverse flow into the yarn will support the saturation of the yarn [2]. Accordingly, the typically used yarn-based textile reinforcing structure results in a dual-scale impregnation consisting of micro impregnation within the constituent yarns of the textile structure and a macro impregnation between the yarns [3-5]. Due to challenging flow conditions significant risk of void formation is given [6]. Whether inter-yarn or intra-yarn void formation is dominant, depends mainly on the average flow front velocity during saturation. Accordingly, void formation can be minimized by infiltration with an optimized flow front velocity resulting in
comparable timely saturation development in the yarns and in between [7]. For complex shaped geometries such approach will not work since a controlled and defined flow front velocity in all regions is hardly reachable. A more detailed understanding about the mechanisms during saturation is necessary. Especially textile reinforcing structures have to be characterized since such structures are mainly used in LCM processes.

In the present paper the capillary driven saturation of textile reinforcing structures is investigated by using a custom-made test rig. Furthermore, an improved modeling based on an extension of the Lucas-Washburn equation is proposed.

**Experimental Procedure**

For the capillary rise experiments a custom-made test rig [8, 9], continuously developed to fulfill all requirements to ensure reliable measurements is used. Details about test setup, test fluid selection, weight and flow front detection are described in [9]. The advantage of the given setup is the use of a flat areal cavity in which the textile structure is placed. The cavity is made of covering 30 mm thick glass blocks and changeable cavity distance frames. This way homogenous sample thickness is ensured and screwed clamping frames allow to apply required compression even if high fiber volume fraction is aimed during the testing.

**Figure 1**: Experimental setup of the capillary rise experiment (top) and a representative data set gained.
Capillary flow modeling and data analysis

Lucas [10] and Washburn [11] investigated the dynamics of capillary flow and proposed the well-established Lucas-Washburn equation (Eq. 1):

\[ h^2(t) = \frac{\gamma_l \cos(\theta) r_c^2}{2 \eta} t \]  

With:
- \( h \): Fluid height
- \( t \): Time
- \( \gamma_l \): Surface energy of the liquid
- \( \theta \): Contact angle
- \( r_c \): Capillary radius
- \( \eta \): Dynamic viscosity

This L-W-equation is based on following assumptions:
- The porous structure consists of constant shaped cylindrical tubes
- An incompressible Newtonian fluid is used
- The fluid exhibits a laminar and viscous flow
- A one-dimensional flow is modeled
- Gravitational effects are neglected

Gravitational effects can be considered by determining the equilibrium reached when hydrostatic pressure and Laplace pressure become equal. According to Jurin’s law [12] this end height is (Eq. 2):

\[ h_{jurin} = \frac{2 \gamma \cos(\theta)}{r_c \rho g} \]  

With:
- \( \rho \): Fluid density
- \( g \): Gravitational acceleration

For small capillary radii the effect is less pronounced and one might neglect the gravitational effect.

From the literature [9, 13-15] it is well known, a difference between experimental results and a fit using Eq. 1 is often found. Consideration of gravity typically improves the quality of the fit. A further improvement is aimed by implementing a ‘porous capillary wall model’ proposed by Blößl [9] (Fig. 2), in which a flow through the walls of the capillary tube, as described by Deen [16], is covered. This way, an extended Lucas-Washburn equation is gained (Eq. 3)

\[ v_z = \frac{r_c}{4 h} \frac{\gamma_l \cos(\theta)}{\eta} - \frac{r_c^2}{8 \eta} \rho g - \frac{v_w h}{r_c} \]  

With:
- \( v_z \): Velocity in z-direction of the fluid inside the capillary
- \( v_w \): Velocity through the porous wall

To determine the parameters for the model, experiments have to be performed. Parallel weight and optical recording allow to double-check the results. As depicted in Fig. 1, the flow front is detected by digital image processing. Difference images of the greyscale values provide the flow front position for every time instance. The quality of this processing step can be improved with various filters and consequently be adapted to many materials. Since the detected flow front points show significant scatter, a method is necessary to reliably describe the flow front line. The empirical cumulative density function (ECDF) with a cumulative distribution limit of 95% was found to be the most suitable [9]. The influence of edge effects as well as a faster flow in hidden layers can be minimized.
The porous capillary wall model can then be generated from the ECDF-flow front data. First, a power law fit is executed to compensate for missing data points and to gain a smooth curve for the flow front.

\[ v_z = \frac{r_c \gamma \cos(\theta)}{4 \eta} - \frac{r_c^2 \rho g}{8 \eta} - \frac{v_w h}{r_c} \]

**Figure 2**: Porous capillary wall model.

Determination of the capillary radius is the next, most critical, step. It can be calculated from the LW equation after determining the slope of the squared height over time progression. Since the slope in a given area needs to be linear and all relevant equations rely on the capillary radius, the selection of a proper value for this area is critical for further calculations. In a series of experiments, the capillary radius depending on porosity ($\phi$) or fiber volume content (FVC) is described by an exponential function.

The parameters fluid viscosity, fluid density, and surface energy are generated separately or taken from literature. Temperature influence on these parameters is considered during the experiments. With all these values, both the LW model and the porous capillary wall model can be described.

The peripheral fluid velocity is determined rearranging Eq. 3 and then applying an exponential fitting function to this data. The model parameters for this function can then be fitted again considering different porosity levels.

**Model Validation**

In the following section, capillary rise experiments performed with different materials are presented and compared with both modeling approaches. A twill 2/2 weave was chosen as reinforcing structure for all materials since it is often used in the processing of composites. Due to its structure, it is also well suited to test the model predictions when peripheral fluid flow is involved. The most common fiber materials carbon, glass, and natural (here: flax) were used. Further information on the materials is given in Table 1.

**Table 1**: Material used in the validation experiments.

| Material | Structure | Supplier | Name           | FAW [g/m²] |
|----------|-----------|----------|----------------|------------|
| Carbon   | Twill 2/2 | C. Cramer, Weberei, GmbH u. Co. KG | Style 423-1 | 400        |
| Glass    | Twill 2/2 | Hexcel Corporation | HexForce01202 | 290        |
| Flax     | Twill 2/2 | Composites Evolution Ltd. | Biotex 200 | 200        |

For all materials, five to seven experiments were performed to ensure statistical reliability and to compensate for possible damaged data sets. As test fluid, Decane was chosen analogous to [9].
After the evaluation of the flow front progression, both the Lucas-Washburn and the porous capillary wall model were applied.

Fig. 3 shows the results of the carbon fiber weave. The blue and the red line depict the mean of the measurement values for nine and eight layers respectively. The small dotted lines show the standard deviation. It can be seen that the fitting curves of the porous capillary wall model approach lie almost exactly on those lines. This demonstrates, the model prediction is accurate but can still be improved slightly. When using (exponential) fitting models’, small deviations in the measured or calculated parameters can lead to some inaccuracy of the results.

Compared to that, the Lucas Washburn fit displayed in orange is only remotely accurate in the initial phase of the measurement. Then its slope completely overestimates the flow front velocity. This shows how the consideration of peripheral flow and gravity can greatly improve the model. The used parameters for the porous capillary wall model are given in the figure.

![Figure 3: Validation of the porous capillary wall model on carbon twill 2/2 weave. Nine (blue) and eight (green) layers and their respective standard deviation (dotted) compared to the Lucas-Washburn (LW, orange) and the porous tube model fit (black).](image)

In further experiments, nine layers of glass and flax fiber twill weaves were used. Analogous to the previous material, the modelling approaches should be validated. In Fig. 4, it can be seen that for both materials the porous capillary wall approach is close to the actual flow front progression. It slightly underestimates the flow velocity. As mentioned before, this can be attributed to small deviations in the calculation of the modelling parameters.

Again, the Lucas Washburn model shows good accuracy in the initial phase of the capillary rise but then overestimates it. As before, it can be stated that the LW model lacks precision when describing reinforcing materials where the capillary flow is not only directed strictly in vertical direction.
Figure 4: Validation of the porous capillary wall model on glass and natural (flax) twill 2/2 weaves. Nine layers of glass (blue) and natural fibers (green) and their respective standard deviation (dotted) compared to the Lucas-Washburn (LW, orange) and the porous capillary wall model fit (black).

Conclusion

The porous capillary wall model approach was introduced to consider more complex capillary flow effects in typically used composite reinforcing textile materials. As an extension of the well-known Lucas Washburn equation, it is capable to describe peripheral fluid flow. The model is validated by the analysis of capillary rise experiments of three different reinforcing materials. It could be proven that the consideration of peripheral flow and gravity has significant effect on the quality of the model. In consequence, regarding common composite reinforcing materials, this model should be preferred to describe the capillary flow in LCM processes.

Acknowledgement

Most part of this work has been elaborated in frame of the project “Reliable and Sustainable composite production for Biobased Components”, funded by the Austrian Ministry for Transport, Innovation and Technology (BMVIT) in frame of the program “Produktion der Zukunft” [Contract no. 858688], which is kindly acknowledged.

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