IN SITU 3D OBSERVATIONS OF THE IMPREGNATION OF MODEL FIBROUS NETWORKS

Chiara Balbinot[1,2], Florian Martoïa[1], Laurent Orgéas[2], Pauline Carion[3], Frédéric Flin[4], Sabine Rolland du Roscoat[2], Jean-Francis Bloch[2], Maxime Terrien[3], Pierre J.J. Dumont[1]*

[1] *Univ. Lyon, INSA-Lyon, CNRS UMR5259, LaMCoS, F-69621 Lyon, France*
[2] *Univ. Grenoble Alpes, CNRS, Grenoble INP, 3SR, UMR5519, F-38000 Grenoble, France*
[3] *Univ. Grenoble Alpes, CNRS, Grenoble INP, LGP2, UMR 5518, F-38000 Grenoble, France*
[4]*Météo-France, CNRS, CNRM, UMR 3589, CEN, F-38000, Grenoble, France*
*Corresponding author: pierre.dumont@insa-lyon.fr

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Introduction

Fibre-reinforced polymer composites are increasingly used in aeronautic, automotive and energy industries to obtain structural or multi-functional parts. An impregnation phase is usually needed for nearly all composite forming processes. It consists of the flow of a polymer matrix through an anisotropic, deformable, and multiscale porous medium made of more or less ordered or disordered networks of fibre bundles, i.e., arrays of aligned fibres. This phenomenon involves complex mechanisms, i.e., the permeation of the polymer matrices, capillary effects and the deformation of fibrous networks due to the fluid interstitial pressure, viscous and capillary effects [1]. A poor control of the impregnation phase is responsible for several defects, such as voids formation or poor fibre wetting in the composite parts. These defects severely affect the mechanical properties and the durability of composites. To understand the formation of voids, it is crucial to understand how the fluid front propagates within the reinforcements, at various scales and in particular within the fibre bundles. The evolution of the flow front is mainly studied using 2D optical visualisation techniques. Thus, there is still a critical need to obtain relevant observations [2,3] of the impregnation phenomena at the fibre pore scale, and to integrate them in macroscale impregnation constitutive models so that the flow front can be described with a more precise geometry than in the actual approaches. Therefore, in this study, 3D X-ray microtomography in situ impregnation experiments of model bundles with various viscous fluids were performed allowing to follow the complex geometry of the flow front.

Materials and methods

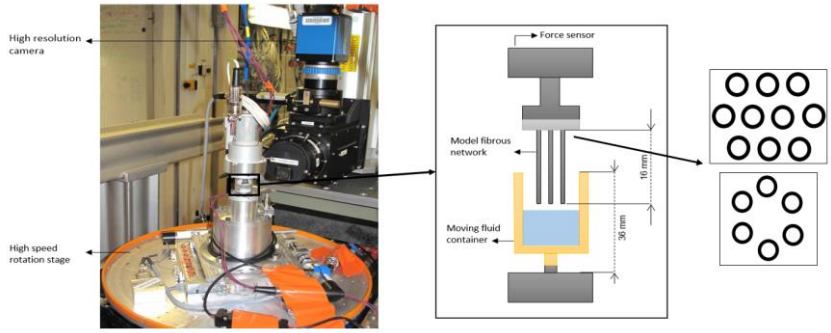

Figure 1: *Photograph of the mini impregnation device that was installed on the microtomograph of the ID19 beamline at ESRF (Grenoble, France) and example of two different architectures of the fabricated model networks of parallel capillary tubes. Note that the tubes were sealed at their upper extremities.*

High-resolution X-ray synchrotron microtomography (ID19 beamline at ESRF) was used to perform in situ impregnation experiments, using a specially designed mini impregnation device (Figure 1). This imaging technique allowed original 3D observations of impregnation phenomena in various model bundles to be obtained. In this study different simply-ordered arrangements of parallel capillary tubes were used (Figure 1). The tubes used to fabricate these rigid porous networks were made of borosilicate glass (1.6 mm diameter). The surface properties of the capillary glass tubes (Figure 2e) were modified using chemical (vapor-phase silanisation with fluoroalkyl trichlorosilane, "Si-F treatment"), or physical

treatments (plasma treatment) to control their wettability. The impregnation experiments were performed using various model fluids with controlled rheological properties and surface tensions: distilled water (γ_L=73 mN/m, μ=0.89 mPa·s) and two kinds of silicone oils with close surface tensions but different viscosities (γ_L =19.9 mN/m, μ=20 mPa·s and γ_L =21.1 mN/m, μ=1000 mPa·s). The impregnation process was performed plunging the fibre network in the fluid moving at different heights the level of the reservoir while the sample was scanned. The X-ray energy and the number of radiographs were set to 19 keV and 500, respectively. A voxel size of 5.1^3 μm^3 was chosen to obtain accurate representation of the geometry of the free surface of the impregnating fluid arising, at the fibre and network scales. The 3D images were reconstructed, from the radiographies acquired, using the Paganin procedure (which is based on the use of the phase contrast in the images). Then, the 3D grey scale images were segmented using software Fiji to distinguish the three phases that form the system: fluid, air and glass tubes (Figures 2a-h).

Results

Figure 2 shows several images obtained for two different testing conditions: a network of glass tubes with a not treated surface, impregnated with distilled water (Figures 2a-d) and a network with the same geometry, but where the capillary tubes were subjected to a Si-F treatment to obtain non wetting conditions (Figures 2e-h). In the first case the fluid (represented in orange) well impregnated the network of glass tubes. The shape of the fluid front showed a capillary rise higher in the centre of the sample with a dome shape. In the second case a 3D concave surface of the fluid front was observed (Figure 2f) and the inner volume of the network was not impregnated, due to poor wettability of the tubes. The image resolution allowed computing both the variations of curvatures of the free surface of impregnating fluid and the contact angles between the liquid phase and the tubes.

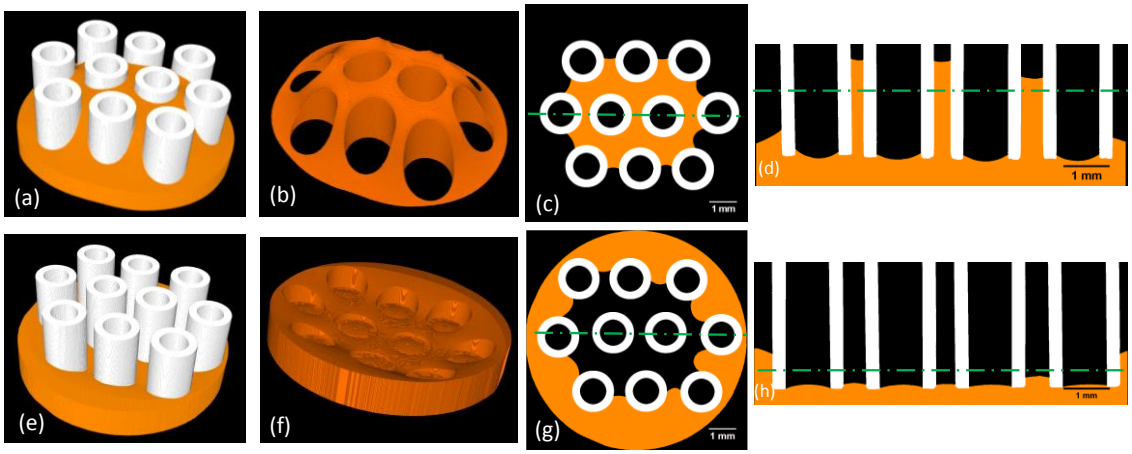

Figure 2: *(a) 3D view of a model network made of capillary tubes (white) impregnated with distilled water (orange) obtained using X-ray microtomography imaging (voxel size of 5.1^3 μm^3). (b) Corresponding 3D view of the flow front between the capillary tubes. (c,d) Corresponding horizontal and vertical cross sections. First row: case of non-treated tubes. Second row (e-h): same type of views as (a-d), but for the case of Si-F treated capillary tubes.*

Conclusion

This study shows that synchrotron X-ray microtomography is a suitable and efficient tool to enhance the description of impregnation phenomena at the fibre scale within model fibre bundles. The obtained 3D images unveil the role of capillary forces (through the measurements of curvatures maps) as well as the effects of the wettability of fibres.

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