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An *in situ* investigation of microscopic infusion and void transport during vacuum-assisted infiltration by means of X-ray computed tomography

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ABSTRACT

In situ vacuum-assisted infiltration experiments were carried out using synchrotron X-ray computed tomography (SXCT) to study the mechanisms of microfluid flow within a fiber tow. A single tow of E glass fibers was infused with a water and syrup blend using an apparatus designed and built for this purpose. The high resolution of the SXCT images allows the detailed reconstruction of individual fibers within the tow while the contrast between the different phases (air, fluid and fibers) was enough to track the fluid front position and shape as well as the void transport during infiltration. The ability of this technique to provide detailed information of microfluid flow and void transport in composite materials is clearly established. The fluid propagation at the microscopic level as well as the mechanisms of void transport within the tow were related to the wetting between the fluid and the fibers, the rheological properties of the fluid and the local microstructural details (fiber volume fraction, fiber orientation) of the fiber tow.

1. Introduction

Liquid molding technologies, such as resin transfer molding (RTM) and vacuum resin infusion (VARI), are widely used for manufacturing of complex composite parts in different industrial sectors. In this strategy, resin is infiltrated into the dry fiber preform by means of a pressure gradient between the inlet and outlet gates. A major limitation of liquid molding is the generation of voids and air entrapments during infiltration, which reduce significantly the mechanical properties of the composite. Macrodefects, resulting from dry – or poorly impregnated – zones, appear when the resin flow reaches the outlet gate before completely filling the component. They are due to an inadequate distribution of the injection/infusion/venting ports in the component. In addition, microdefects (voids) can also appear, even if the part has been completely filled with resin during infusion. Standard reinforcements used in composite manufacturing are formed by tows containing thousands of fibers in specific fabric architectures (unidirectional, woven, non-crimp, stitched, etc.), leading to microporosity (the free space in between individual fibers within the tow) and macroporosity (the free space between tows). Viscous flow dominated by the resin pressure gradient takes place through the high permeable channels between adjacent fiber tows while tow impregnation perpendicular to the fiber is mainly driven by capillary forces if the fibers are wetted by the liquid. This dual-scale flow (micro-meso) is partially responsible for the generation of voids during liquid molding of composite parts because of the competition between viscous and capillary forces [14,15,26,27].

Previous experimental observations have demonstrated that void formation in engineering composites depends on the ratio between viscous and capillary forces through the non-dimensional modified capillary number, Ca^* [9,24,25,29]

$$Ca^* = \frac{\mu \bar{v}}{\gamma \cos\theta} \quad (1)$$

where μ and $\bar{v}$ stand, respectively, for the resin viscosity and the

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average resin velocity, while γ and θ are the fluid surface tension and the contact angle, respectively. An optimum capillary number to minimize the void content has been found in specific material systems [9]. Viscous forces are dominant with respect to capillary ones for large capillary numbers ($Ca^* > 10^{-2}$) and the rapid flow of the resin through the tow-to-tow free gaps leads to the formation of voids within the tows, as entrapped air cannot scape. On the contrary, the fluid velocity between tows is smaller for low capillary numbers ($Ca^* < 10^{-3}$), and the wicking effects caused by the intratow capillary forces become dominant. Under these conditions, fiber tows are rapidly filled with resin and are generated between tows.

Obviously, understanding the resin flow mechanisms is crucial to manufacture composite parts with minimum porosity by liquid molding and the selection of the appropriate experimental technique to monitor accurately void generation and transport is a critical part of this process. Macroscopic resin flow measurements can be performed by direct image monitoring in transparent acrylic molds during RTM [13,23,25] or on the surface of the vacuum bag in VARI [15]. Vilà et al. [36] tracked the position of the resin flow front in a VARI process by measuring the changes in the fabric thickness, associated with the resin-fabric stress transfer during infusion, by means of digital image correlation. Dielectric sensors [22] or embedded optical Bragg sensors, based on the change of the refractive index of the surrounding media when the fabric is infiltrated, can also be used to track resin flow and curing at discrete points [8,18]. Other authors have used C-scan ultrasounds when resin progress through the fabric [34]. The infusion experiments were carried out with the vacuum bag immersed in a water tank and the resin flow was stopped at different times. Fully and partially saturated regions were tracked by C-scan by averaging the signal attenuation in the through-the-thickness direction of the laminate. This technique can also be used to monitor the flow propagation in the through-the-thickness direction in thick fabrics to determine the out-of-plane permeability [33]. All these techniques provide valuable information about the macroscopic flow propagation in a composite part during the infusion/injection process but their low spatial resolution hinders the analysis of the microflow process within the tow.

To overcome these limitations, Magnetic Resonance Imaging (MRI) can be used to track the flow front by mapping the fluid concentration inside porous samples of cm dimensions [17]. Neacsu et al. [21] carried out MRI measurements to study capillary-driven transversal flow in bundles of aligned fibers using blends of water and corn syrup with protonated liquids. The evolution of the wet portion of the fiber bundle with respect to time was obtained and compared with analytical models of fluid propagation. Endruweit et al. [5,6] determined the local fluid concentration using MRI during impregnation of several fabrics, including woven, non-crimp and triaxial braids, detecting local variations attributed to the microvoid formation at the tow level as well as dry spots.

X-ray radiography has also been used to track the progress of the fluid during infiltration due to the difference of X-ray absorption coefficients between the fibers and the fluid [2]. The two-dimensional information provided by radiography can be very useful to track the progress of the fluid front but it is not suitable to analyze the development of porosity, particularly at the microlevel. X-ray computed tomography (XCT), in which a set of radiographies obtained during sequential sample rotation are used to obtain a three dimensional reconstruction of the object, is much more appropriate for tracking porosity during infiltration. This technique has been successfully applied to study damage in composites [31,32], as well as to analyze the effect of processing conditions in the porosity of composite materials [3,4,10,11]. Moreover, the resolution of this technique (in the range of μm) is adequate to detect intratow and intertow voids, including information about their size, shape and spatial distribution.

To the authors' knowledge, XCT has not been used to monitor the infiltration process at the microscale in composite materials and this work is intended to establish the advantages and limitations of this technique for this particular problem. To this end, a miniaturized apparatus for vacuum infusion was designed and built to study *in situ* the infiltration mechanisms operating at the microscale in an XCT synchrotron beamline. These conditions are representative of the fluid flow in textile preforms under low capillary numbers in which the wicking effects due to capillary forces are dominant with respect to viscous flow. The fluid propagation at the microscopic level as well as the mechanisms of void transport within the tow were related to the wetting between the fluid and the fibers, the rheological properties of the fluid and the local microstructural features (local fiber volume fraction, fiber orientation) of the fiber tow.

2. Experimental techniques

The vacuum infusion apparatus was designed in such a way that the fluid is infiltrated from the top of a vertical single fiber tow while the outlet is placed at the bottom, Fig. 1a). Thus, the device can be installed in the rotation stage of a XCT system. The miniaturized device allows to study small specimens consisting of one to three fiber tows within the detector field of view of the X-ray system (approximately 40 mm in length by 3.8 mm wide). An important feature of the device is the miniaturization of all the parts of the infusion process, including the inlet and outlet ports. The inlet was manufactured using a syringe needle of 0.8 mm in diameter which provides a reservoir of 20 ml and a piston system to control accurately the volume of infused fluid. The outlet was made using an open syringe of 0.8 mm in diameter previously cut and adjusted to the support, which is connected to the vacuum system.

The sample was a single fiber tow of 2K (1000 tex) extracted from a plain woven fabric of E glass (average fiber diameter 16 ± 2 μm), which was placed in between two standard vacuum bag films of about 64 μm in thickness (NBF-540-LFT) thermally sealed at the edges, Fig. 1b). The vacuum bag and fiber tow assembly was 25 mm in length with an irregular cross-section of ≈ 4 mm in width and 0.5 mm in thickness, Fig. 1c). The sample was placed between the inlet and the outlet, connecting both ends with the respective syringe needles, and sealed with standard tacky tape (LTT-90B). Thus, the sample contained in the vacuum bag film was completely sealed at the edges (by thermal sealing) and borders (by tacky tape) at the infusion and vacuum points. The whole system was mounted on an aluminum support connected to the vacuum system. Vacuum pressure of $\approx 0.91 \times 10^5$ Pa was applied with a rotatory vane pump connected to the vacuum line to avoid leaks during the tomographic measurements. The vacuum pressure was continuously monitored during the *in situ* experiment. The infusion tow was encapsulated within a PMMA cylinder (at atmospheric pressure inside the cylinder) to provide mechanical stability to the infusion device and to protect the X-ray system against fluid spills without significant X-ray attenuation.

The fluid infusion is controlled by a simple regulation system made up of a screw and a nut with washer. Initially, the nut and the washer block the syringe piston preventing fluid propagation driven by the vacuum. The infusion driven by vacuum can progress by rotating the nut of the screw. In this way the flow front can be controlled accurately by adjusting the nut rotations and the total amount of fluid infused can be controlled step-by-step with the thread pitch. This is critical because any parasitic fluid propagation through the sample during the acquisition time of the tomograms (due to negligible leaks at the vacuum and infusion ports or other