Vacuum assisted cellulose infusion: A technique for nano-reinforcements deposition for multiscale composites.

Francisco Javier Goyo-Brito.; José Ricardo Tarpani.

Sao Carlos School of Engineering (EESC-USP), 13566-590, Sao Carlos-SP, Brazil
goyo-brito@usp.br, jrpan@sc.usp.br

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Abstract. *Unsized carbon fiber fabrics (CFF) were impregnated with three different concentrations of an aqueous suspension of microfibrillated cellulose (MFC), 0,1; 0,2, and 0,4 %wt. Vacuum-assisted infusion was used to make the suspensions penetrate and deposit the cellulose on carbon fiber (CF) surfaces, the effect of three different preform assembly configurations (PAConfig) on cellulose deposition was evaluated. Resin infusion under flexible tooling was used to manufacture multi-scale MFC-CF-Epoxy composites. The infusion of MFC was an effective method for its penetration, forming a tridimensional network of MFC between CF, transferring the stress between them, and serving as a node at the intersection of 0-90 bundles. The effect of MFC concentration on the improvement of the mechanical properties is dependent on the deposition efficiency provided by the PFConf. The use of distribution media (DM) promotes superficial deposition of MFC on CFF improving the flexural strength when 01 and 0.2 wt% MFC, while the infusion without DM provides more effective amounts of MFC for increasing the elastic modulus of the composites.*

1. INTRODUCTION

In the last decade, it was identified that micro/nanofibrils of cellulose (MFC) might increase the choices of fibers in composites and expands their use because of their excellent mechanical properties [1]. Furthermore, coating continuous fibers through impregnation with MFC aqueous suspensions has proven to be an effective, simple, and low-cost method for improving the mechanical properties of composites [2].

In carbon fiber reinforced polymers (CFRP) has been determined that adhesion between MFC and carbon fibers occurs by Vander Waals interaction[3] and that it also depends on CF roughness, improving the toughness of epoxy matrix composites MFC-CF-EP [4]. Methods as dipping coat, spray coat, or electrophoresis have been developed for the manufacture of these multi-scale composites that have been developed using MFC aqueous suspensions by coating carbon fiber fabrics (CFF) [5].

This work proposes vacuum-assisted infusion as a rout for adding MFC that not only coat CFF but provides the deepest penetration of MFC into them, therefore, better mechanical properties of MFC-CF-EP composites.

Since preform assemblies configurations (PAConfig) are determining on the flow and impregnation of fibers when infused by resins [6][7], three layouts were studied for impregnation with MFC suspensions, as well as the effect three different concentrations of MFC were used.

2. EXPERIMENTAL

2.1. Infusion of Cellulose process

MFC water paste was transformed into 0,1; 0,2; and 0,4 wt. % of MFC/water suspensions by dispersing 10 wt.% MFC in distilled water with an Ultra-Turrax during 4 minutes @ 10.000 rpm. Those suspensions were used to coat CFF (previously dried for 3 hours at 100 ° C) using a vacuum-assisted MFC infusion process, similar to the vacuum-assisted transfer molding process.[8]

Three PAConfig were used for the diffusion of MFC suspensions through the carbon fiber fabrics, as shown in Figure 1. The first, only the carbon fiber fabrics between the mold and the vacuum bag or Non-Mesh (NoM), the second, identified as On-Top (OnT), by placing a distribution media (DM) on the carbon fiber fabric, and third, Sandwich (San) configuration with a distribution network above and below the carbon fiber.

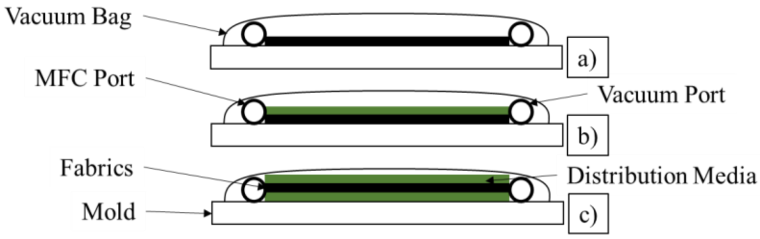


Figure 1 - Schematic of PAConfig:(a) NoM arrangement, (b) OnT arrangement, (c) San arrangement.

MFC suspensions were infused through the CFFs using a vacuum of -40 kPa at ambient temperature, controlli[8]ng the inflow in a rate of 100 mm/min, the process stops once the entire 50 ml of suspension pass through. CF-MFC infused was subsequently demolded and dried in an oven for 3h at 100°C.

2.2. Manufacturing of CF-MFC/EP composites

Resin infusion process under flexible tooling [9] was used to permeate the preforms containing the non-coated CFF (Reference or Ref) as well as the two-plies CFF-MFC coated and dried of each preform arrangement. Under a vacuum bagging pressure of −80 KPa, filling time for the entire preform was 10 min, under de same pressure curing was carried out for 24 h at room temperature. Post curing was performed in an oven for 5 h at 70 °C.

2.3. Flexural Properties

Three-point loading tests were performed according to the ASTM D7264-15 standard to determine the flexural strength (MPa) and the flexural modulus (GPa) of the CF-MFC/EP composites. The testing machine was 5969 Universal Material Testing by INSTRON (load cell: 5 kN). The samples were two-ply laminates with dimensions close to 60x15x0.5 mm3 and a span to thickness ratio of 40:1.

2.4. Microscopy

Morphological characteristics of MFC deposition onto CFF and as fractographic aspects of CF-MFC/EP specimens were examined in a field emission microscope (FEM) FEI Inspect F50™. An electrically-conductive ultra-thin carbon layer was sputtered over the sample surfaces to achieve the appropriate electron flow.

3. RESULTS AND DISCUSSION

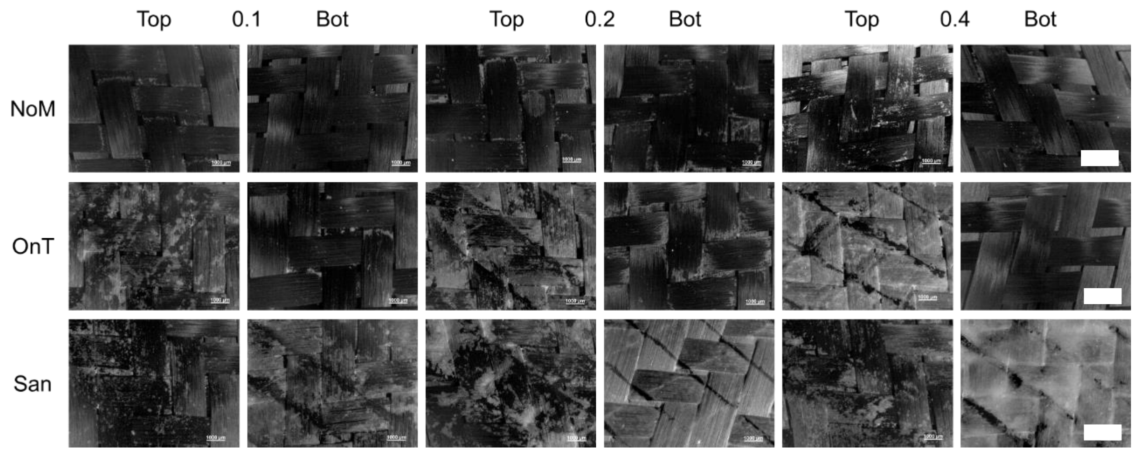


Figure 2 - microscope images of the top and bottom of MFC coated CFF (CF-MFC) through the infusion process. MFC suspension concentration ordered in pairs of images. PAConfig images ordered by rows. Scalebar equivalent to 2 mm.

Figure 2 shows images of the upper and lower surfaces of the MFC-infused CFFs, ordered by MFC suspension concentration (columns, left shows the top of the fabric, right It bottom) and PAConfig used (lines).

It is observed that the deposition of MFC on CFF is heterogeneous, this is due to the strong entanglement that links firmly MFC fibers of different lengths and diameters contented in the suspension [10].

In the cases of NoM, few dispersed superficial deposition is observed, for 0.1 and 0.2 wt% MFC is observed mainly in the cross-linking valleys between fibers, and less in the parts that are in contact with the vacuum bag or the mold, suggesting that the MFC follows internal paths, between bundles, instead of superficial.However, for NoM0.4, a more superficial MFC is observed, in the lower part of the fabric, it is not observed. Grimsley et al [11] indicate that (in no DM systems) when local fluid pressure increases, a spring-back effect occurs, it reduces the decompression of the vacuum bag, letting the suspension flow through the upper part of the CFF.

In OnT and San PAConfig, the CFF surfaces in contact with the DM are shown coated with MFC, it is observed that low planar permeability of DM permits a greater superficial deposition [12].

A greater surface deposition is observed in the SAN configuration on both sides, given the preferential flow of the suspension through the DM, which could disadvantage the penetration through the bundles. Moreover, marks left by the contact of the DM are observed, which indicates that when removed DM carries with it the MFC deposited on it, indicating that the MFC flows even above it.

Figures 3 show an SEM micrograph of top views of CFF infused with 0.2 wt.% MFC, in NoM, OnT and San PAConfig, those show the web-like structure of a wide range of diameters (white arrows indicating the direction of infusion flow), a wide network of nanofibrils was observed, which would present great interdependence for mobility between them. Chen et al claim that getting loose fibrils in MFC suspensions is unlikely because they tend to tangle with each other and form networks.[13].

As indicated by the red arrows in Fig. 3.b, it can be seen that MFC not only connect with other MFC in the same plane but also penetrate and interconnect with networks that flow in other planes throughout the thickness of the preform and between CF plies. The MFC forms a continuous three-dimensional network, one that penetrates the layers between the CFs, embracing and interconnecting them. Furthermore, it is observed that MFC serves as a link node between CF crossing bundles, in the cases of the OnT and San arrangements it is characterized by the creation of interconnecting bridges of this MFC network that seem to cover superficially and then they penetrate through the fibers.

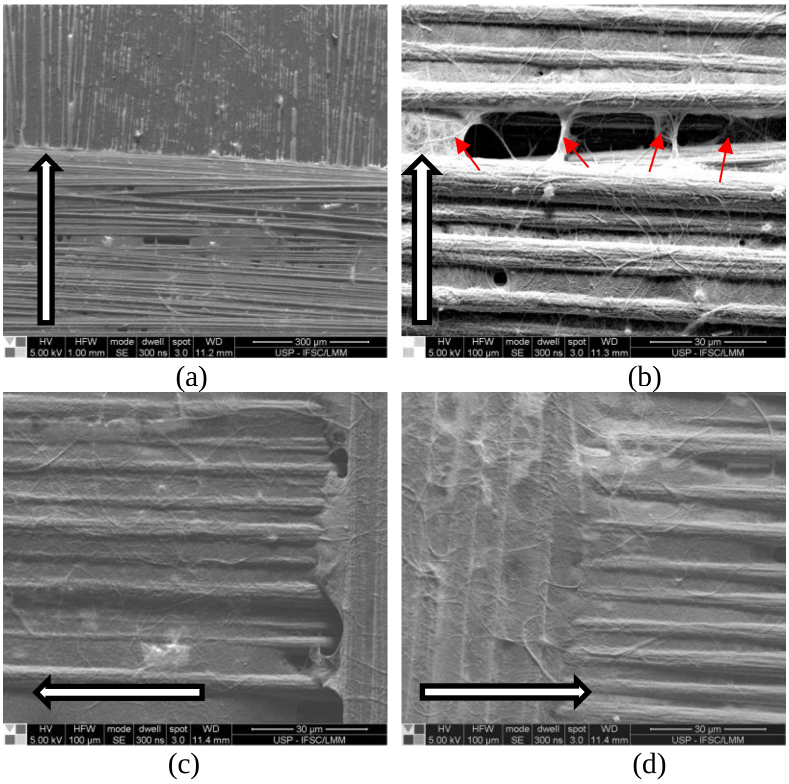


Figure 3 - SEM imaging of dried MFC-CFF infused with 0.2 wt. % suspensions; (a, b) NoM CFF top view (c) OnT and (d) San, top view. Arrows indicate suspension flow direction.

The effect of MFC content on the flexural properties of infused MFC-CF-Epoxy composites are presented in the Fig. 44. Error bars are 1 standard deviation. It is observed that the modulus of elasticity values improved with the addition of MFC, in relation to the MFC concentration, it is observed that 0.2 wt.% presents the highest values as a group, that is, independent of the PAConfig used for the infusion.

PAConfig analysis shows that for 0.4 wt.% NoM shows best results, nevertheless, for the concentrations of 0.1 and 0.2 wt.% OnT results are better. OnT and NoM values are similar when compared for 0.1 wt.% and 0.2 wt.% of MFC. This may be related, to the direction of flow penetration; in NoM the MFC suspension initially penetrates into the CFF in a transverse direction, as the viscosity increases or the permeability decreases, the local pressure in the fluid increases and the spring-back effect in the vacuum bag occurs [14].

The MFC nodes that interconnect the carbon fiber 0-90 bundles can be a point of transmission and distribution of loads, likewise, the penetration between plies of CFF can also be a factor that provides greater resistance to delamination.

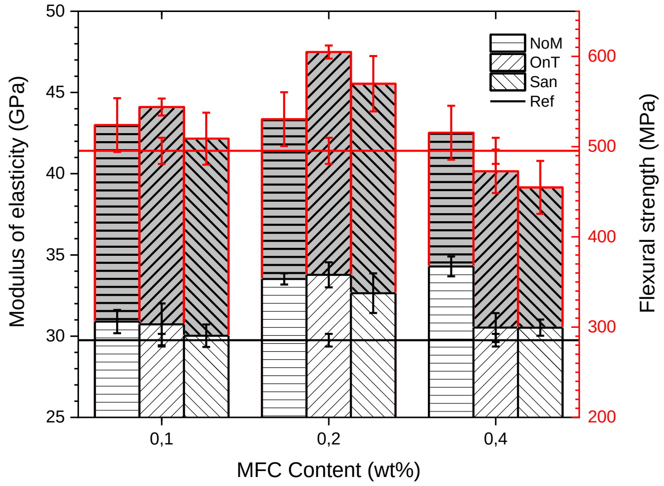
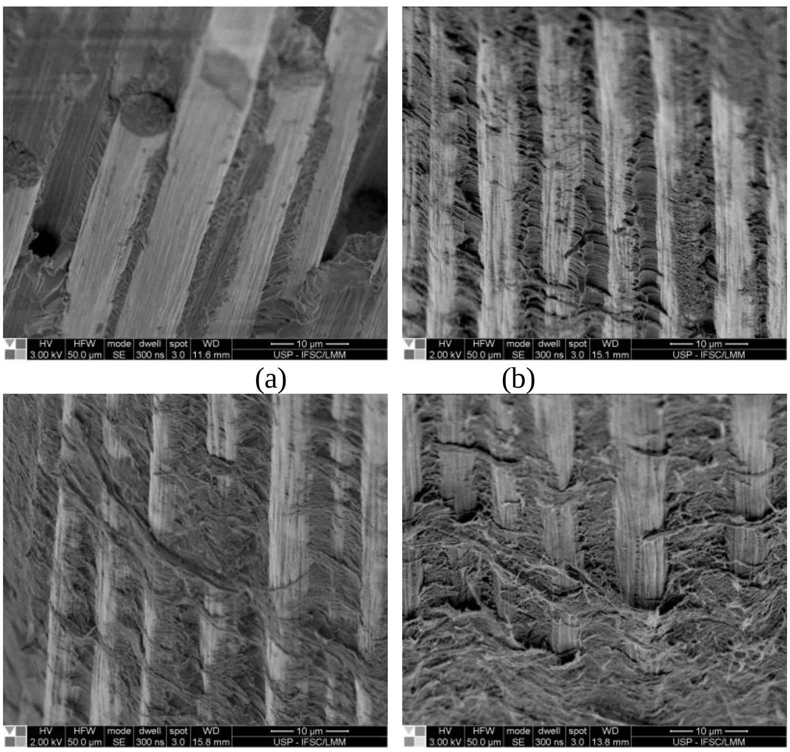


Fig. 4 - Effect of the MFC deposition on MFC-infused/CF/Epoxy composites on flexural properties.



(c) (d)

Figure 5 - Fractography of: delaminated inter-plies surface of NoM infused MFC/CF/EP laminates: (a) 0 wt. %MFC and (b) 0.1 wt. %MFC, (c) 0.2 wt. %MFC and (d) 0.4 0 wt. %MFC, MFC-infused/CF/Epoxy laminates. Side view of fractured OnT0.2 laminate showing

As observed in Fig.5, composites with MFC present a rougher fracture surface, with fibril remnants still adhering to the CF surface, there is a tendency for the permanence of fibers oriented in the same direction. Furthermore, when analyzing the surface between CFF plyes, the presence of MFC in this region indicates that the cellulose infusion method allows the penetration of MFC into regions not reached by conventional methods.

Likewise, it can be seen in the morphologies of the MFC were frayed during the fracture, a part of these remained in the epoxy and another part in the CF, indicating good adherence to both. Alienation of the fibers that remained adhered to the CF is also observed, which may indicate that fibers were pulled by the matrix in a preferential direction according to the stress.

4. CONCLUSIONS

The infusion method was effective for MFC penetration into CFF, forming a 3D network in between CF, transferring the stress between them, and serving as a node at the intersection of 0-90 bundles. The effect of MFC concentration on the improvement of the mechanical properties is dependent on the deposition efficiency provided by the PFConf. The use of distribution media (DM) promotes superficial deposition of MFC on CFF improving the flexural strength when 01 and 0.2 wt% MFC, while the infusion without DM provides more effective amounts of MFC for increasing the elastic modulus of the composites.

Highest concentrations can saturate CFF surfaces reducing permeability, and the penetration of subsequent MFC. Deposition efficiency does not necessarily mean an increase in mechanical properties. The specific distribution of the deposited MFC may be more efficient.

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