# STUDY OF THE FLUID-STRUCTURE INTERACTION OF A MAT REINFORCEMENT WITH A THERMOPLASTIC RESIN

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## ABSTRACT

The aim of this work was to study the compression behavior of a mat reinforcement. The compaction stages of the dry and wet reinforcement and its relaxation over time as a function of several pressure levels have been characterized with a custom-designed bench. The compression behavior of the reinforcement can be modeled by a power law. The coefficients of the law are changing with the rupture of the fibers during compression. The study of the reinforcement compressibility enables an optimized composite manufacturing, to obtain materials without porosity and without increasing the thickness induced by a polymer excess.

## 1 INTRODUCTION

Impregnation is one of the major steps in the composite manufacture. It transforms the assembly of reinforcement and matrix layers into a rigid composite. The flow of resin through a dry reinforcement is usually modeled as a saturated flow through a porous media, thanks to Darcy's law [1]. The high viscosity of thermoplastic matrix requires consolidation pressures of several dozens of bars. The application of Darcy's law to film stacking consolidation must therefore be adapted for being used with thermoplastics because of very strong fluid-structure interactions, involving hydro-mechanical coupling [2]. In particular, the pressure gradient within the melted polymer must be updated by introducing the stress-strain behavior of the fibrous reinforcement. Indeed, the reinforcement is first compressed by the liquid bed when the consolidation pressure is applied and then relaxes as the fluid permeates the reinforcement [3] following Terzaghi's relation. This balance of forces states that the total applied pressure $\sigma_{tot}$ is balanced by the average effective stress in the preform $\sigma_f$ and by the average resin pressure $\overline{p_r}$:

$$\sigma_{tot} = \sigma_f + \overline{p_r} \tag{1}$$

This phenomenon related to the elasticity of the reinforcement when compression occurs, considerably modifies the permeability of the reinforcement during its impregnation and must be considered for a proper modeling of the impregnation process [4]. For this purpose, the hydro-mechanical behavior of a compressible carbon fiber (CF) mat with a thermoplastic matrix was investigated.

## 2 EXPERIMENTAL

### 2.1 Materials and methods

One grade of CF reinforcement in the form of a non-woven mat with a nominal thickness of 2.5 mm and an areal weight of 250 g/m² was studied.

In order to study the fluid-structure interaction between the reinforcement and the thermoplastic matrix, composite plates were manufactured with Poly(Vinylidene Fluoride) (PVDF) combined with the CF mat.

The panels were manufactured using thermocompression molding process based upon the film stacking method. This method consists of heating and compressing a stack of alternating layers of polymer film and dry reinforcement.

## 2.2 Characterization

2.2.1 Compressibility

A compaction rig was designed and manufactured to perform the compression tests on dry and wet mat fibers. The mat samples were cut to a format of 5 cm x 5 cm using a punch die and a swing arm press. After measuring the machine compliance, a normal compression load was applied to the samples up to 10 MPa using a compaction platen with a diameter of 15 cm. The compression set-up is installed in a 30 kN Instron testing machine.

For each test, 4 layers of fabrics were used. Before the compression measurement, the actual areal weight was calculated from the mass of each layer used. The compression test was carried out in three stages. Firstly, the reinforcement was compressed at speeds of 2 and 10 mm/min until the target pressure was reached. The relaxation phase was then observed for 5 min under a constant force. Unloading was then performed at the same rate as the compression stage until zero stress was reached.

For the wet fiber compression, each reinforcing ply was soaked in a bath of distilled water for at least 15 min to ensure complete wetting of fibers. Excess liquid was then drained from the wet plies by placing them on a grid. This step was performed to minimize the effect of excess liquid during the compaction test. After the wetting procedure, the plies were tested in compression mode using the same protocol as for dry samples.

2.2.2 Tensile tests

The tensile tests were carried out on an Instron 5800-R electromechanical machine with a 30 kN load cell, equipped with self-tightening jaws, and an extensometer. The breaking stress was calculated for different manufacturing configurations of the composites. For each configuration, three specimens with a 2 mm tensile area, were tested. The average stress at break was used to observe the effect of the manufacturing configurations on the plates mechanical properties.

# 3 RESULTS AND DISCUSSION

## 3.1 Compressibility

Reinforcement impregnation modeling is performed most of the time assuming that the preform remains rigid. However, in practice, a porous media is compressible and the pressure gradient guiding the impregnation front can significantly deform the reinforcement. In the case of CF mat, the low in-plane orientation of the fibers makes them very compressible and prone to flow induced deformations.

The compressive behavior of the reinforcement can be described by means of the stress required for increasing the fiber volume fraction ($V_f$) within the reinforcement, which can be calculated from the thickness variation of the sample (Figure 1).

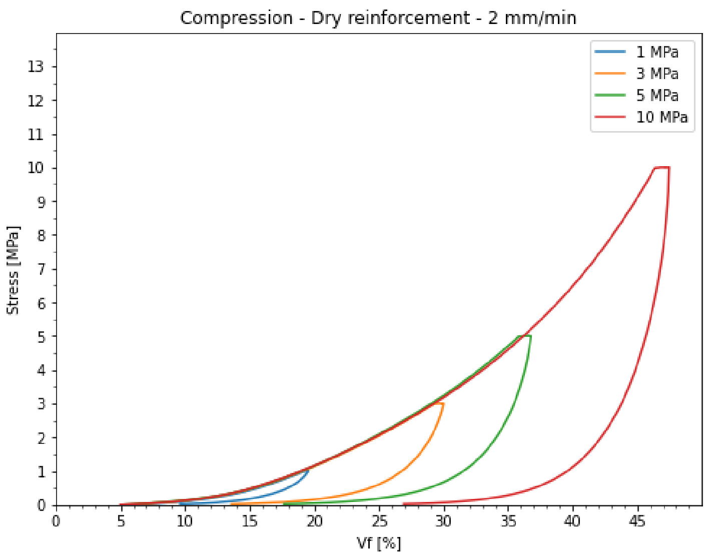


Figure 1: Stress versus $V_f$ curves for the CF mat preform at 4 different compression levels.

As shown in Figure 1, the fiber volume fraction of the reinforcement increases from 6% (*i.e.*, 94% of open porosities in the initial state), until gradually reaching 46.51% under a pressure of 10 MPa and 47.41% after 5 min of creep under the same load.

This particular mechanical behavior can be attributed to fiber rearrangement into a more compact organization during compression. The contacts between fibers take up some of the pressure and change the distribution of the fibers through the thickness of the plate.

A significant hysteresis is observed between the compression and unloading curves, which indicates that the fiber reorganization occurring during compression is not fully reversible. The irreversible behavior of the reinforcement must therefore be considered, in particular for the relaxation phenomena occurring during impregnation. However, during impregnation, the relaxation occurs when the reinforcement is saturated by the melted polymer and the relaxation behavior must be distinguished from the compression step.

In order to analyze the relaxation of the reinforcement in saturated regime, compressibility tests were performed on wet reinforcements. Results show that the maximum $V_f$ reached is higher for the wet reinforcement (29.7%) compared to the dry reinforcement (29.3%) for the same load level (Figure 2). These effects are mainly due to the lubrication which enables an easier rearrangement of the fibers at low stresses and reduces the rupture of fibers at high stresses. The thin layer of lubricant on the surface of the fibers acts as a protective agent, reducing the stress acting on the fibers [5].

In addition to reaching a higher $V_f$ in the case of the wet reinforcement, the creep is more pronounced in wet configuration. During the 5 minutes of holding the load, the fibers reorganize more easily, increasing the $V_f$ after relaxation (+ 0.7% for the dry reinforcement against + 1.4% for the wet reinforcement).

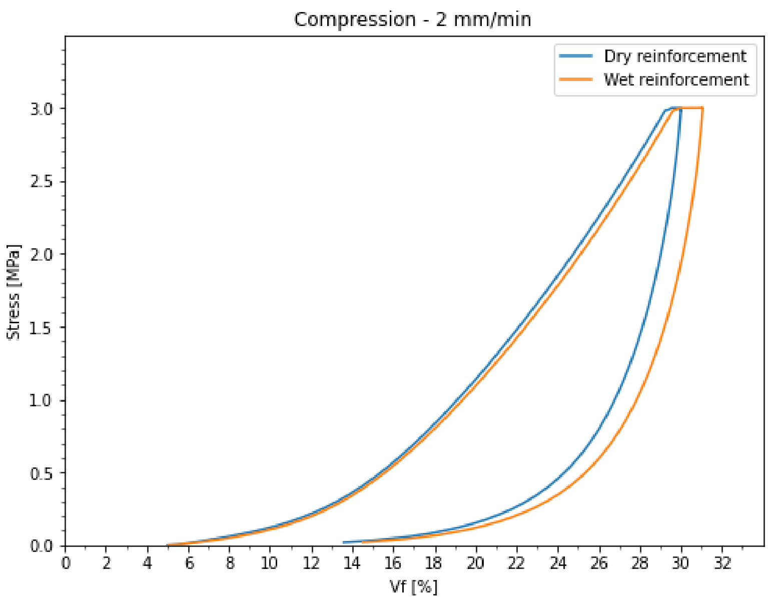


Figure 2: Stress versus $V_f$ curve for the dry and wet CF mat preform under 3 MPa, holding force.

As shown in Figure 3, the effect of the fiber's lubrication is also noticeable during the relaxation phase under a displacement hold, i.e., at constant fiber volume fraction ($V_f = 35\%$). The stress on the dry reinforcements remains higher than for wet one, which confirms the lubricating effect on carbon fibers reorganization. This effect is a crucial element to consider when studying the compression behavior of the material.

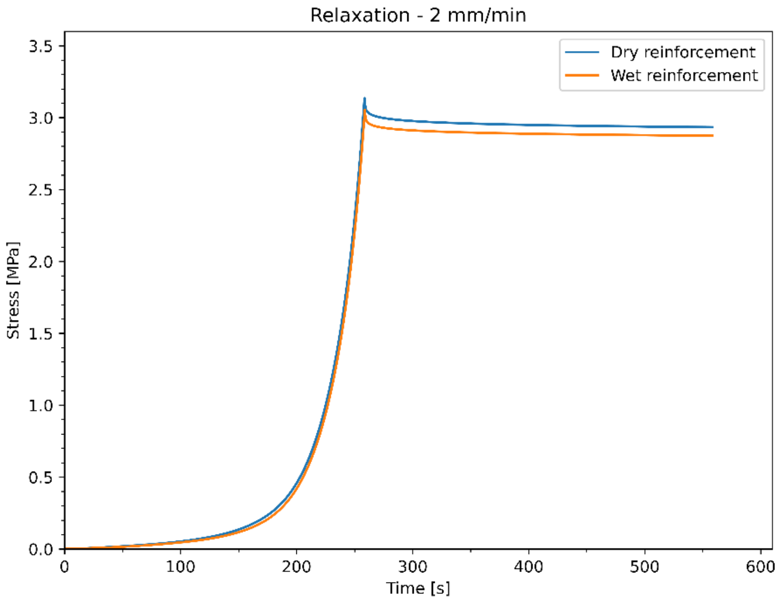


Figure 3: Evolution of the stress during the relaxation phase, holding displacement at $V_f$ = 35%.

### 3.2 Fiber breakage

The irreversible behavior observed during the compression-relaxation tests can partly be attributed to fibers breakage at some fiber-fiber contact points. Indeed, the mat architecture of the reinforcement results in a high level of fiber entanglement (Figure 4). The organization of the fibers leads to many contact points between the fibers during compression. Then, multiple bending stresses occur locally, leading to fiber breakage.

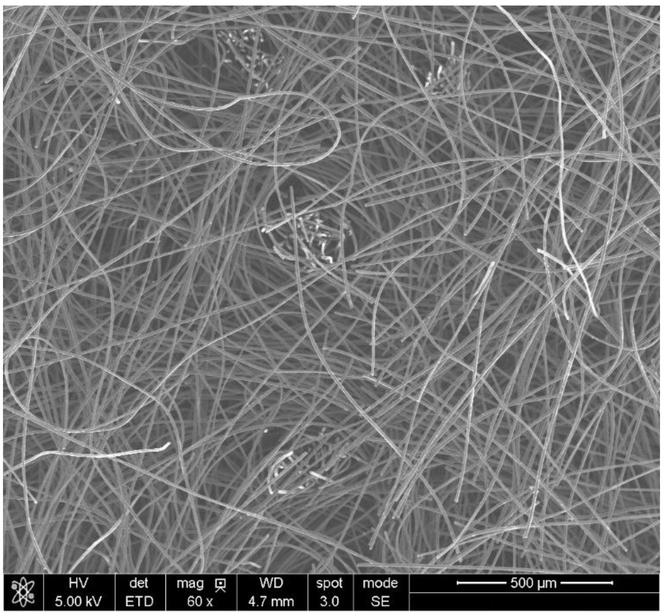


Figure 4: Scanning Electron Microscope (SEM) observation of the carbon-fiber architecture.

The breakage of the fibers during processing is a crucial point of control because it not only influences the consolidation process through the compression and relaxation behavior, but it also affects the final performances of the final composites (mechanical, electrical, physicochemical).

Since compression tests do not allow clear identification of the stress level at which the rupture of carbon fibers occurs, mechanical testing was used to reveal the level of fiber breakage as a function the applied load. To do so, flat composite panels were consolidated under press using a film stacking configuration, but instead of applying the consolidation pressure once the polymer is melted, a first loading and unloading at different levels was achieved at room temperature before heating. All samples were then consolidated at the same consolidation pressure after the initial compression of the CF were carried out. Tensile specimens were then cut from the composite plate and the composites stress-at-break measurements were recorded as they give an indication as to whether the fibers have been broken: the higher the percentage of broken fibers in the material, the lower the stress at the break of the composite.

The results in Figure 5 show that the fiber breakage starts from the first load levels, even low ones, and increases with the applied pressure up to 10 MPa, which is the maximal pressure applied in this study. Thus, for $V_f = 47\%$ corresponding to a compression pressure at room temperature of 10 MPa, the decrease in stress to failure is 19%, while for $V_f = 39\%$, corresponding to a compression pressure at room temperature of 6 MPa, the decrease in stress to failure is 5%.

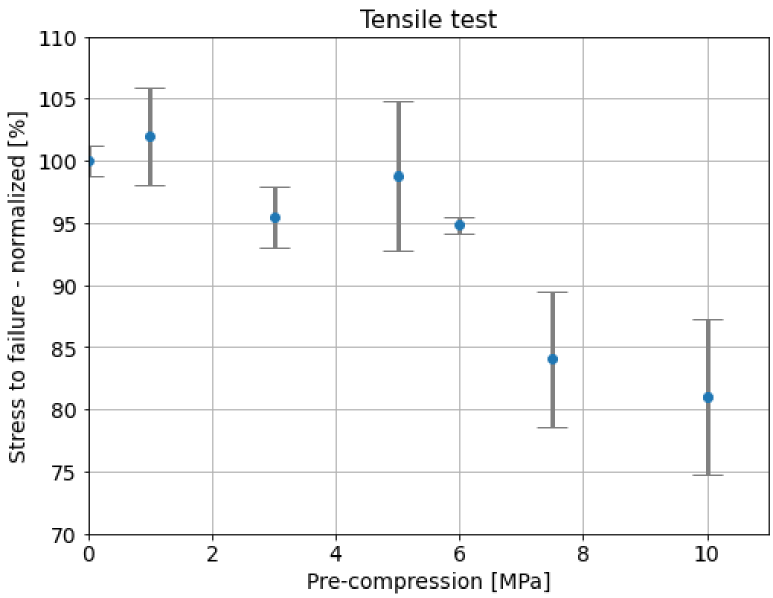


Figure 5: Effect of fiber breakage on tensile stress at break of composites.

In order to consider the effect of fiber breakage in impregnation models, the compression behavior of the reinforcement was modeled with the following power law, where $c$ and $d$ are the coefficients of the power law (Figure 6):

$$\sigma = c\, V_f^d \tag{2}$$

In equation 2, $d$ is a coefficient based on the beam theory and the bending behavior of the fiber. It varies according to the nature of the reinforcement. In the case of mat-type, with a poor organized architecture, the coefficient classically varies between 5 and 8 while it is between 9 and 15 in the case of more ordered reinforcements such as unidirectional (UD) or woven reinforcements [6], [7]. The value of the coefficient $c$ is highly dependent on the compression ratio, due to the viscoelastic response of the reinforcement [5].

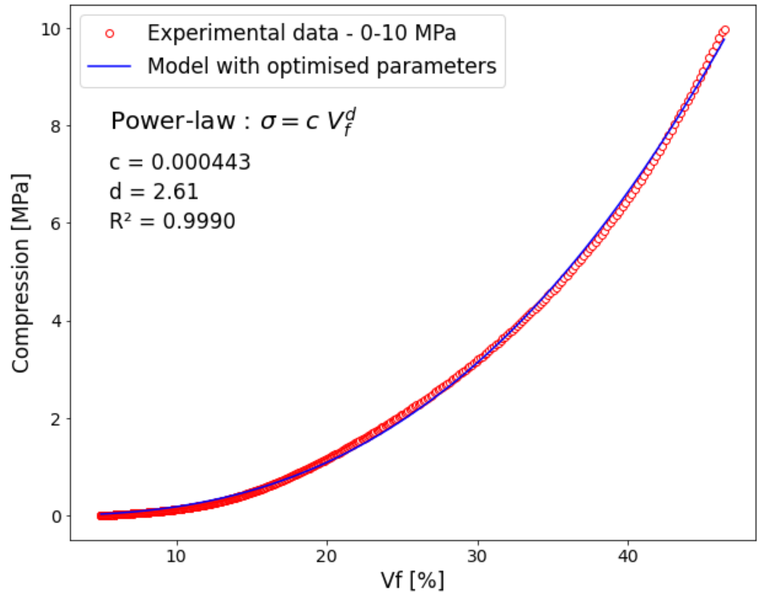


Figure 6: Modeling of the compression behavior of dry reinforcement.

The coefficient $d$, then can be used as an indicator of the level of fiber breakage. It was thus identified for different ranges of pressure starting from the unloaded state, and the values of d coefficients are shown in Figure 7 as a function of the pressure range considered. Globally, in the case of the studied reinforcement, the coefficient is lower than that of more classical mats [6]. The studied mat architecture is indeed less organized, leading to a high reinforcement compressibility. But the coefficient $d$ varies according to the pressure applied to the reinforcement. It is maximal for low pressures and decreases with increasing pressure. As discussed above, the CF breaks progressively with loading (Figure 5). As the fibers break, the architecture of the reinforcement is modified and the value of the $d$ coefficient decreases. The disorganization is increased with the pressure due to the breakage of the fibers.

The effect of lubrication on the compression behavior is small. It does not considerably affect the value of the coefficient $d$, contrary to the compression speed which tends to increase it.

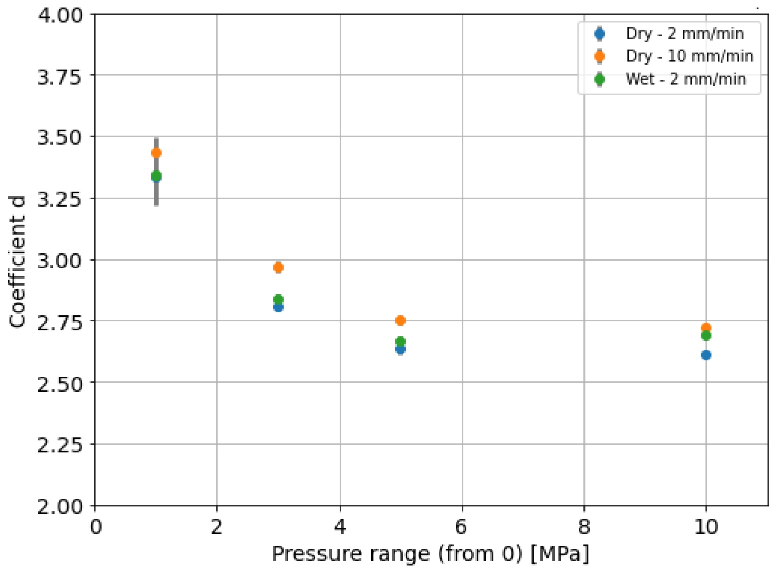


Figure 7 : Evolution of the coefficient $d$ versus the pressure range.

### 3.3 Comparison with manufactured composites

The study of fluid-structure interactions through compression of the reinforcement enables to understand the phenomena occurring during impregnation through the effect of the applied pressure. The results obtained from the characterizations are compared with the actual composites plates manufactured under different processing conditions (Figure 8).

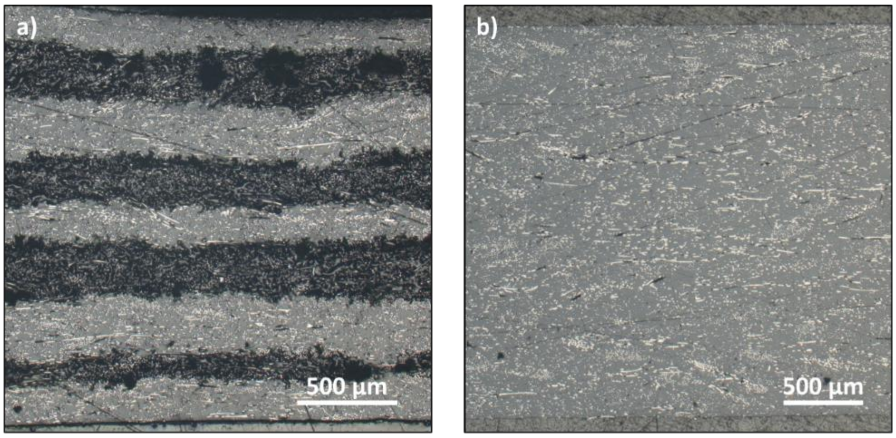


Figure 8: Optical microscope observations of composites manufactured under 1 MPa with a) $M_f$ = 30% et b) $M_f$ = 20%.

The compression behavior of the reinforcement enables to optimize the composite processing conditions. A composite, classically manufactured with a fiber volume fraction of 30%, under a thermocompression pressure of 1 MPa leads to a porosity rate of 33% (Figure 8-a). The observation of the microstructure under optical microscope enables identifying important dry zones, related by 4 dark stripes in the thickness corresponding to the 4 non-impregnated CF plies. In this case, the porosity is not only related to an impregnation problem described by Darcy's law. Indeed, the compression behavior of the reinforcement (Figure 1) indicates that the volume fraction of fibers corresponding to a pressure of 1 MPa is 20%. Thus, the applied pressure does not allow obtaining a composite of 30% mass fraction of fibers with zero-porosity because the amount of polymer is not sufficient to obtain a fully impregnated composite. Indeed, it is mandatory to balance the amount of matrix to the maximum compression level the reinforcement can reach under a specific load. In the present case, at least 80%v of polymers must be introduced so as to fully impregnate the reinforcement under a pressure of 1MPa. By modifying the material ratios of CF and polymer from the compression behavior under 1 MPa, *i.e.*, 20%v reinforcement and 80%v polymer, the resulting composite has no porosity (Figure 8-b). This results in a 12% thickness reduction as the pores occupy more space than the polymer.

## 4 CONCLUSIONS

In order to model the impregnation of a mat reinforcement by a thermoplastic matrix, the compression behavior of the mat was studied. The compression phase showed no significant difference between dry and wet reinforcements. However, the relaxation phase showed the better ability of the fibers to reorganize when lubricated. Similarly, the load carried by the CF is higher when the reinforcement is lubricated. The thin layer of lubricants protects the fibers during compression. The compression step leads to a progressive rupture of the fibers, modifying the value of the coefficient $d$ in the power-law. Measurements of the transverse permeability of the mat, combined with the compression behavior, will enable to set up an advanced model of the permeability up to a model of the impregnation, thanks to Darcy's law.

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