



Effect of mechanical consolidation on adhesion mechanism of hybrid non-woven Alfa fibers reinforced unsaturated polyester composite

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ABSTRACT

Vibrational analysis based on FTIR measurements were performed on hybrid non-woven Alfa fibers reinforced unsaturated polyester composite (HNWAFRUP) and its constituents. The reinforcement was made up of Alfa and wool fibers in the relative volume fractions ratio 4:1. These non-woven fibers sheet were consolidated mechanically by means of the needle punching. This study revealed that the increase of passages numbers in this device allowed the separation of the Alfa fibers and the reduction of the lignin's component in these fibers which decreased its hydrophilic character. Chemical reactions between wool and Alfa fibers were based mainly on the appearance of new vibrations originating from wool fibers in the reinforcement. Adhesion mechanism between the matrix and the reinforcement was established by chemical bonds formed by secondary bonding. Tensile testing performed on this composite revealed the enhancement of its mechanical properties attributed to a less fiber to fiber contact of wool fibers. Nevertheless, the decrease of its specific Young's modulus revealed the mechanical consolidation effect on Alfa fibers/matrix adhesion.

Keywords: Adhesion; Mechanical consolidation; FT-IR; Wool fibers; Hybrid composite.

1. INTRODUCTION

Bio-composites have attracted the attention of many researchers and industrials due to the numerous advantages of natural fibers as reinforcement in these polymeric materials [1]. These composite materials are mostly used in construction [2-6] and automotive [7-9] fields. Nevertheless, the growth of natural-fiber composites cannot be carried out without any challenge. Indeed, the hydrophilic character of this reinforcement could limit its compatibility with the hydrophobic polymeric matrix and hence reduces the mechanical properties of the composite if moisture is absorbed as they age [10]. Interfacial adhesion and resistance to moisture absorption of natural fiber composites can be improved by the suitable modification of fibers surface [11-13] or by modifying matrix to make it more compatible with cellulose fibers [14]. Further, physical treatment based on hybridization with glass fibers provides a method to improve the mechanical properties of natural fiber composites [15]. In our previous work, we have evidenced that hybridization with wool fibers could be considered as ecological surface treatment of Alfa fibers in the unsaturated polyester hybrid composite [16].

The present work is focused on the effect of the mechanical consolidation of non-woven Alfa fibers, using a needle punching, on their adhesion in unsaturated polyester composite. In this study, hybridization treatment of Alfa fibers with wool fibers in the relative volume fraction ratio 4:1 was taken into account. Vibrational analysis based on FT-IR measurements was performed on the composite and its constituents so as to evidence the effect of the passages numbers increase of needle punching on hydrophilic character of the reinforcement and on the chemical interactions occurred between Alfa and wool fibers. The adhesion mechanism was determined by this vibrational study. Tensile testing was carried out on this composite in order to confirm this study.

2. MATERIALS AND METHODS

2.1 Materials

The unsaturated polyester (UP) resin used as the matrix was the same as that used in our previous study [17] and was supplied by Cray Valley/Total. The reinforcement of non-woven fibers (Alfa, wool) was elaborated in the similar steps described in our previous study too [18]. The HNWAFRUP composite was manufactured using the classical 'contact mould method' [19]. Fibers were deposited on the mould and then impregnated with the liquid resin mixed with suitable proportions of methyl ethyl ketone peroxide and cobalt octanone as hardener and catalyst, respectively. The saturated material was then pressed by a roller to remove bubbles. After the hardness of the resin, the composite was withdrawn from the mould. The volume fraction of the reinforcement for this HNWAFRUP composite was 7.4 %.

2.2 Methods

The IR spectra were recorded on a Perkin Elmer UATR two spectrometer, in a reflection mode, between 4000 and 400 cm^{-1} with a resolution of 2 cm^{-1} . The preparation of the sample for this characterization was explained previously [20].

Tensile testing was carried out with a Lloyds dynamometer universal testing machine as per NF T 57-301 at a crosshead speed of 5 mm/min and a gripping length of 100 mm. The composite was cut out in the direction of nonwoven production. Obtained results were calculated as the average of ten samples.

3. RESULTS AND DISCUSSIONS

Figure 1 illustrates FTIR spectra of the reinforcement and its constituents (Alfa fibers and wool fibers). Substantial changes between spectra of Alfa fibers and the reinforcement were noticed. These could be attributed to the mechanical consolidation effect using the needle punching and the hybridization effect with wool fibers. The principal IR absorbance peaks obtained for Alfa fibers were identified as follows: (i) the broad band at 3330 and 3284 cm^{-1} attributed to the stretching vibration of the O–H bonds, (ii) the bands at 2916 and 2850 cm^{-1} assigned to the methylene C–H asymmetric and symmetric stretching vibrations of cellulose, respectively, (iii) the bands at 1590 and 1423 cm^{-1} due to the C=C bond elongation of the lignin aromatic ring [21], (iv) the band at 1638 cm^{-1} attributed to the stretching vibration of water molecules due to the hydrophilic character of the lignocellulosic fibres [22], (v) the alcohol C–O stretching vibration observed at 1025 cm^{-1} [23], (vi) the 1335 and 1369 cm^{-1} bands assigned to the bending of –OH groups in the cellulose component [24], (vii) the band at 898 cm^{-1} attributed to the C-1 group frequency or ring frequency, characteristic of β -glycosidic bonds between glucose units [25], (viii) the 875 cm^{-1} band due to the C-H out-of-plane bend of the lignin aromatic ring [26] and (ix) the band at 660 cm^{-1} associated with the OH out-of-plane bend.

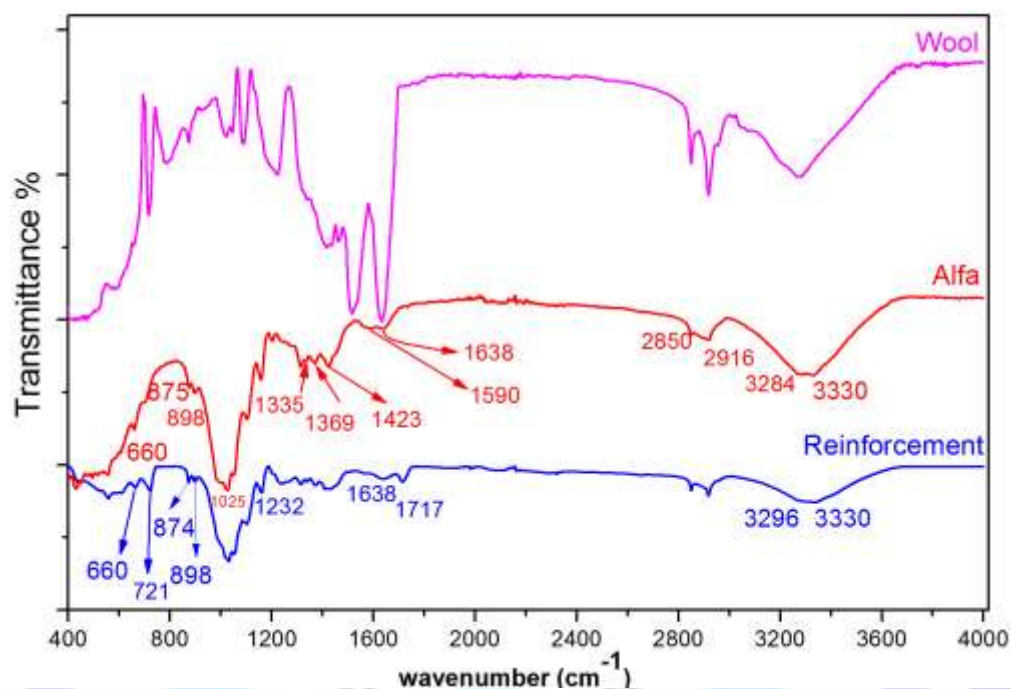


Fig 1: FTIR spectra of the reinforcement and its constituents

The non-woven Alfa fibers were consolidated mechanically using the needle punching four times. The increase of the passages numbers in this device, in comparison with our previous work [16] led to the decrease of the ν_{OH} vibration intensity located at 3330 and 3284 cm^{-1} . Hence, the disruption of hydrogen bonding in the network structure, allowed the separation of Alfa fibers. Similar behavior is obtained by the alkaline treatment which removes a certain amount of lignin, wax and oils covering the external surface of the fiber cell wall [27]. Analysis of the vibration σ_{OH} at 1335 cm^{-1} confirmed also the separation between the Alfa fibers. Indeed, the corresponding spectra are compared assuming Gauss- and Lorentz-shaped bands for the Alfa fibers and the reinforcement, respectively, as depicted in figure 2-(a, b). The relative intensity $\left(\frac{I_{\text{reinforcement}}}{I_{\text{Alfa}}} \right)_{\sigma_{\text{OH}}}$ was reduced to 0.54 which could be attributed to the disruption of hydrogen bonding

occurred in web structure of Alfa fibers. Further, the disappearance of the vibration $\nu_{\text{C=C}}$ at 1590 cm^{-1} indicate the elimination of a certain amount of lignin. This remove gave rise to a new vibration at 1717 cm^{-1} originating from the cellulose [26]. The fit of the band at 1638 cm^{-1} for Alfa fibers and the reinforcement were illustrated in figure 3-(a, b), respectively. Hence, the obtained relative intensity $\left(\frac{I_{\text{reinforcement}}}{I_{\text{Alfa}}} \right)_{\text{absorbed water}}$ is reduced to 0.19. This reduction

was attributed to a less hydrophilic character of the reinforcement due to the remove of a certain amount of lignin. The presence of wool fibers in the reinforcement gives rise to additional vibrations such as: (i) the peak at 1232 cm^{-1} attributed to the N-H deformation of the amide III band and (ii) the peak at 721 cm^{-1} associated with the primary amide, NH bend [26]. Also, it gives the intensification of some vibrations such as : (i) the band at 874 cm^{-1} which can originate from the symmetric stretching CNC and (ii) the band at 660 cm^{-1} which originating from the C–S stretching vibrations.

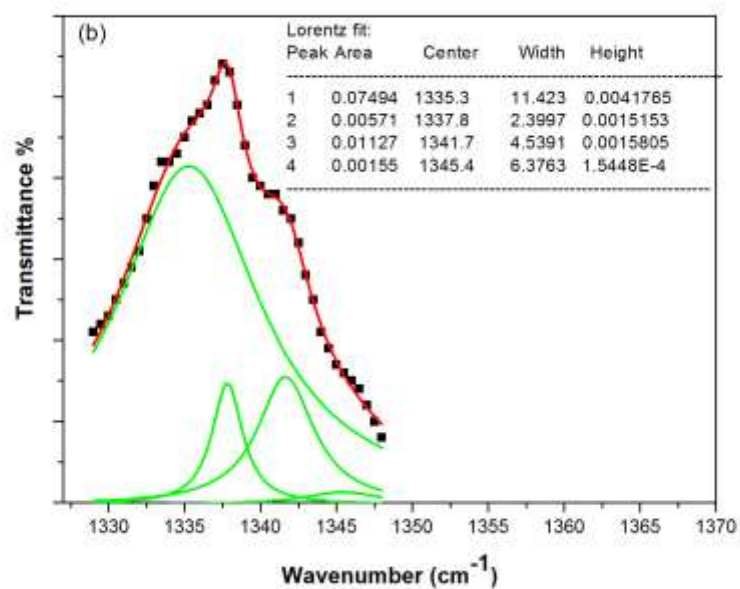
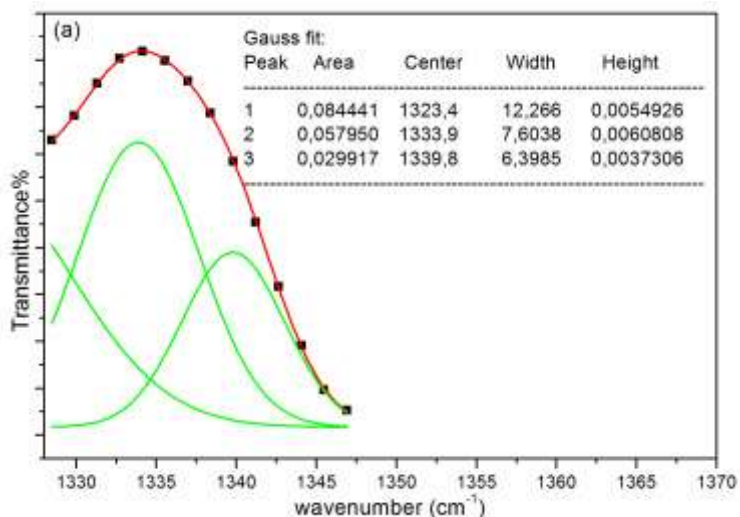
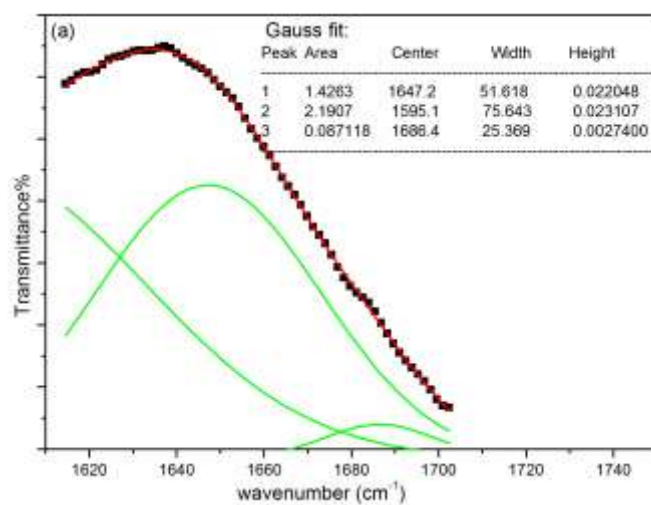


Fig 2: Peak fit of OH groups bending at 1335 cm⁻¹: in Alfa fibers (a) and the reinforcement (b)



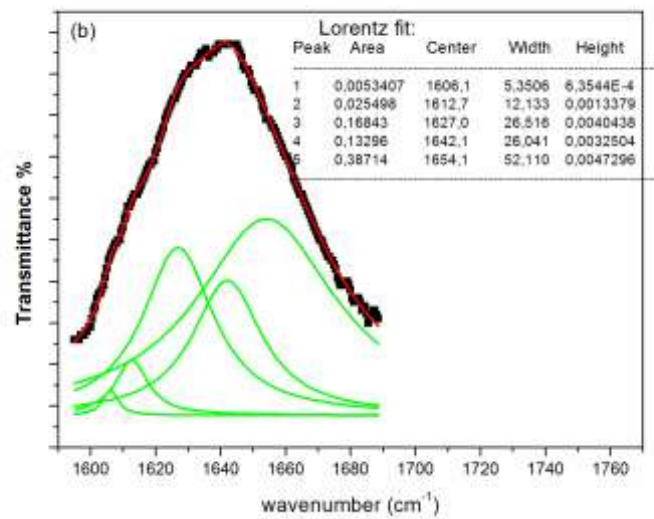
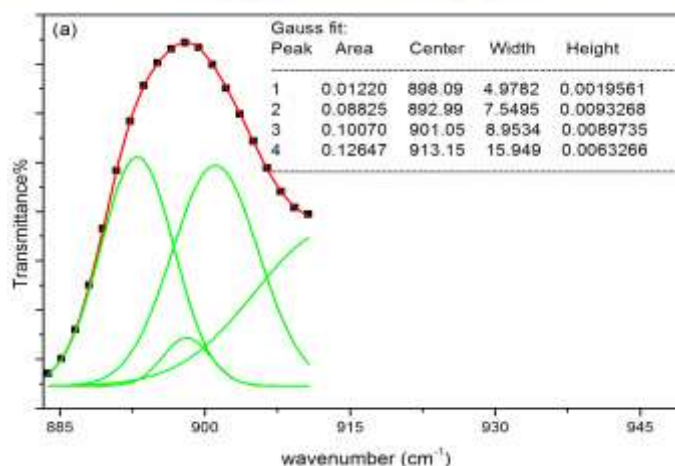


Fig 3: Peak fit of water stretching vibration situated in 1638 cm⁻¹: in Alfa fibers (a) and the reinforcement (b)

The analysis of the band at 897 cm⁻¹ is illustrated in figure 4-(a, b) by the comparison of the spectra assuming Gauss- and Lorentz-shaped bands for Alfa fibers and the reinforcement, respectively. Consequently, the relative intensity $\left(\frac{I_{\text{reinforcement}}}{I_{\text{Alfa}}}\right)_{\beta\text{-glycosidic bonds}}$ is enhanced to 1.59. As a result, both mechanical consolidation and hybridization

with wool fibers effects can be considered as surface treatments of Alfa fibers. In the purpose of confirming the effect of these surface treatments on adhesion of Alfa fibers in the matrix, FT-IR spectra of the UP and its HNWAFRUP composite are shown in figure 5, with the reinforcement spectrum as illustration. The principal vibration peaks obtained for the UP matrix are as follow: (i) the peak around 3524 cm⁻¹ attributed to the stretching vibrations of hydroxyl (OH) groups, (ii) strong peaks at 3081, 3059 and 3025 cm⁻¹ assigned to C-H stretching, (iii) the peaks at 2849 and 2924 cm⁻¹ assigned to the symmetric and anti-symmetric stretching vibrations of the methylene group (CH₂) respectively, (iv) a very intense peak observed at 1720 cm⁻¹ due to the carbonyl (C=O) stretching from the ester linkage, (v) peaks at 1600, 1580, 1493 and 1451 cm⁻¹ attributed to the C=C stretching vibrations within the aromatic ring, (vi) several and strong peaks at 1256, 1118, 1065, 1040 and 982 cm⁻¹ due to the aromatic C-H in-plane bend and (vii) peaks at 844, 741, 697 and 651 cm⁻¹ assigned to the aromatic C-H out of plane bend [12]. It is to be noted that the band characteristic of the valence vibration of the O-H bond in UP matrix at 3524 cm⁻¹ is shifted to lower wavelengths, around 3452 cm⁻¹ for its composite. In addition, the width of this band is larger in composite than in pure UP. These results indicate the presence of hydrogen bonds between the O-H bonds of the matrix and (O-H, N-H) bonds of the reinforcement. There is also a decrease in the intensity of the valence vibration band of the C=O at 1720 cm⁻¹. Indeed, its relative intensity $\left(\frac{I_{\text{composite}}}{I_{\text{matrice}}}\right)_{\text{C=O}}$ is reduced to 0.54. The obtained

fit spectra are compared assuming Gauss-shaped bands as depicted in figure 6-(a, b). This indicates the hydrogen bond formation between the reinforcement and the matrix [26, 28]. This hydrogen bond interaction could improve their interfacial interaction and enhance compatibility. Moreover, the decrease of the bands intensities at lower frequencies can be explained by the existence of a chemical reaction between the matrix and the reinforcement.



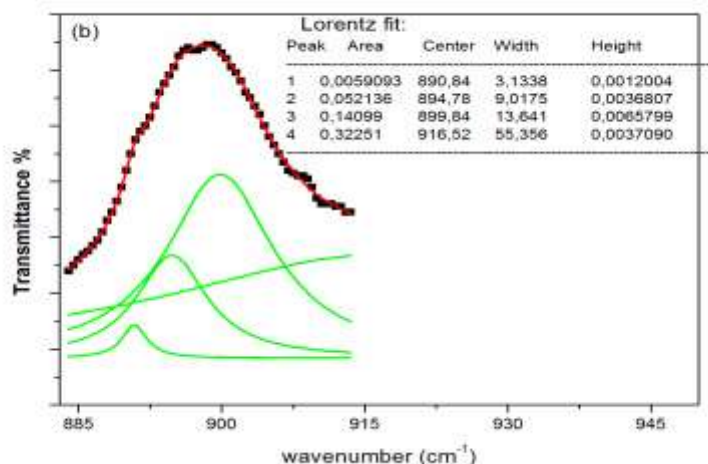


Fig 4: Peak fit at 897 cm^{-1} : in Alfa fibers (a) and the reinforcement (b)

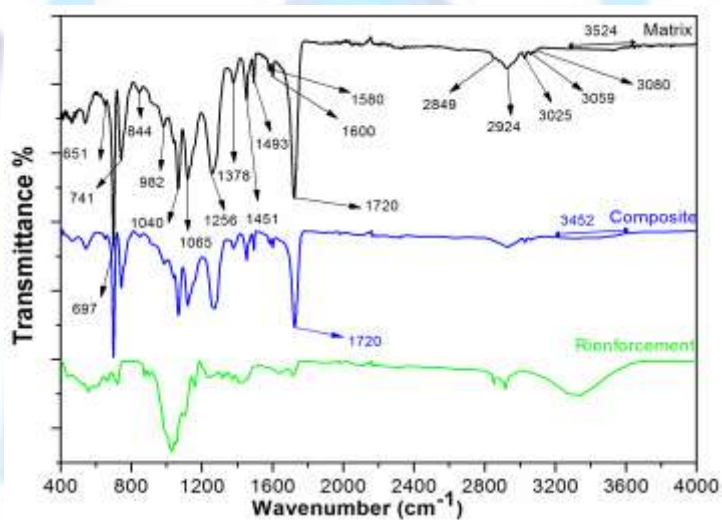
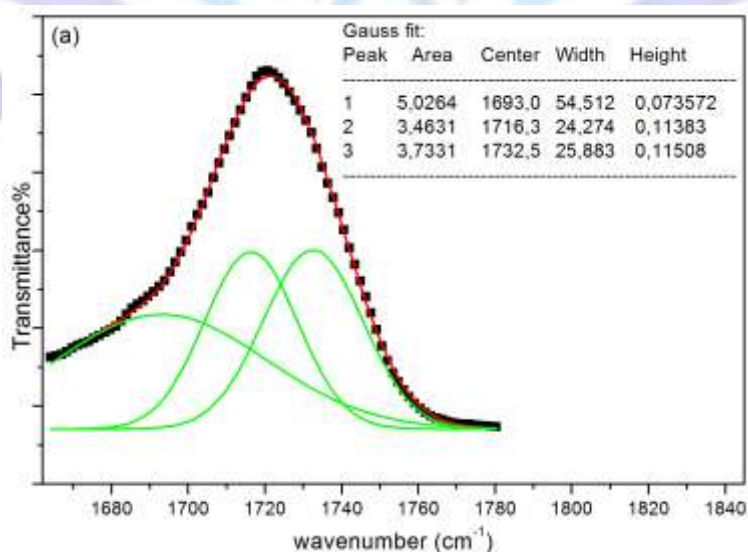


Fig 5: FT-IR spectra of the HNWA FRUP composite and its constituents (the matrix and the reinforcement) in the wavenumber range 400-4000 cm^{-1}



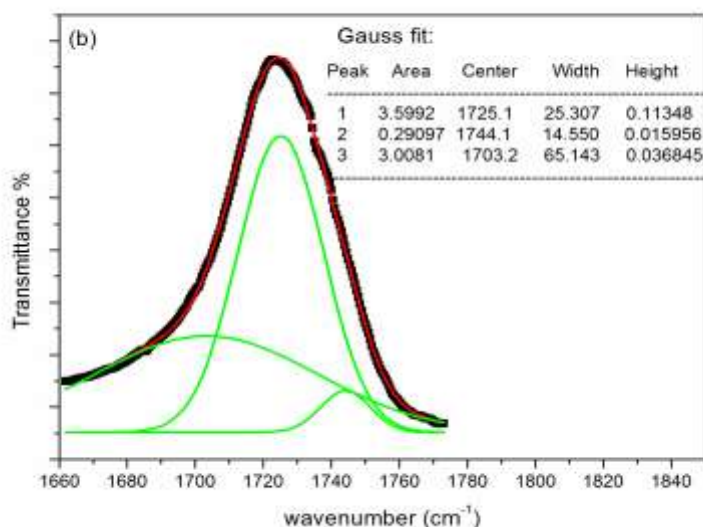


Fig 6: Peak fit of carbonyl stretch situated in 1720 cm⁻¹ in: matrix (a) and HNWAFRUP composite (b)

Tensile properties of the HNWAFRUP composite were illustrated in Table 1. These results were compared to those in our previous study [16]. This comparison revealed an enhancement of the tensile properties of the HNWAFRUP composite which can be attributed to the less fiber to fiber contact of wool fibers. Hence the increase of passages numbers in the needle punching improves the interfacial bonding between wool fibers and the matrix. As results the failure tensile strength, stress and failure increase. However, the decrease of the specific Young's modulus of HNWAFRUP composite could be explained by a lower adhesion of Alfa fibers in the matrix in comparison to that occurred in the Alfa fibers reinforced unsaturated polyester (AFRUP) composite [16]. As a result the hybridization effect gave rise a better adhesion in the AFRUP composite. This effect was overlapped by the mechanical consolidation one in the HNWAFRUP composite.

Table 1. Tensile properties of HNWAFRUP and AFRUP composites.

Composites	ϵ (%)	F (N)	σ_t (MPa)	E_t (GPa)	σ_t/ρ (MPacm ³ /g)	E_t/ρ (GPa cm ³ /g)
HNWAFRUP	2.97	761.57	16.90	0.801	21.2	1.01
AFRUP [16]	2.29	547.24	8.76	0.697	18.3	1.45

F (N), σ_t , ρ and E_t are the tensile strength at break, the stress at break, the density and the Young's modulus of the composites, respectively.

4. CONCLUSIONS

FTIR analysis of the HNWAFRUP composite and its constituents revealed that mechanical consolidation of the non-woven Alfa fibers, using the needle punching, contributes to its adhesion in the UP matrix. Indeed, this analytical study revealed that the increase of passages numbers in the needle punching could reduce the hydrophilic character of Alfa fibers by removing certain amount of lignin. Furthermore, it allows Alfa fibers separation which affects its chemical interactions with wool fibers. Tensile testing showed an improvement of HNWAFRUP composite tensile properties attributed to wool fibers/matrix adhesion enhancement. Nevertheless, its specific Young's modulus decrease in comparison to our previous study could be associated with a lower Alfa fibers/matrix adhesion occurred by the mechanical consolidation.

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