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A Pull-out Fiber/Matrix Interface Characterization of Vegetal Fibers Reinforced Thermoplastic Polymer Composites: The Influence of the Processing Temperature

Duy Cuong Nguyen, Ali Makke, Guillaume Montay

***Abstract*—This work presents an improved single fiber pull-out test for fiber/matrix interface characterization. This test has been used to study the Inter-Facial Shear Strength 'IFSS' of hemp fibers reinforced polypropylene (PP). For this aim, the fiber diameter has been carefully measured using a tomography inspired method. The fiber section contour can then be approximated by a circle or a polygon. The results show that the IFSS is overestimated if the circular approximation is used. The Influence of the molding temperature on the IFSS has also been studied. We find that a molding temperature of $183°C$ leads to better interfacial properties. Above or below this temperature the interface strength is reduced.

***Keywords*—Interface, pull-out, processing, temperature, hemp, polypropylene,composite.

I. Introduction

THE vegetal fibers such as hemp,sisal,bamboo... have become an attractive candidates to substitute the conventional fibers commonly used in polymer based composites. The use of such fibers as a polymer reinforcement leads to many environmental advantages and economic benefits. (i) The vegetal fibers originate from renewable resources. (ii) There is no transformation industry required. (iii) In some cases the resulting composite material can be recycled several times.

It's well known that the mechanical properties of composite materials depend strongly upon the coupling between the fibers and the matrix at the interface. A weak interface coupling decreases severely the composite strength. This issue requires more attention in our study because of the weak coupling between the hemp fibers and the PolyPropylene 'PP' matrix. Therefore, the composite damage is triggered by the interface fracture in the most cases.

To understand and improve the mechanical behavior of hemp fiber reinforced polypropylene composites, a through investigation of fiber/matrix interface properties is required. Then, InterFacial Shear Stress 'IFSS' has to be characterized carefully.

M. Duy Cuong Nguyen is with the Laboratory of Mechanical Systems and Concurrent Engineering LASMIS, University of Technology of Troyes, 10004 Troyes Cedex, France (e-mail : duy_ cuong.nguyen@utt.fr).
M. A. Makke EPF-Ecoles d'Ingénieurs, and LASMIS of University of Technology of Troyes, 10004 Troyes Cedex.
M. G. Montay is with the Laboratory of Mechanical Systems and Concurrent Engineering LASMIS, University of Technology of Troyes, 10004 Troyes Cedex.

To this end, several experimental methods have been developed such as: the fragmentation test [3], [16], the pull-out test [13]–[15], the micro-bond test [9], [11], the push-out test [1], [8].

However, these methods are usually expensive and need a long time to prepare carefully the samples. These shortcomings limit severely the use of such tests for industrial purpose. To address this issue, a single fiber pull-out test has been developed in order to simplify the interface characterization process.

Another problem encountered when IFSS is calculated is how the fiber/matrix contact area is determined. In the most research works, the fiber section is usually supposed circular and homogeneous along its length [6], [10]. In addition, the fiber diameter is only measured in some points and averaged along the fiber length. Recently, Ilczyszyn F. and al. [5] have developed a new approach to better describe the fiber geometry. This approach takes into account the irregular shape of the fiber section.

In this work, we use the developed single fiber pull out test in order to determine the IFSS in the hemp/PP composite. First, we describe the sample preparation procedure and the developed pull out test. After, we study the mechanical response of a sample and we show how the IFSS can be determined from the test results. The influence of molding temperature on the IFSS will also be presented. At the end the results are summarized and discussed.

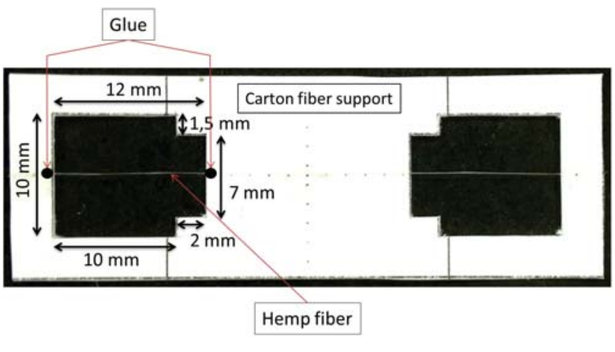

Fig. 1: The carton fiber support. The two gaps allow one to prepare two samples paper support

II. Materials and Method

A. Specimens Preparation

Hemp fibers have manually been extracted from stalks nuance E40 (cultivated in Champagne-Ardennes region in France) and glued on a carton support as shown in Fig. 1. The carton gap dimensions was taken equal to $10 \times 12mm$ in order to have $2mm$ free length (The length of the free part of the fiber as shown in Fig. 6). Each carton support is used to prepare two samples, thus two fibers are glued per support.

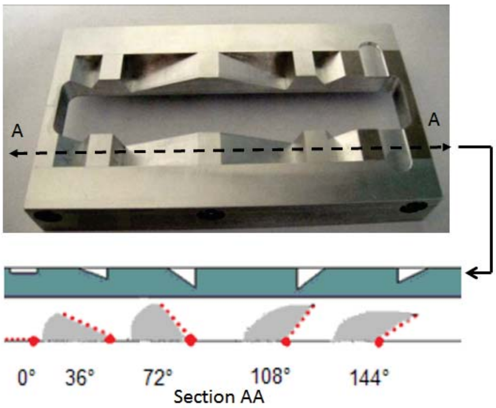

Fig. 2: The photo shows the stand which is used to orient the fiber carton support in five distinct angles. This stand is inserted under a digital microscope to capture the fiber profile in each angle as shown in the scheme. This figure is reproduced from [4]

1) Fiber Diameter Measurement: The carton support is mounted on a specific stand to capture the fiber profile (as shown in Fig. 2). This stand allows one to rotate the carton support in five angles: $0°$, $36°$, $72°$, $108°$ and $144°$ (as shown in Fig. 3). A digital microscope is used to get the fiber profile. Thus five photos that correspond to five projection angles were taken for each fiber. A digital image processing algorithm has been developed to analyze these photos. Thanks to this algorithm, the fiber surface is carefully determined.

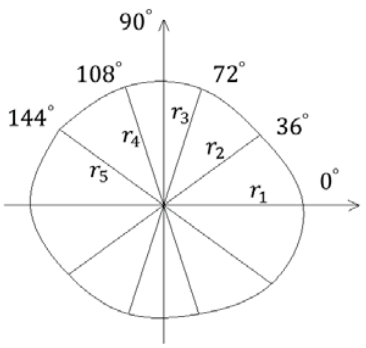

Fig. 3: Schematic representation of a hemp fiber section. The lines indicate the fives measured diameters

Two methods can be used to approximate the fiber section contour length:

1) The fiber section contour can be considered as a circle. The contour length is the circle perimeter.

$$P = 2\pi.\langle \overline{r} \rangle \tag{1}$$

where $\langle \overline{r} \rangle$ is the double averaged radius of the fiber. First, the radius is averaged along the fiber length ‘l’.

$$\overline{r_i} = \frac{1}{l}\int_0^l r_i(l)dl \tag{2}$$

The sub-index ‘i’ designates the capture angle index of the fiber. $1 \leq i \leq 5$. Secondly, the radius is averaged over all capture angles.

$$\langle \overline{r} \rangle = \frac{1}{5}\sum_{i=1}^{5} \overline{r_i} \tag{3}$$

2) In the second method, the fiber contour is approximated by a decagon. The contour length is taken as the sum of the decagon sides length as follow:

$$P = \sum_{i=1}^{5} \left(\overline{r_i}^2 + \overline{r_{i+1}}^2 - 2.\overline{r_i}.\overline{r_{i+1}}.\cos(\overline{r_i}, \overline{r_{i+1}}) \right) \tag{4}$$

The two methods are compared in the next. It should be noted that for both methods the fiber geometry is supposed to be a homogeneous extrusion of the averaged contour. Then, the contact area between the fiber and the matrix can be calculated as follow:

$$S_{int} = P \times l_e \tag{5}$$

where l_e is the fiber length embedded in the matrix.

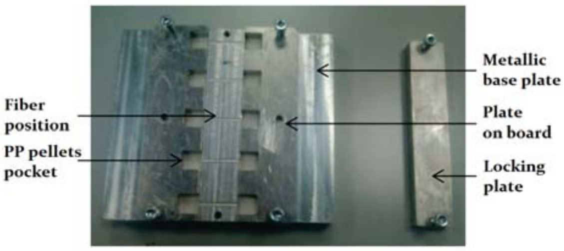

Fig. 4: The specific mold used to melt the PP granules and to elaborate the samples. 10 samples are prepared on each molding step

2) Molding Procedure: In order to increase the molding efficiency, a high number of samples has to be produced simultaneously. To this end, a specific mold has been designed. This mold allows the molding of ten samples in parallel. Fig. 4 shows the used mold.

The sample fabrication is performed as described in the following steps:

- Five carton supports are mounted in the mold. Two hemp fibers are glued on a support. The supports are maintained on the mold by a locking plate as shown in the Fig. 4.
- The mold cavities is filled by PP granules.
- After that, the mold is introduced in an oven. The oven temperature is maintained at $173°C$, during 42 minutes to ensure a full melting of the PP granules.

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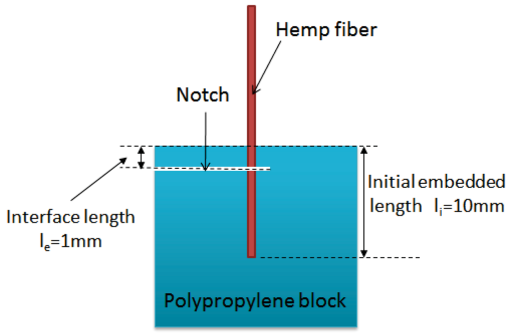

Fig. 5: A sketch of the resulting sample, the scheme is no at scale

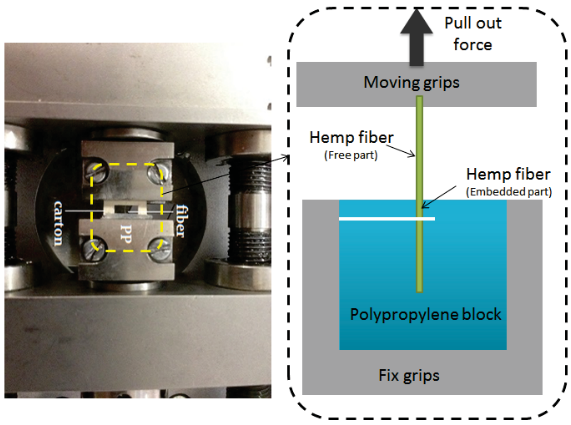

Fig. 6: Pull-out test : the photo and the sketch show how the sample is mounted in the tensile test machine

- After melting, the mold and specimens is cooled at room temperature.
- At the end, the embedded length of the fiber in the matrix have to be adjusted. Thus, The sample is cut at a level of 1mm from the PP boundary with the help of low speed saw. At the end we get a 1mm embedded length (called also interface length l_e hereafter).
  Fig. 5 shows a resulting sample sketch.

The fabrication procedure described above allows one to fabricate up to 40 samples per day.

B. Pull-out Test Procedure

In order to measure the IFSS, we must measure the maximum force that should be applied on the fiber to extract them from the matrix block. To this aim, an adapted tensile test machine has been used. Fig. 6 shows the pull out test procedure. The tensile speed is very low $1\mu m/s$. The tensile test is stopped when the fiber is fully extracted from the PP block.

The pull out test was followed by a digital microscope. The length l_1 of the fiber will be measured after the test is finished. The real embedded fiber length l_e can then be calculated as follow :

$$l_e = l_1 - l_0 \tag{6}$$

where l_0 is the length of the free fiber part of the sample at the beginning of the test.

The apparent IFSS (τ) is calculated following the formula:

$$\tau_{app} = \frac{F_{max}}{S_{int}} \tag{7}$$

where F_{max} is the maximum force reached in the pull-out test prior to the fiber extraction. and S_{int} is the contact area between the fiber and the matrix defined above in (5).

III. Results and Discussion

A. Fiber Diameters

The vegetal fibers have a strong fluctuation in their geometry. To calculate accurately the interfacial shear strength IFSS, the fibers diameter has been carefully measured before and after the pull-out test. These measures were averaged over 13 samples that exhibit a full fiber extraction in the pull-out test. We obtain a diameter of $58.73 \pm 19.11\mu m$ (before) and $54.89 \pm 16.65\mu m$ after the fiber is completely pulled out.

This decrease in fiber diameter can be caused by several factors such as: (i) the thermal shrinkage due to matrix cooling after the molding. (ii) the material loss due to the inter-phase layer or the inter-diffusion phenomenon (iii) The radial contraction of the fiber due to the Poisson effect.

B. The Interfacial Shear Strength - IFSS

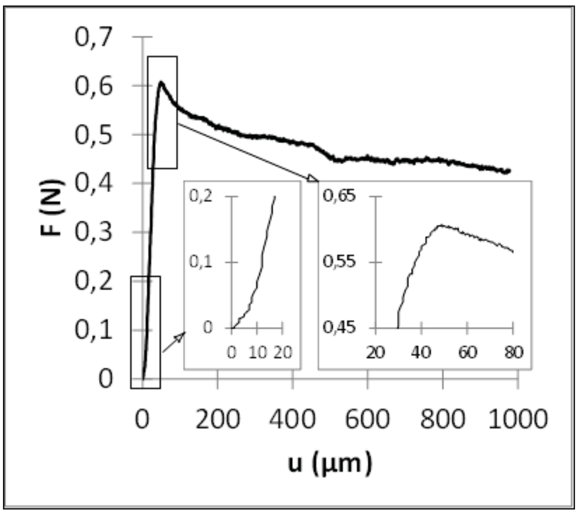

Fig. 7: A typical force-displacement curve of a sample. Two regimes can be clearly distinguished: (i) $0 \leq u \leq 49\mu m$ The force increases linearly when the displacement increases. This behavior is caused by the elastic response of the sample (ii) The force reaches a maximum at $u = 49\mu m$; after that, it starts to decrease. This behavior corresponds to the fiber extraction from the PP block. The measured force in this regime corresponds to the friction between the fiber and the matrix at the interface

TABLE I: Comparison between the average of the IFSS values calculated with circular and polygonal

-	Circular contour	Polygonal contour
IFSS (Mpa)	3.26 ± 1.15	3.09 ± 1.11

1) Pull-out Test Result: The force-displacement curve that results from the pullout test shows three different loading stages: (i) fibrils loading: The fibrils are the constitutive elements of a fiber. when the fiber is submitted to a tensile strain, the fibrils become oriented according to the loading force direction. This behavior can be shown at the beginning of the tensile curve, when the force increase is non-linear. The second stage is (ii) the fiber and the interface loading. The force increase linearly with respect to the displacement. This increase results from the elastic behavior of the sample.

The force reaches a maximum $F_{max} = 0,61N$ when $u = 49\mu m$ and after it starts to decrease (as shown in Fig. 7). In the third stage, (iii) the decrease of the tensile force is triggered by the onset of fiber extraction from the matrix block. At the beginning of this stage, an adhesive fracture takes place at the interface, then the fiber is detached from the matrix. With increasing the tensile displacement, the fiber starts to slip in the matrix. The measured force in this case corresponds to the friction between the fiber and the matrix. These stages have been also observed in several works [2], [7], [12].

2) Maximum Pull-out Force Dependence: The pull out test has been performed on 19 samples. In all tests, the fiber was fully extracted from the block of PP.

The force threshold to extract the fiber F_{max} is found to be largely dispersed. However, it depends upon the fiber diameter $2\langle\overline{r}\rangle$, the interface length l_e and the contact area S_{int}. Fig. 8 shows these F_{max} dependence with these parameters.

3) The Influence of the Contact Area Calculation Method on the IFSS: Equation (7) shows that the IFSS depends upon the fiber-matrix contact area. In section II-A1, we have detailed two methods to calculate the fiber section contour (1,4) and then the contact area (5). The two methods are compared. The resulting IFSS averages are shown in table I. The contact fiber/matrix area calculated by the polygonal approximation leads to a lower IFSS value. A consistent results were found in reference [7].

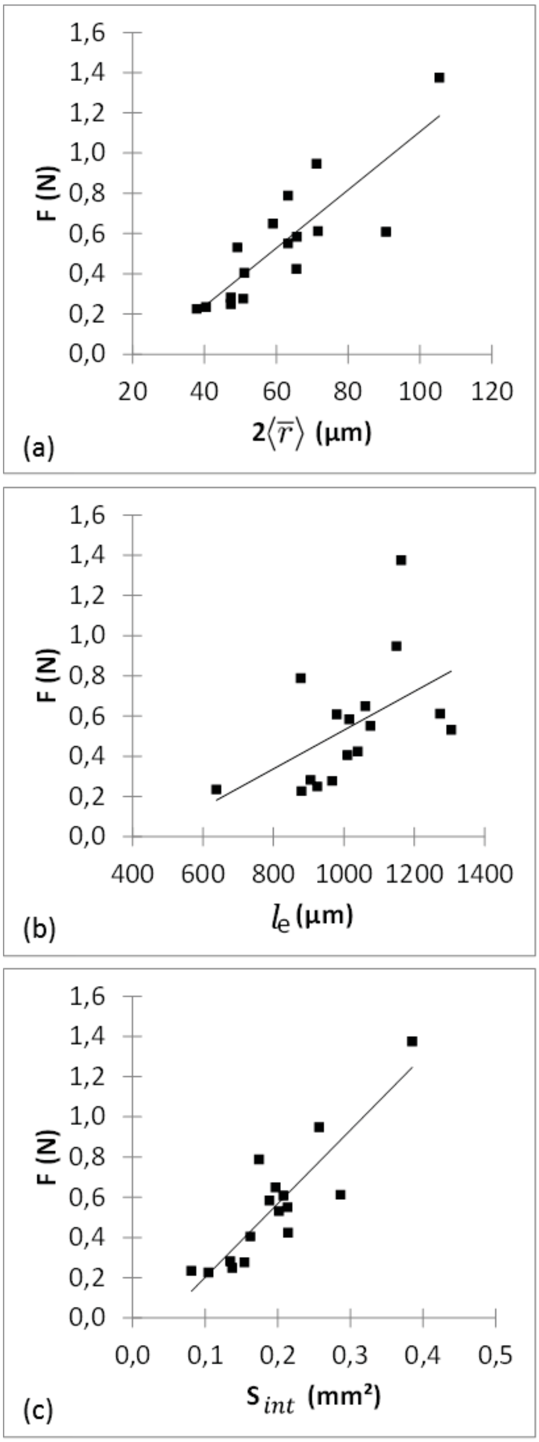

Fig. 8: The force with respect to the fiber diameter (a), Interface length (b) and fiber matrix contact area (c). Each point corresponds to the peak of the force-displacement curve shown in figure 7. The lines capture the points tendency.

C. Influence of the Fabrication Temperature on the IFSS

The influence of the fabrication temperature on the IFSS has been studied. Samples have been made using the same preparation process described above but the molding temperature is modified. Three molding temperatures were considered: $173°C$ (results above), $183°C$ and $202°C$. Pull-out tests have been performed on these samples. As a consequence, we get subsequently 19, 20 and 16 samples that exhibit a full fiber extraction from the matrix. The averaged IFSS values were calculated for each case. The results are compared in table II.

The results shows that the molding temperature $T = 183°C$ leads to optimal interface properties in our case. The highest $\langle IFSS\rangle$ was found at $T = 183°C$, $\langle IFSS\rangle = 4,0 \pm 1,70Mpa$. The $\langle IFSS\rangle$ value decreases below and after this temperature.

The increase of molding temperature from $173°C$ to $183°C$

TABLE II: IFSS with respect to the molding temperatures

Temperature	$173°C$	$183°C$	$202°C$
IFSS (Mpa)	3.1 ± 1.1	4.0 ± 1.7	3.9 ± 1.03

will improve the IFSS. This can be interpreted by several factors:

- The increase of molding temperature will decrease the PP melt viscosity. This effect will enhance the polymer melt diffusion in the fiber asperities. As a consequence, the hanging between the fiber and the matrix is improved.
- In the other hand, the increase of temperature will increase the cellulose chains mobility of the fiber wall. During the molding, an inter-diffusion occurs between the cellulose chains of the fiber and the PP chains of the matrix. Thus, a strong inter-phase results. In this case, the cellulose chains behave as strong anchors that bond the fibers with the matrix.

In contrast, an excessive increase of the molding temperature will annihilate the fiber mechanical properties. This phenomenon has several origins:

- At high temperature, the molecular weight of the cellulose chains in the cell wall of fiber decreases. Therefore the fiber is weaken.
- The loss humidity in the fiber at high temperature increase the fiber embrittlement. The higher the temperatures at which molding performed, the stronger this effect becomes.

The alteration of fiber properties at high temperature leads to a weak interface.

As discussed below, the temperature has two contradictory influences on the IFSS. Therefore, an optimal molding temperature is requested for a better composite mechanical properties.

IV. Conclusion

An improved pull-out test has been used in order to measure the InterFacial Shear Strength IFSS of hemp fiber reinforced polypropylene. The sample preparation procedure was simplified and the preparation time is reduced. The fibers geometry has been characterized using a digital microscope with the help of an image processing algorithm. To measure the fiber section contour, two methods were compared: The circular and the polygonal approximation. The results show that the commonly used circular approximation leads to an overestimation of the IFSS.

The pull-out force to extract the fiber from the PP block shows an important statistical fluctuation depending on the fiber geometry (fiber diameter, embedded length and the contact area between the fiber and the matrix).

We have found also that the molding temperature has a strong influence on the IFSS values. The temperature $183°C$ leads to a better IFSS and then an improved interface. Above or below this temperature the IFSS decreases. This observation was interpreted with respect to the PP melt viscosity and the fiber cell wall degradation.

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