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### ► To cite this version:

M C Seghini, F Touchard, F Sarasini, L Chocinski-Arnault, D Mellier, et al.. Interfacial adhesion assessment in flax/epoxy and in flax/vinylester composites by single yarn fragmentation test: Correlation with micro-CT analysis. *Composites Part A: Applied Science and Manufacturing*, 2018, 113, pp.66 - 75. 10.1016/j.compositesa.2018.07.015 . hal-03026038

**HAL Id: hal-03026038**

**<https://hal.science/hal-03026038>**

Submitted on 26 Nov 2020

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# **Interfacial adhesion assessment in flax/epoxy and in flax/vinylester composites by single yarn fragmentation test: correlation with micro-CT analysis**

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## **Abstract:**

Despite the academic interest in using plant fibres as reinforcement in polymer composites to replace glass fibres, the industrial exploitation of resulting composites in semi- or structural applications is still limited. This is mainly due to the poor adhesion at the plant fibre/polymer matrix interface dictated by their surface chemistry and strong hydrophilic behaviour. In the present work, an assessment of the interfacial adhesion at the yarn scale has been carried out.

Fragmentation tests have been performed on flax/epoxy and flax/vinylester single yarn composites. High-resolution microtomography has allowed a 3-D reconstruction of the breaking area of the flax yarn. The flax/epoxy system has shown the lowest values of critical fragment length and interfacial debonding length, and the highest values of IFSS. For both epoxy and vinylester samples, it was found that the breakage of flax has been mainly concentrated in the peripheral zone of the yarn.

**Keywords:** A. Natural fibres; B. Interface/Interphase; B. Fragmentation; D. CT analysis

## **Introduction**

It is very well-known that the global mechanical properties of composite materials are strongly dependent on the adhesion quality of fibre/matrix interface and this is even more stringent for natural fibre composites due to the inherent hydrophilic nature of plant fibres. As a result, the good specific mechanical properties of the fibres are far from being efficiently exploited, thus leading to an unsatisfactory mechanical performance of the resulting composites that cannot be reliably used in semi-structural applications. Therefore, it is not surprising that a great deal of research efforts has been directed towards the improvement of natural fibre/polymer matrix interface compatibility [1][2]. However, these efforts need to be supplemented with a reliable and quantitative assessment of interfacial adhesion in order to optimize the surface treatments and broaden the applications of natural fibre composites. For the analysis of traditional composites based on synthetic fibres such as glass or carbon, several tests have been developed, including the pull-out test, the microbond test, the push-in test and the single fibre fragmentation test (SFFT) [3][4]. The applicability of these methods has been the subject of extensive debate in literature and some of them have been also extended to the characterisation of the fibre/matrix interactions of lignocellulosic fibre-reinforced polymers [5]–[9]. The results clearly show that when dealing with natural fibre composites, which are highly heterogeneous in nature, additional care and caution should be paid in assessing the interfacial adhesion through single fibre model composites. Single fibre fragmentation test is one of the most widely used tests mainly because of practical advantages related to the simplicity of sample preparation and the possibility of producing many fragments in a single specimen and then, through statistical analysis, a valid distribution can be obtained from a limited number of test samples. Nonetheless, this test method applied to natural fibre composites suffers from several limitations including the need to estimate accurately the fibre strength at the critical fibre fragment length and the significant variation of the fibre diameter not only among fibres but also within the same fibre. It is worth noting that an adequate test configuration should involve a stress distribution similar to that in a real composite and in natural fibre composites the potential of plant fibres can be only explored by using yarns, where short fibres are twisted with an angle to the yarn axis in order to provide axial strength to the yarn [10]. Therefore, the interfacial adhesion should be assessed at

the yarn scale and not only at the technical fibre scale. In this regard, Spārniņš et al. [11][12] proposed a method for IFSS estimation using the pulled-out fibre length distribution of oriented flax fibre reinforced composite incorporating a modified Weibull strength distribution. The authors obtained values of IFSS lower than those determined by fragmentation tests, which were ascribed to a defective impregnation of flax yarns and low mutual adhesion of elementary flax fibres. To partially address some of the mentioned issues, it could be possible to extend the SFFT at the yarn scale, thus embedding one single yarn and eliminating problems related to imperfect impregnation of plant yarns. This approach has been investigated in previous works [13][14], where detailed experimental, numerical and full-field strain measurements by digital image correlation helped in getting insight into the strong heterogeneity of strain fields that develop with the applied stress and in providing a first step towards the assessment of local behaviour. The complexity of such approach stems also from the cross-sectional area of the yarn that typically includes voids between the fibres which are then replaced by matrix during the impregnation of the single yarn. In this framework, the purpose of this work is to investigate further the possibility to use fragmentation test to assess the adhesion of flax yarns to different thermoset resins. In particular, epoxy and vinylester matrices have been selected. A specific metallic mold has been designed to manufacture monofilament composite samples, where the filament is represented by a single flax yarn embedded in the polymer resin. The adhesion quality between flax yarn and both epoxy and vinylester matrices has been assessed in terms of critical fragment length, debonding length and interfacial shear strength. This experimental characterization has been supplemented for the first time, to the best of authors' knowledge, with microcomputed tomography. High-resolution microtomography (1.5  $\mu\text{m}$ ) allowed the measurement of the debonding length between flax yarn and polymer matrices. Using the AVIZO 9.0 software, it was also possible to perform a volumetric reconstruction of the breaking area of the flax yarn.

## **1 – Materials and methods**

### **1.1 Materials**

Individual flax yarns have been extracted by hand from Biotex fabric. The Biotex flax fabric (*Linum usitatissimum*) was supplied by Composites Evolution (UK). It is a 2×2 twill fabric (200 g/m<sup>2</sup>), commercialized without any specific sizing agent. The thermoset resins used for the study were epoxy and vinylester. Concerning the epoxy resin, Prime 27 infusion resin and PRIME 20 slow hardener, both delivered by GURIT, were selected. The curing process was carried out with a recommended mixing ratio of 100:28 by weight. A post-curing procedure of 7 h at 90 °C was optimized to obtain a fully cross-linked epoxy. Regarding the vinylester resin, the Advalite VH-1207 infusion resin, supplied by Reichhold, was chosen. This is a styrene free resin, formulated for use in resin vacuum infusion process. The Norpol Peroxide P MEC N24 catalyst, supplied by Reichhold, was used as curing agent. The curing process was carried out with a mixing ratio of 1 phr. No post-curing treatment was performed for the vinylester resin.

## **1.2 Tensile testing**

A mechanical characterization of each constituent has been carried out by performing tensile tests on single flax yarns and neat epoxy and vinylester resins. Single yarn tensile tests were performed according to ASTM C-1557. Three different gauge lengths were tested, namely 20, 30, and 40 mm. Individual yarns were glued onto card tabs with a central window cut out to match the desired gauge length for the test. Tensile tests were carried out at room temperature with a Zwick/Roell Z010 equipped with a 1 kN load cell. Tests were performed in displacement control with a cross-head speed of 2 mm/min. Before testing, all the yarns were heat treated at 45 °C for 24 h for moisture elimination. Thirty yarns were tested at each gauge length. Neat resin specimens were loaded in uniaxial tension using an Instron E1000 ElectroPuls machine equipped with a load cell of 2 kN. In order to reproduce the same conditions used during the fragmentation tests, a crosshead speed of 0.005 mm/min has been used for the tensile tests. For each resin, a minimum of five tests have been performed.

## **1.3 Single yarn fragmentation tests**

### **1.3.1 Specimen preparation**

A specific metallic mould has been designed to manufacture single yarn composite specimens with the yarn aligned in the loading direction. Some little slots were designed at each end of the negative samples to facilitate the flax yarn placement in the middle of the specimen. The whole surface of the mould was prepared by applying a release agent. Before casting, flax yarns were conditioned at 45 °C for 24 h for moisture elimination. Each flax yarn was positioned in the mould and held in place by adhesive tape. In order to avoid the leakage of the resin from the mould and simultaneously ensure the right quantity of resin to reduce as much as possible the formation of a meniscus due to the shrinkage during the crosslinking process of the resin, the mould has been filled up with resin using a syringe. The metallic mould used for the sample manufacture and the specimen geometry can be seen in figure 1.

### **1.3.2 Test set-up**

The fragmentation testing was performed with an Instron E1000 ElectroPuls test machine with a load cell of 2 kN and using a crosshead speed of 0.005 mm/min. The test was carried out at a rather low loading rate and the loading phase was stopped if the specimen failed, or when the fragmentation saturation level was achieved which, in the present case, was defined as occurring when no new yarn breaks appeared during a subsequent strain increase by 0.5%, as frequently reported in literature (R. Joffe, J. Andersons, and L. Wallström, “Interfacial shear strength of flax fiber/thermoset polymers estimated by fiber fragmentation tests,” *J. Mater. Sci.*, vol. 40, no. 9–10, pp. 2721–2722, 2005). Some epoxy- and vinylester-based specimens were loaded up to failure at strains higher than 9 and 11%, respectively, in order to be sure that the saturation had occurred. The average flax yarn diameter was evaluated by optical microscopy before each test. For each specimen, the yarn diameter has been measured in at least ten points along the gauge length (15 mm).

## **1.4 Optical and FE-SEM observations**

The flax yarn diameters and all the fragment length measurements were carried out by using a ZEISS Axio Imager optical microscope. The morphology of the single yarn composite fracture surfaces was investigated by a field-emission gun scanning electron microscope (JEOL JSM-7000F). All specimens were sputter coated with gold prior to FE-SEM observations.

### **1.5 Micro-CT apparatus**

Image acquisition has been performed using an UltraTom CT scanner manufactured by RX Solutions (France). The system consists in a Hamamatsu open type microfocus X-ray tube operating at 20-100 kV / 0-200  $\mu$ A, within a maximum power of 20 W (Configuration with a Lanthanum hexaboride filament). A precision object manipulator with two translations and one rotation facilitates rotating the sample for acquisition of tomographic data, and displacement along the optical axis to adjust the magnification. X-rays generated by the source diverge at an angle providing a cone-beam, thus samples can be imaged at various geometric magnifications by moving the sample close to the source to provide high resolution mode or close to the detector to provide low resolution measurements. The generator and the detector are also mobile to cover a large range of magnification. A 1.5  $\mu$ m resolution has been used in this work, with an accelerating voltage of 50 kV and a beam current of 157  $\mu$ A. This X-ray detector consists in an X-ray CsI scintillator screen which is settled on an amorphous silicon layer. For 3D reconstruction, X-ray images were acquired from 1120 rotation views over 360° (approximately 0.32° rotation step). The reconstruction was performed using an algorithm based on the filtered back-projection procedure for Feldkamp cone beam geometry. The analysis of the micro-CT pictures has been performed by using the Avizo 9.0 software.

## **2- Results and discussion**

### **2.1 Characterization of composite single constituents**

A mechanical characterization for both types of polymer resin has been done by performing tensile tests. Table 1 summarizes the mechanical results. High failure strain values were found for both epoxy and vinyl ester resins. This behaviour may be explained by the low crosshead speed used

during tensile tests. In particular, vinylester resin displayed a markedly ductile behaviour. Unlike synthetic fibres, one of the main characteristics of the plant ones is their extreme heterogeneity. One of the parameters of greatest variability of this type of reinforcement is surely the diameter. In this study, a statistical analysis based on measurements at a hundred points was performed to define the diameter value of flax yarn. The Gaussian probability distribution of flax yarn diameter is defined by the equation (1) [13]:

$$P(d) = \frac{1}{\sqrt{2\pi} \cdot \sigma} e^{-\frac{(d-\mu)^2}{2 \cdot \sigma^2}} \quad (1)$$

where  $d$  is the diameter value of flax yarn,  $\mu$  is the mean of the distribution, and  $\sigma^2$  is the variance of the distribution. In figure 2 is reported the Laplace-Gauss distribution results for the yarn diameter. A very large diameter distribution was found for this kind of flax, with an average value of 327  $\mu\text{m}$  and a standard deviation of 96  $\mu\text{m}$ . These results confirm the significant scattering in the measured diameter values and were used for evaluating the yarn cross-sectional area assuming a circular cross-section. Table 2 presents the tensile strength of flax yarns evaluated at three different gauge lengths, namely 20 mm, 30 mm and 40 mm. As reported in table 2, the strain to failure value for all the yarns was between 3.4 % and 3.8 %. It can be seen that with longer gauge length there is a small decrease in the yarn strength, from 272 to 236 MPa. The scatter of the tensile strength of flax yarns was statistically analysed using a two-parameter Weibull distribution, according to the equation (2) [15]:

$$Pr(\sigma) = 1 - \exp \left[ - \left( \frac{\sigma}{\sigma_0} \right)^m \right] \quad (2)$$

Where  $Pr(\sigma)$  is the cumulative probability of failure,  $m$  is the Weibull modulus (related to the dispersion of the data) and  $\sigma_0$  is a scale parameter (characteristic strength). The probability of failure was estimated by the estimator  $F$  reported in equation (3):

$$F = \frac{i-0.5}{N} \quad (3)$$

where  $N$  is the number of filaments tested and  $i$  is the rank of data point for each filament. The graphs in figure 3 represent the results of the two-parameter Weibull distribution applied to all the gauge lengths. The Weibull modulus and the characteristic life values found for the flax yarns are reported in table 2. A comparison between the stress-strain curves of flax yarn and polymer resins may be observed in figure 4. It can be seen that flax yarn is characterized by a clearly higher tensile strength in comparison to both epoxy and vinylester resins, and a far smaller strain at failure. These differences in mechanical behaviour between the flax yarn and the polymer matrices are essential for carrying out the fragmentation tests. As reported by Zafeiropoulos [9], only in monofilament composites characterized by a very ductile matrices and a brittle filament, it is possible to reach a significant strain higher than the strain to failure of the filaments that ensures a full debonding/yielding of the filament fragments. This will provide a constant shear at the filament/matrix interface and so a full satisfaction of the Kelly-Tyson requirements.

## 2.2 – Determination of critical fragment length

According to the Kelly and Tyson model [16], if a long filament is embedded in a resin matrix and the resultant system is elongated, the stress transferable to the filament at a distance  $x$  from its end may be evaluated by the equation (4):

$$\sigma_x = \left(\frac{4\tau}{d}\right) \cdot x \quad (4)$$

where  $d$  is the diameter of the filament and  $\tau$  the shear strength at the filament/matrix interface.

This equation comes from a simple force balance at filament/matrix interface and it is valid only under the condition of assuming a constant shear stress  $\tau$  at the interface. When the monofilament composite is elongated further, the tensile stress transferable to the filament increases until it reaches the ultimate tensile strength of the filament, equation (5):

$$\sigma_f = \left(\frac{4\tau}{d}\right) \cdot x_0 \quad (5)$$

where  $x_0$  is the value of the distance  $x$  from the end of the filament at which the tensile stress reaches the ultimate tensile strength of the filament. If a broken piece of filament exceeds  $2x_0$  in length, the breakage process repeats itself. On the contrary, if the broken piece is characterized by a length shorter than  $2x_0$ , the tensile stress transferable to the filament is not able to reach the ultimate tensile strength of the filament and hence no further fragmentation occurs. As reported by Ohsawa et al. [17], the length of filament fragments ( $l$ ) follows the uniform distribution characterized by a range  $x_0 \leq l \leq 2x_0$ , and the average value of the fragment length is given by the equation (6):

$$\bar{l} = \frac{1}{2}(x_0 + 2x_0) = \frac{3}{2}x_0 \quad (6)$$

Since  $x_0$  is the length needed for the stress on filament to reach the ultimate tensile strength, the critical filament length  $l_c$  is equal to  $2x_0$ . By introducing this relationship into eq. (6), it is possible to evaluate the critical filament length from equation (7):

$$l_c = \frac{4}{3} \cdot \bar{l} \quad (7)$$

In figure 5 is reported an example of a typical optical micrograph obtained for the VF\_3 fragmented flax yarn. The yarn diameter and fragment lengths were measured in the gauge length region of the specimen. The fragment lengths have been determined as the average values between the internal and the central points of the breaking zones (reported in dotted red lines). Moreover, around each yarn break, the length of the black zone (debonding length) observed on micrographs (figure 5) has also been measured (reported in blue line). As it will be demonstrated by the micro-CT analysis in section 2.5, this black zone corresponds to the debonding length. Therefore, the black zone length values are noted as  $l_{debonding}$  in figure 5 and table 3. Regarding the fragmentation tests, the two thermoset systems exhibited different results. All the critical length measurements are reported in table 3. A larger number of fragments was found for flax yarn embedded in epoxy matrix than in the case of vinylester matrix. Flax yarn has suffered a greater number of cracks with critical fragment length value lower than that found in the case of vinylester resin. The critical filament length plays an essential role in determining the adhesion quality in a composite. In fact,

the measurement of the critical filament length is directly correlated to the shear strength value transferred at filament/matrix interface. As reported by Zafeiropoulos [9][18], a very interesting approach to assess the interface properties in a composite material is the model proposed by Fraser et al. [19]. In this approach, the adhesion quality is not estimated from the measurement of the interfacial shear stress but rather by the fragment length values. The principle of the approach introduced by Fraser and co-authors is that, at saturation level, the critical filament fragment length reflects the stress transfer efficiency at the interface and thus it offers a qualitative value of the interfacial bond. However, it must be emphasized that, with this method, it is possible to compare the adhesion properties of different filament/matrix systems through the comparison of the filament fragment length values only in the case that the filament strength values are the same for all the systems. In this study, both epoxy and vinylester composites have been realized using the same type of flax yarn. This allows us to use the approach proposed by Fraser and co-authors to determine the differences in interfacial bond in terms of critical yarn fragment length. From the results reported in table 3, it is possible to see that the flax/epoxy system is characterized by smaller values of critical length. This indicates a better adhesion quality between flax yarn and epoxy matrix contrary to what has been observed for the vinylester matrix. As previously reported, flax yarn is a highly heterogeneous material, especially for its diameters values (figure 2). For this reason, the  $l_c/d$  ratio has been calculated in order to compare normalized values (see in Table 3). Also in this case the flax/epoxy system displays the best results in terms of interfacial adhesion.

### **2.3 – Calculation of interfacial shear strength**

The filament/matrix interface adhesion quality is usually described by determining the interfacial shear strength value (IFSS). According to Kelly and Tyson [16], the IFSS may be estimated using equation (8):

$$IFSS = \frac{\sigma_f(l_c) \cdot d}{2 \cdot l_c} \quad (8)$$

where  $d$  is the filament diameter,  $l_c$  is the critical fragment length and  $\sigma_r(l_c)$  is the filament strength at a length equal to the critical filament length. Because of the small  $l_c$  values, it is impossible to measure experimentally the tensile strength of a single filament at a length equal to the critical one. To overcome this problem, an extrapolation of strength at critical length  $l_c$  was realized. In this study, two different ways of extrapolation have been taken into consideration. The first method has been proposed by Asloun et al. [20]. According to this study, it is possible to extrapolate the maximum stress of flax yarn at small gauge lengths from tensile tests performed at several large gauge lengths. It is possible to do this extrapolation by taking the logarithm of stress in equation (9):

$$\ln(\sigma_f) = -\frac{1}{m_i} \ln(L_0) + \ln[\sigma_0^i \Gamma(1 + \frac{1}{m_i})] \quad (9)$$

where  $m_i$  and  $\sigma_0^i$  are the shape and the scale parameters of the equivalent Weibull distribution respectively,  $L_0$  the gauge length,  $\sigma_f$  the tensile strength and  $\Gamma$  the Gamma function. Thus, the tensile strength of a flax yarn at a given gauge length can be easily estimated by linear extrapolation using equation (9). As it is possible to see in figure 6, the graph plotting  $\ln(\sigma_f)$  against  $\ln(L_0)$  is linear with a slope of  $-\frac{1}{m_i} = -0.2$ . The second extrapolation method that has been taken into account in this study is based on the Weibull cumulative distribution function determined for only one given filament gauge length  $L_0$ . This method is widely used in literature [9], [13], [21]–[23]. Using the obtained two-parameter Weibull distribution, the average filament strength may be evaluated by equation (10):

$$\langle \sigma_f \rangle = \sigma_0 L_0^{-\frac{1}{m}} \left[ \Gamma\left(1 + \frac{1}{m}\right) \right] \quad (10)$$

where  $\Gamma$  is the Gamma function and  $\sigma_0$  and  $m$  are the characteristic life and the shape parameter of Weibull distribution for the tested gauge length  $L_0$ , respectively. In this study, the chosen value for  $L_0$  is 40 mm. Therefore, the used Weibull parameters for this second method are those obtained

with tensile tests on flax yarns of 40 mm length (Table 2). By applying equation (10) to  $L_0$  and to the critical length  $l_c$ , it is possible to obtain the following equation (11):

$$\sigma_f(l_c) = \sigma_f(L_0) \left( \frac{L_0}{l_c} \right)^{\frac{1}{m}}$$

(11)

For comparison purposes, in this study both the linear extrapolation method with equation (9) and the Weibull weakest link theory with equation (11) have been used to determine the yarn strength at a length equal to the critical yarn length,  $\sigma_f(l_c)$ , and so the IFSS values. In table 4 are reported the results obtained with both methods. Comparing the results of the two methods, a negligible difference has been found for the flax yarn  $\sigma_f(l_c)$  values. These results prove the validity of both calculation methods. Concerning the analysis of the yarn/matrix adhesion quality, from table 4 it is possible to see that the IFSS values reflect the results already found by comparing the critical fragment lengths. In fact, from the IFSS values, it is possible again to conclude that the flax yarn/epoxy system is characterized by a better interface quality than the flax/vinylester one.

#### **2.4 - Fractographic analysis**

After fragmentation tests, a morphological investigation of the fracture surface has been performed using the FE-SEM. The fracture surface of both single flax yarn samples with epoxy and vinylester resins was investigated. From the direct analysis of the fracture surfaces (figure 7), it can be concluded that the yarn failure is concentrated mainly in the peripheral zone of the yarn. This phenomenon is particularly evident for the flax/epoxy samples, figure 7-A. Concerning the vinylester resin, a partial tearing of the yarn, occurring at the moment of failure was found for most of the samples. This different behavior can be considered as a further confirmation of the lower adhesion quality between the flax yarn and the vinylester matrix. At a smaller scale, in figure 8, it is possible to see that there are differences between the two systems at the fibre/matrix interface. Indeed, for the flax/epoxy sample the presence of little resin ligaments, albeit minimal, was found between the matrix and the flax fibres. It has not been observed for the flax/vinylester composite.

These little connections may explain the better adhesion that the flax yarn displayed in presence of the epoxy resin.

## **2.5 – Fragment debonding analysis**

An in-depth analysis of the fracture zone distribution along the flax yarn was also carried out. A post-mortem examination of the monofilament composites has been performed through the use of micro computed tomography. It is important to point out the great novelty of the use of this type of analysis for the assessment of the interfacial properties in monofilament composites. For both EF\_1 (flax/epoxy) and VF\_3 (flax/vinylester) specimens, microtomographic image acquisitions have been made with a resolution of 1.5  $\mu\text{m}$ . Using the AVIZO 9.0 software, it was possible to perform a 3-D reconstruction of the breaking area of the flax yarn (figure 9). Raw images were pre-treated using smoothing filters and an image segmentation was carried out. Black voids (breaking zone) were identified from the background (the rest of flax yarn) on the basis of the different grey threshold values. The analysis carried out on both types of samples has shown that the breakage of flax is mainly concentrated in the peripheral zone of the yarn. This result is clearly visible in the 9-E and 9-F images that show the 3-D reconstruction of the breaking zone, and it is totally in accordance with what was observed by FE-SEM analysis on fracture surfaces, figures 7-A and 7-B. The 3D micro-CT images also allow the measurement of the debonding length between the flax yarn and the polymer matrix. Indeed, the debonding zone is detectable by the black voxels (corresponding to voids) between the periphery of the yarn and the matrix. This measurement was possible through the high resolution micro-CT observations (voxel size of 1.5  $\mu\text{m}$ ). In figure 10 are reported two examples of x-slices obtained for both flax/epoxy and flax/vinylester single yarn samples. Debonding length values of  $530\mu\text{m} \pm 70\mu\text{m}$  and  $624\mu\text{m} \pm 76\mu\text{m}$  were found for the flax/epoxy (EF\_1) and the flax/vinylester (VF\_3) monofilament composites, respectively. These values of the debonding lengths are consistent with the black zone lengths measured by optical microscopy on the same samples:  $504\mu\text{m} \pm 102\mu\text{m}$  for EF\_1 and  $599\mu\text{m} \pm 181\mu\text{m}$  for VF\_3 (Table 3). This demonstrates that the observed black zone on micrographs coincides with the debonding

zone between the flax yarn and the polymer matrix. According to Kim et al. [24] and Ramirez et al. [21], debonding represents a very important phenomenon in studying the interface quality of polymer composites. These authors have performed fragmentation tests on single carbon and glass fibre composites. They measured the debonding length around each fragment by using photoelastic birefringence patterns. As explained by Kim and Nairn [24], the interfacial zone between the filament and the polymer matrix may be considered as a set of bonding lines. When the monofilament composite is loaded, the molecular bonded lines become tense until filament fractures and some lines near the filament break are disconnected. They defined the debonding zone as the region containing both the filament breaking gap and the breaking zone of the molecular bonding lines. They showed that a large value of the debonding zone is typical for weak interfacial bonds. Thus, the higher debonding length value found for the system constituted by flax yarn and vinylester matrix displays, once again, the lower adhesion quality of flax yarn with this type of polymer matrix. This result reflects perfectly the calculated IFSS values. A comparison between the debonding length measurements, performed by optical microscopy and micro-CT, and the inverse of the IFSS values obtained for both EF\_1 and VF\_3 samples has been carried out and reported in figure 11. As it is possible to see in table 3 and in table 4, the IFSS values are directly linked to the debonding length between the flax yarn and the polymer matrix, and thus to the adhesion quality at the yarn/matrix interface. The greater the adhesion interfacial properties and the IFSS values, the lower the debonding lengths. It is also worth noting that micro-CT is able to provide a more accurate measurement of the debonding length with a reduced dispersion of data.

### **3 - Conclusions**

This study has been focused on the characterization of the adhesion properties between flax yarns and polymer (epoxy and vinylester) matrices. Experimental investigation was performed on single flax yarn composite specimens. Fragmentation tests allowed the determination of the interfacial shear strength at yarn/matrix interface. The adhesion quality between flax yarn and both epoxy and vinylester matrices has been assessed in terms of critical fragment length, interfacial shear strength

(IFSS) and debonding length. A link between these parameters was demonstrated. The best adhesion properties have been obtained for the flax/epoxy system. In fact, this composite is characterized by the lowest values of critical and debonding lengths and the highest values of IFSS. For the first time, micro computed tomography has been used as analysis technique in the study of the interfacial properties after performing fragmentation tests. High-resolution microtomography allowed to measure the debonding lengths and to correlate it with the black zone lengths observed by optical microscopy. Using the AVIZO 9.0 software, it was possible to perform a 3-D reconstruction of the breaking area of the flax yarn. For both epoxy and vinylester samples, it was found that the breakage of flax has been mainly concentrated in the peripheral zone of the yarn, which confirms that fragmentation tests on single yarn composites allow the analysis of the interface adhesion quality between yarn and matrix in view of the optimization of woven composite materials.

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## Figure Captions

**Figure 1** (A) Metallic mould used for sample manufacture: the resin casting stage using a syringe; (B) Specimen geometry for the single yarn fragmentation tests.

**Figure 2** Gaussian distribution of measured diameters of flax yarn.

**Figure 3** Cumulative distributions of the failure probability values measured for three different gauge lengths (20, 30 and 40 mm) for flax yarn.

**Figure 4** Stress-strain curves for flax yarn (gauge length of 40 mm), epoxy and vinylester resins obtained at 0.005mm/min.

**Figure 5** Typical optical micrograph of a fragmented flax yarn. In particular this image shows the yarn diameter  $d$  (green colour), the fragment length  $l$  (red colour) and the debonding length  $l_{\text{debonding}}$  (blue colour) measurements for the VF\_3 sample.

**Figure 6** Evolution of tensile strength of flax yarn versus gauge length.

**Figure 7** FE-SEM-micrographs showing the fracture surface for flax/epoxy (A) and flax/vinylester (B) single yarn composites.

**Figure 8** FE-SEM-micrographs showing the different interfacial morphology of flax/epoxy (A) and flax/vinylester (B) single yarn composites after testing.

**Figure 9** Volumetric reconstruction of the flax yarn and the fracture zone for both flax/epoxy (A-C-E) and flax/vinylester (B-D-F) systems. The flax yarn is represented in yellow, the fracture zone in blue and red respectively for flax/epoxy and flax/vinylester samples.

**Figure 10** High resolution micro-CT images for EF\_1 flax/epoxy (A) and VF\_3 flax/vinylester (B) single yarn samples. The red dotted lines show the debonding length measurements obtained by micro computed tomography.

## Tables

**Table 1** Tensile properties of epoxy and vinylester resins obtained at 0.005mm/min

	$\sigma_f$ [MPa]	$\epsilon_f$ [%]
<i>Epoxy Resin</i>	58.19 ± 0.75	8.29 ± 0.15
<i>Vinylester Resin</i>	34.99 ± 0.47	21.21 ± 2.89

**Table 2** Tensile properties of flax yarns as a function of gauge length and the corresponding Weibull parameters. Tests have been carried out at 2 mm/min.

	Gauge length $L_0$ [mm]	$\sigma_f$ [MPa]	$\epsilon_f$ [%]	Characteristic Strength $\sigma_0$ [MPa]	Weibull Modulus $m$
<i>Flax yarn</i>	20	272 ± 46	3.78 ± 0.57	292	6.83
	30	248 ± 46	3.62 ± 0.62	265	6.36
	40	236 ± 57	3.40 ± 0.42	257	5.02

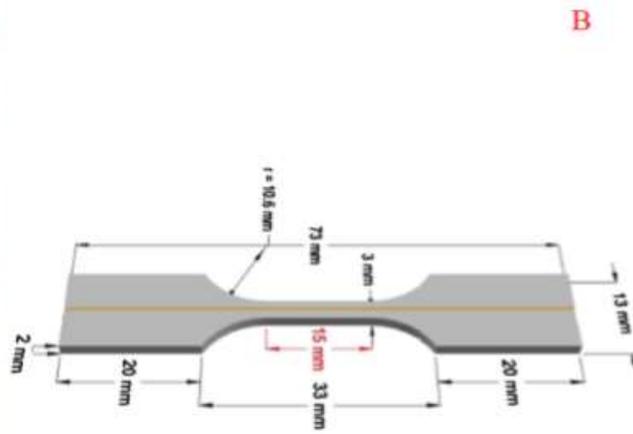
**Table 3** Fragment length values for flax/epoxy and flax/vinylester single yarn composites.

<i>Flax/Epoxy Resin</i>	Yarn diameter $d$ [ $\mu\text{m}$ ]	Number of fragments	Average $l$ [ $\mu\text{m}$ ]	$l_0/d$	$l_{\text{debonding}}$ [ $\mu\text{m}$ ]	$l_{\text{debonding}}/d$
EF_1	210 ± 35	8	1924 ± 639	12.18	504 ± 102	2.39
EF_2	222 ± 32	8	1473 ± 532	8.83	356 ± 155	1.61
EF_3	253 ± 43	7	2345 ± 372	12.35	460 ± 96	1.82
EF_4	247 ± 21	8	1847 ± 486	9.95	465 ± 9	1.88
EF_5	308 ± 25	5	2786 ± 508	12.04	446 ± 128	1.45

<b>EF_6</b>	250 ± 18	8	1717 ± 419	9.15	432 ± 137	1.73
<i>Mean Value</i>			<b>2015 ± 473</b>	<b>10.75 ± 1.61</b>	<b>444 ± 49</b>	<b>1.81 ± 0.32</b>
<i>Flax/Vinylester Resin</i>						
<b>VF_1</b>	238 ± 22	5	2340 ± 799	13.09	1466 ± 294	2.21
<b>VF_2</b>	281 ± 15	4	3560 ± 519	16.84	959 ± 252	2.39
<b>VF_3</b>	271 ± 33	6	2521 ± 807	12.37	599 ± 181	1.92
<b>VF_4</b>	316 ± 23	5	3132 ± 1140	13.21	755 ± 181	2.03
<b>VF_5</b>	336 ± 39	4	3737 ± 1215	14.78	648 ± 156	6.15
<b>VF_6</b>	273 ± 19	6	2447 ± 691	11.95	555 ± 301	3.40
<i>Mean Value</i>			<b>2956 ± 605</b>	<b>13.7 ± 1.81</b>	<b>830 ± 343</b>	<b>3.02 ± 1.62</b>

**Table 4** Interfacial shear strength values for flax/epoxy and flax/vinylester single yarn composites: 1- linear extrapolation method; 2- two-parameter Weibull theory.

	<b>l<sub>c</sub>/d mean</b>	<b>σ<sub>r</sub>(l<sub>c</sub>) 1 [MPa]</b>	<b>σ<sub>r</sub>(l<sub>c</sub>) 2 [MPa]</b>	<b>IFSS 1 (MPa)</b>	<b>IFSS 2 (MPa)</b>
<i>Flax/Epoxy Resin</i>	10.75 ± 1.61	416 ± 19	406 ± 18	19.84 ± 3.88	19.39 ± 3.76
<i>Flax/Vinylester Resin</i>	13.7 ± 1.81	383 ± 16	376 ± 15	14.18 ± 2.93	13.91 ± 2.85



B

Fig. 1. (A) Metallic mould used for sample manufacture: the resin casting stage using a syringe; (B) Specimen geometry for the single yarn fragmentation tests. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

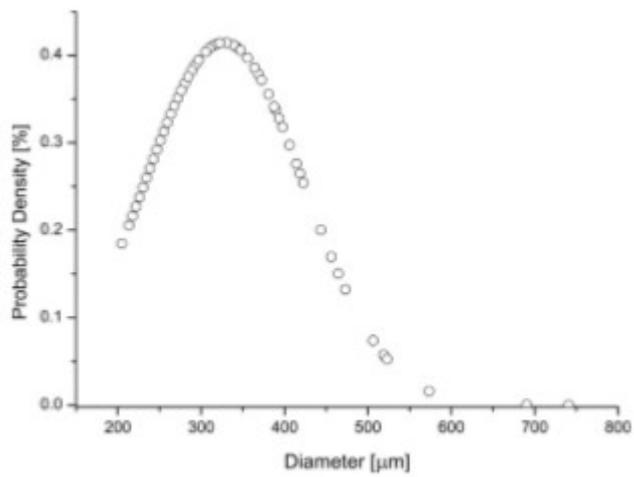


Fig. 2. Gaussian distribution of measured diameters of flax yarn.

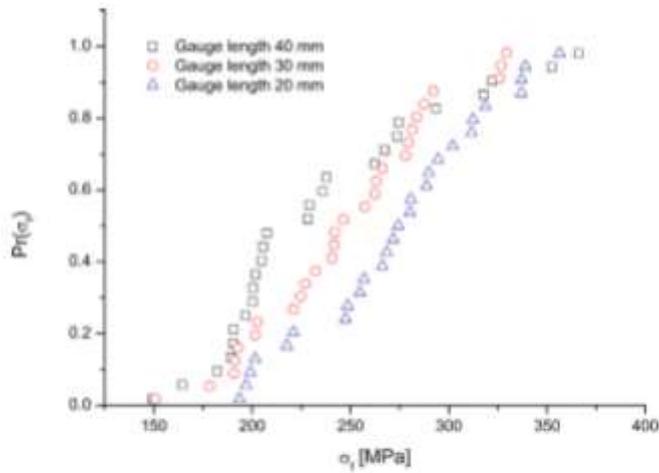


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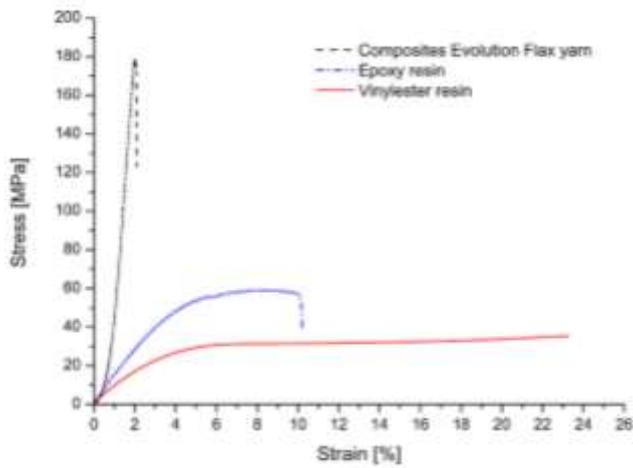


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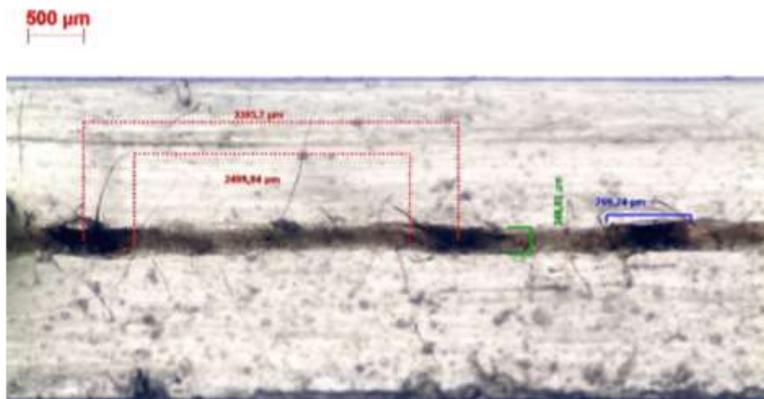
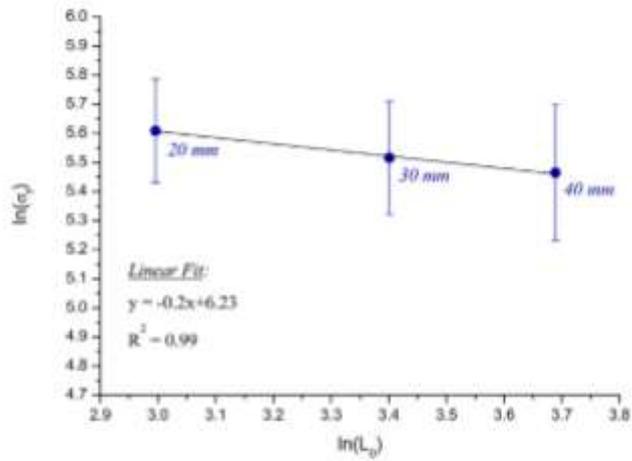
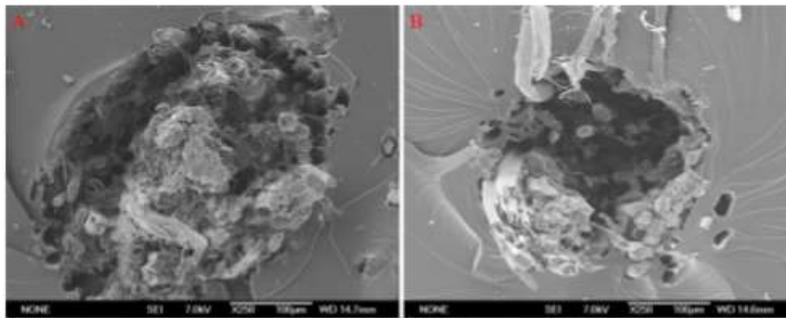


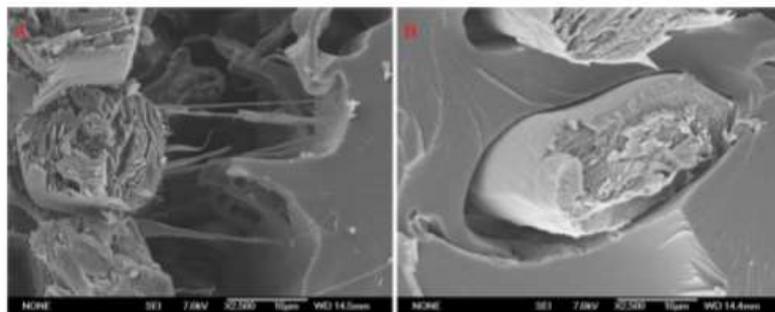
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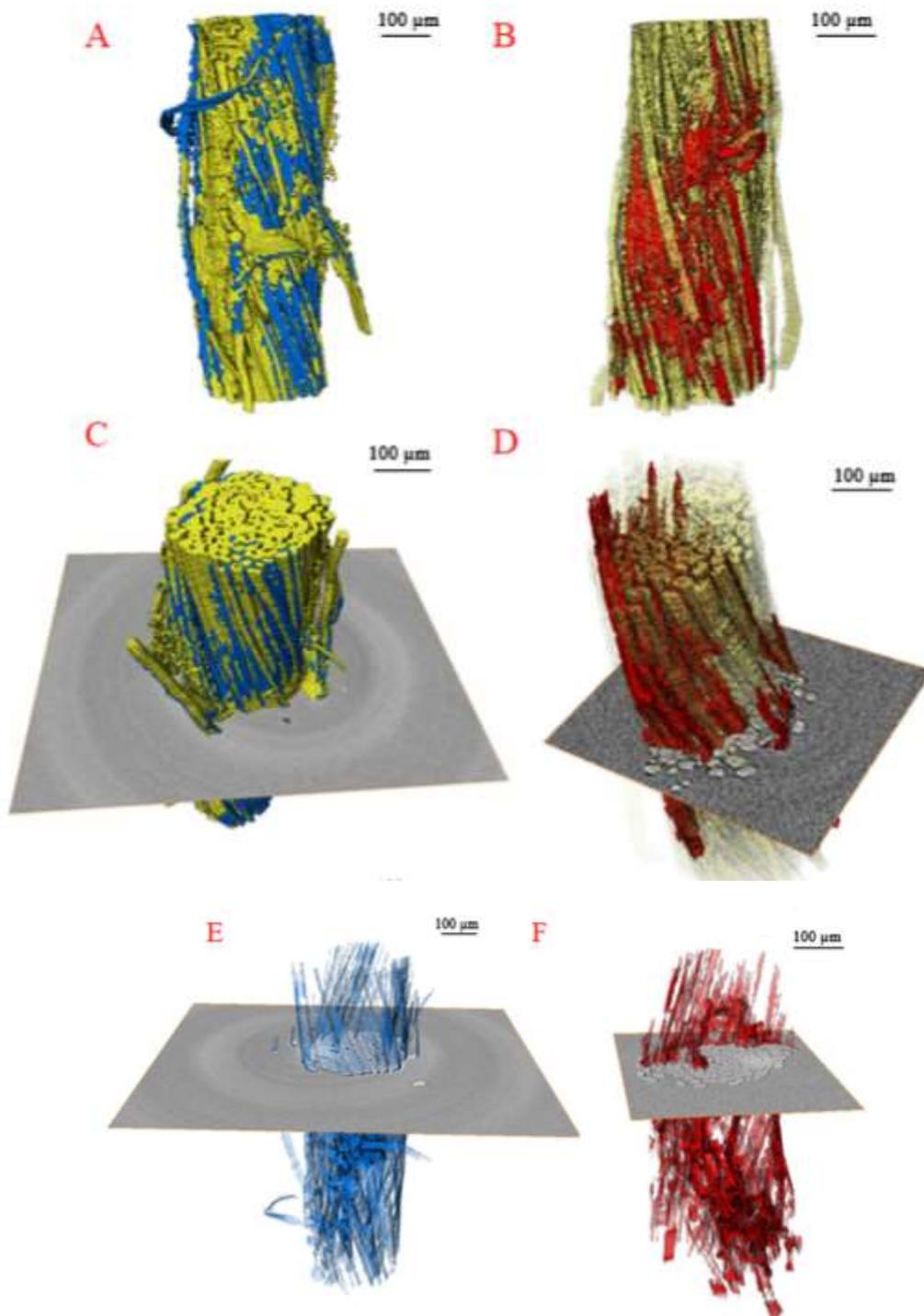
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**Fig. 7.** FE-SEM-micrographs showing the fracture surface for flax/epoxy (A) and flax/vinylester (B) single yarn composites. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)



**Fig. 8.** FE-SEM-micrographs showing the different interfacial morphology of flax/epoxy (A) and flax/vinylester (B) single yarn composites after testing. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)



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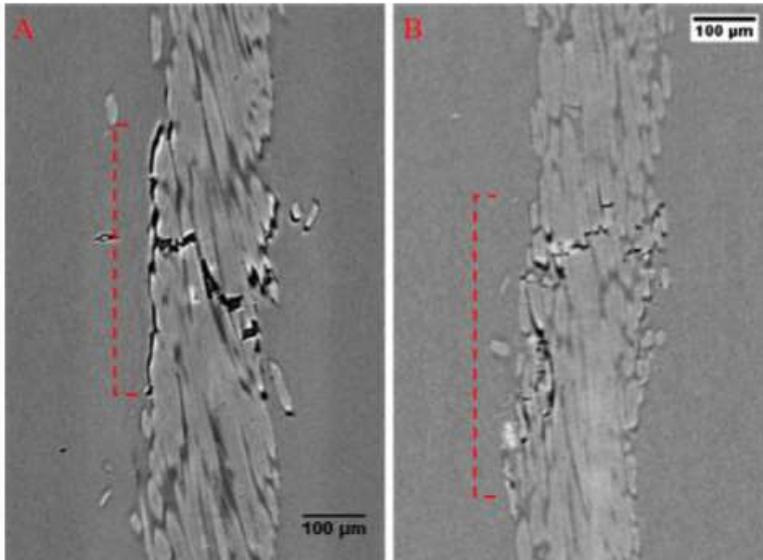


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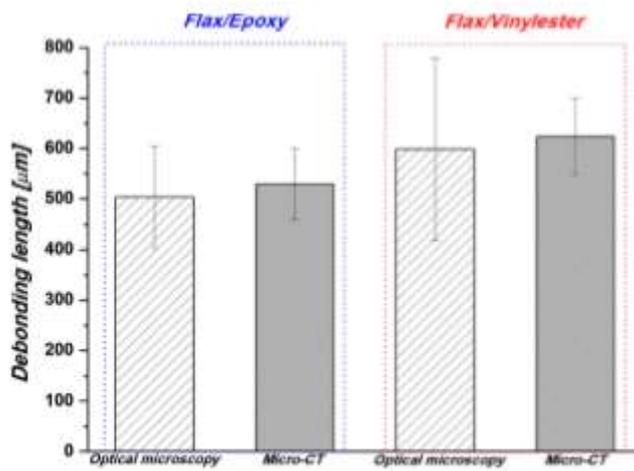


Fig. 11. Bar graph representing the comparison between the different debonding lengths, evaluated by micro-CT and optical microscopy, for both flax/epoxy and flax/vinylester single yarn samples. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)