24th ABCM International Congress of Mechanical Engineering
December 3-8, 2017, Curitiba, PR, Brazil

COBEM-2017-0444
EVALUATION OF INTERFACIAL FIBRE/MATRIX STRENGTH IN NATURAL HYBRID COMPOSITES

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Abstract. *Researches related to the use of Natural Fibre Reinforced Composites (NFRC) have grown considerably in recent years, due to the current society's need to create production methods that require lower energy levels, consume products less aggressive to the environment and preferably biodegradable. Thus, numerous studies were carried out on the influence of fibre / matrix adhesion on the mechanical properties of NFRC in different types of thermoset and thermoplastic matrices. However, the researches related to natural hybrid composites are still little explored. The purpose of this study is to compare the influence of the superficial treatments of mercerization and silanization on the interfacial shear bond strength of jute, sisal and rami fibres in polymer matrices used in the production of hybrid composites (i.e. jute + sisal, jute + rami and jute + fibreglass). Pullout and short-beam tests were performed in order to evaluate the resistance to the real and apparent interfacial shear, respectively. Finally, the fracture surfaces were analysed by optical microscopy. The results of the Pullout tests presented great dispersion and the failure occurred in the fibre and not in the fibre / matrix interface. However, it was possible to determine the influence of surface chemical treatments on fibre strength. From the short beam test, the behaviour of the natural hybrid compound was evaluated for crack propagation and fibre delamination until failure. Both tests presented similar results that demonstrated the efficiency of superficial chemical treatments in improving interfacial adhesion and fibre strength, reducing the presence of voids and consequently increasing the mechanical properties of the natural hybrid compounds. In particular, the composite that presented the best result was jute + ramie silanized, followed by jute + silanized sisal while the hybrid composite jute + fibreglass presented the worst results.*

Keywords: *Natural hybrid composites, superficial treatments, Pullout test, Short beam test*.

1. INTRODUCTION

The development of biodegradable and recyclable materials has increased significantly in recent decades due to global initiatives to reduce environmental impacts and, consequently, to reduce global warming. In this way, several researches and industrial projects have emerged in order to address such directives, such as the use of natural raw materials and the development of lighter automobile and aircraft structures in order to reduce fuel consumption. In this context, research on the use and production of composites with reinforcing fibres of natural origin has increased (Martins *et. al*., 2014; Araújo, 2003; Gurunathan *et. al.*, 2015).

Natural fibre composites when compared to synthetic fibre composites have the main advantages of low cost, lower density and energy consumption required for their processing, resulting in a reduced environmental impact. However, they have as their main disadvantage the low interfacial adhesion between matrix and fibre due to the hydrophobic nature of most polymer matrices whereas the natural fibres are hydrophilic (Gurunathan *et. al.*, 2015; Pavlidou *et. al.*, 2013; Nunna *et. al.*, 2012; Fragassa, 2016). In order to overcome these disadvantages, some authors have proposed the use of superficial chemical treatments in natural fibres (i.e. mercerization, silanization and acetylation) as a solution to improve the interfacial adhesion and consequently increase the mechanical properties of natural fibre composites (Valenza, 2015). The production of hybrid composites with the aim of improving the composite strength by mixing two or more different types of reinforcing fibres, whether natural + synthetic or natural + different natural fibres was also proposed (Pickering, *et. al.*, 2016; Yusoff *et. al.*, 2016; Ornaghi *et. al.*, 2011). Such hybrid composites can be produced in an inter-laminar or intralaminar manner (Fig. 1), the first being easily produced and widely studied.

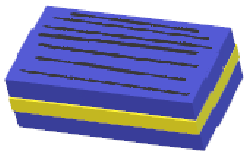
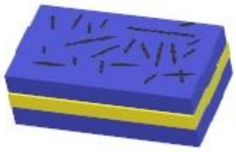
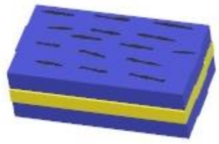
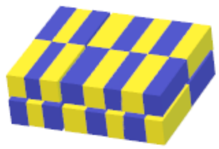
	Continuous fibres	**Discontinuous fibres**	
		Random	**Aligned**
Interlaminated			
Intralaminated			

Figure 1: Configurations for continuous and discontinuous fibre reinforced in hybrid composites.

Several researchers (Mittal *et. al.*, 2016; da Silva *et. al.* 2013) demonstrated the relation between a good interfacial fibre / matrix adhesion and its influence on the increase of the mechanical properties of composite materials. The Pullout, Micro bond and Fragmentation tests are used to evaluate the interfacial fibre / matrix shear strength and the maximum interfacial shear stress, while evaluations regarding the delamination of the composite materials and their behaviour regarding the propagation of cracks can be performed through short beam tests (Tauber *et. al.*, 2013; Almeida *et. al.*, 2012).

The main objective of this work was to evaluate the influence of the use of superficial chemical treatments on natural fibres with the purpose of improving their adhesion to the matrix. Further, the effects of these treatments on the properties of hybrid composites of continuous fibres (Jute + Sisal, Jute + Rami and Jute + Fibreglass) was investigated using pullout and short beam tests. Finally, the strength of the new hybrid composites was compared with the strength of the interlaminar hybrid composites and of composites with only one type of reinforcing fibre.

2. EXPERIMENTAL PROCEDURE

The materials used in the present work were jute fibres, sisal and rami (supplied by Sisalsul, São Paulo - Brazil), fibreglass and epoxy resin - AR260 / AH260 (supplied by Barracuda Advanced Composites, Rio de Janeiro - Brazil) – in the fraction of 100:26 of resin and hardener, respectively.

The chosen jute fibre is a commercial two-way fabric, easily found on the market. The fibres of rami, glass and sisal are presented in the wire configuration. Thus, it was possible to sew these fibres in the void spaces of the jute fabric, in this way creating an intralaminar natural hybrid fabric, as can be seen in Fig. 2. These hybrid fabrics present 60% of jute and 40% of the others types of fibres (i.e. fibreglass, ramie and sisal).

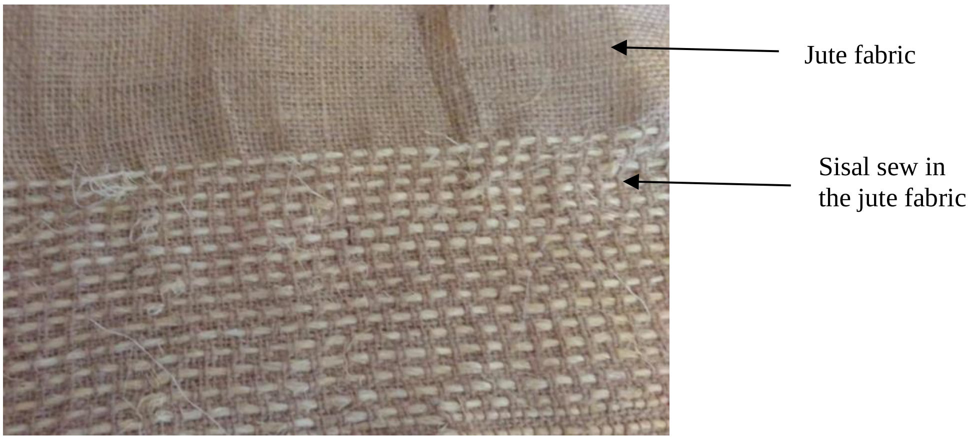


Figure 2: Intralaminar natural hybrid fabric

First, each kind of fibre and the bi-directional hybrid fabrics of jute + sisal, jute + rami and jute + fibreglass were washed and passed through surface treatments:

- Mercerization: The fibres were immersed in alkaline solution (water and NaOH) for 2 hours and with 2% of concentration;
- Silanization: Fibres impregnated directly by manual shaking with silane 2% (Xiameter OFS 6040 – pH = 5.0) in proportion of 1/3 of silane / fibre.

Afterwards the fibres were oven dried at 100° C for 120 minutes. Finally, the moulding process of the specimens was performed by immersing different lengths of embedding the fibres into the mould, soon after the mould was closed with a Teflon tape and its wells were filled with the epoxy resin. As a final point, the mould is heated at 80°C for 6 hours to ensure complete curing of the resin

The samples for the pullout test were manufactured using a metal mould (Fig. 3) and a procedure described in a previous work (Lima *et. al.* 2016), capable of guaranteeing a good reproducibility of the specimens and a good surface finish in the resin blocks, in which the fibres are inserted. The depth of resin cubes is equivalent to 10mm and length of embedding of the fibre varying from 0.5mm to 5mm, as Fig. 4.

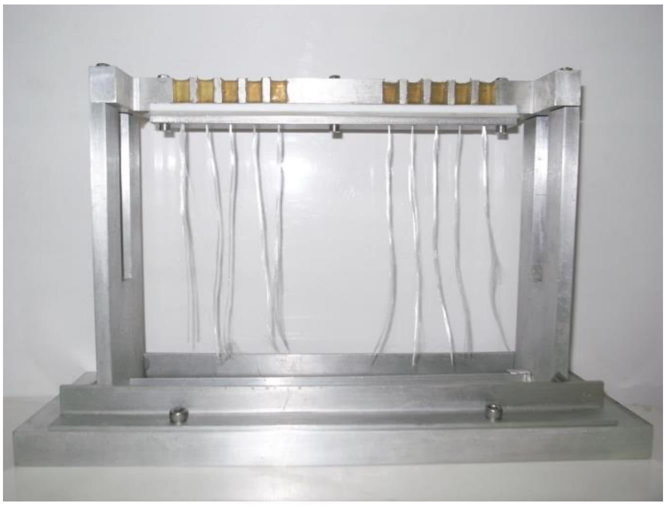


Figure 3: Mould for production of pullout test samples

Figure 4: Pullout test samples

The hybrid natural composites for the short-beam test were fabricated by a hand lay-up process and the curing was done in a hydraulic press at a pressure of 120 kgf/cm² and 80°C in 6 hours. The total volumetric fraction of reinforcement fibres used in the fabrication of the hybrid composites was equal of 30%.

After that, ten specimens of each type of composite, in the conditions without treatment, mercerized and silanized, were prepared in the dimensions of 40 x 10mm and 2mm thickness, according to the dimensions recommended in ASTM D2344 (ASTM, 2000).

For the test a ratio span of thickness of 4:1 was used as recommended by the standard to ensure delamination failure.

The tests were performed in an Instron 5966 (hydraulic, simple multi-axis machine – Instron, Paraná - Brazil) with an advance speed equivalent to 1mm / min (see Fig. 5a). Load-displacement curves were recorded. Subsequently, with the curve of maximum load versus displacement was possible to determinate the interlaminar shear stress (τ – MPa) and the flexural strength (σ – MPa) of the final composite through Eq. (1) and Eq. (2).

$$\tau = \frac{3 * P}{4 * w * t} \quad (1)$$

$$\sigma = \frac{3 * P * s}{2 * w * t^2} \tag{2}$$

Where *P* is the maximum load applied in the test, *w* the width, *t* the thickness and *s* the span of the machine.

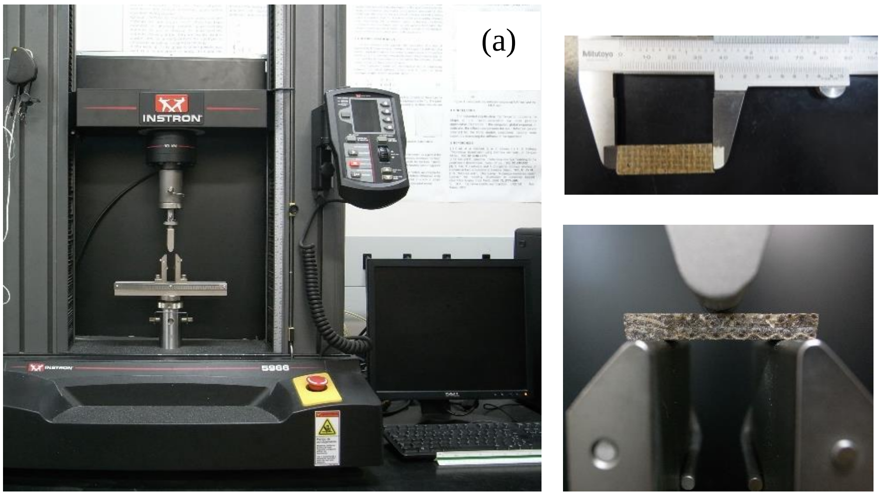


Figure 5: a) Instron machine; b) size of the specimens; c) test detail

3. RESULTS AND DISCUSSION

3.1 Pullout test

After the treatment of the fibres, the first test performed was the pullout test with the objective to study the influence of the superficial chemical treatments in the adhesion between fibre and matrix and consequently the effects on the mechanical resistance of the fibres themselves and the hybrid composites produced.

About 20 specimens of the following types were produced:

- Fibreglass (without treatment);
- Jute (without treatment, mercerized and silanized) ;
- Sisal (without treatment, mercerized and silanized);
- Rami (without treatment, mercerized and silanized).

However, during the tests all failures occurred in the fibre section and not at the interface as intended. It was noticed that after the test that all specimens had an increase of 5mm in their length of embedding, and this might have influenced the results, according to Fig. 6.

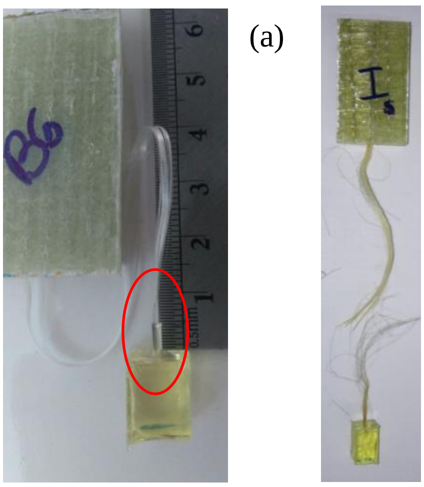


Figure 6: a) extra length of resin; b) fibre's failure

However, from the results of the pullout test it was possible to determine the influence of the surface chemical treatments on the strength of the fibres, as shown in Fig.7.

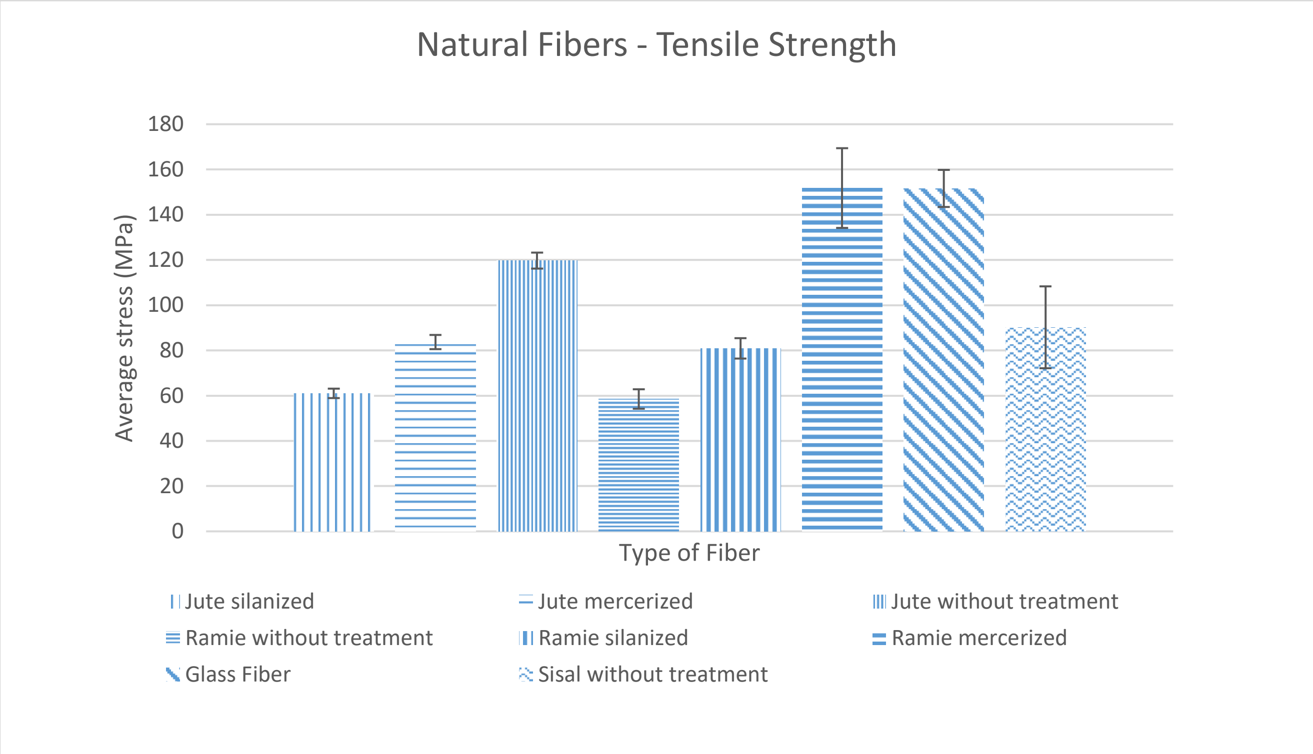


Figure 7: Tensile strength (MPa)

As can be seen from Fig. 7, the different types of surface treatments do not act in the same way in natural fibres. This is due to differences in the morphology of the fibres. The alkaline rami fibre showed excellent results, its tensile strength increased by approximately 60%, reaching resistance values very close to that of glass fibre, for example. While the jute fibre presented better results for the untreated condition, signalling that the treatments were very aggressive to its surface reducing its resistance.

Other relevant information to be drawn from the pullout test is the minimum or critical length of the fibres in the polymer matrix to ensure complete fibre extraction. The greater the critical length the smaller the adhesion between the fibre and the matrix. One of the ways of determining this value is to plot the fracture strength of the fibre by the interfacial shear stress as a function of the length of embedding as can be seen in Fig.8.

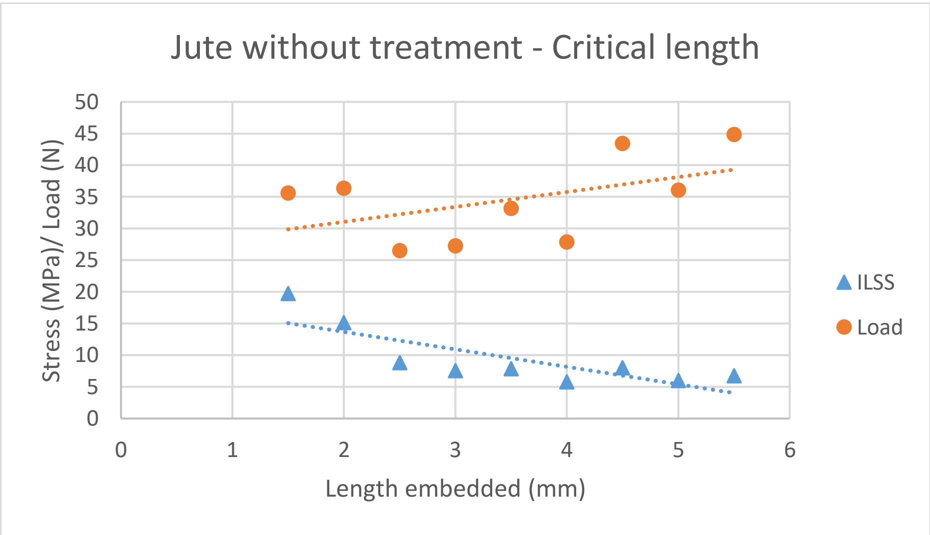


Figure 8: Critical length (mm)

The point of encounter between the curves of the maximum fibre breaking strength and the interfacial shear stress (ILSS) determines the critical length of the embedding. As can be seen in the graph of Fig. 8, the tendency of the curves is to be found in lengths of embedding less than 1mm, which characterizes a good interfacial adhesion between the components. However, due to dimensional constraints of the mould, it was not possible to manufacture again the specimens with smaller lengths.

Therefore, in order to determine the shear stress between the interfaces, the short beam test of three-point bending was chosen.

3.2 Short beam test

The three-point bending short beam test, unlike the pullout test, determines the apparent shear stress between the composite layers. Thus, it is possible to verify the influence of the surface chemical treatments on the resistance to delamination of the composite material and, consequently, to understand the best combination of hybrid composite in order to guarantee a good distribution of stresses between fibre / matrix.

For the short beam tests, the following combinations of hybrid composites were produced:

- J+FG (Jute + Fibreglass);
- J+R (Jute + ramie – without treatment, mercerized and silanized);
- J+S (Jute + sisal – without treatment, mercerized and silanized).

The obtained results can be observed in the Tab. 1, Fig. 9 and Fig.10.

Tabela 1: Short beam results

Type of composites		ILSS (Short beam strength - MPa)	Flexural strength (MPa)	Load (N)
J+GF	Without treatment	15.55 ± 4.43	124.40 ± 35.42	414.67 ± 118.08
J+R	Without treatment (WT)	24.26 ± 3.99	194.09 ± 31.88	646.98 ± 106.29
	Mercerized (M)	21.27 ± 2.54	170.16 ± 20.35	567.22 ± 67.84
	Silanized (S)	27.34 ± 5.25	218.71 ± 41.96	729.04 ± 139.87
J+S	Without treatment (WT)	24.24 ± 5.67	193.89 ± 45.34	646.29 ± 128.60
	Mercerized (M)	21.44 ± 2.44	171.55 ± 19.51	571.85 ± 65.04
	Silanized (S)	24.39 ± 6.41	195.11 ± 41.74	650.36 ± 139.14

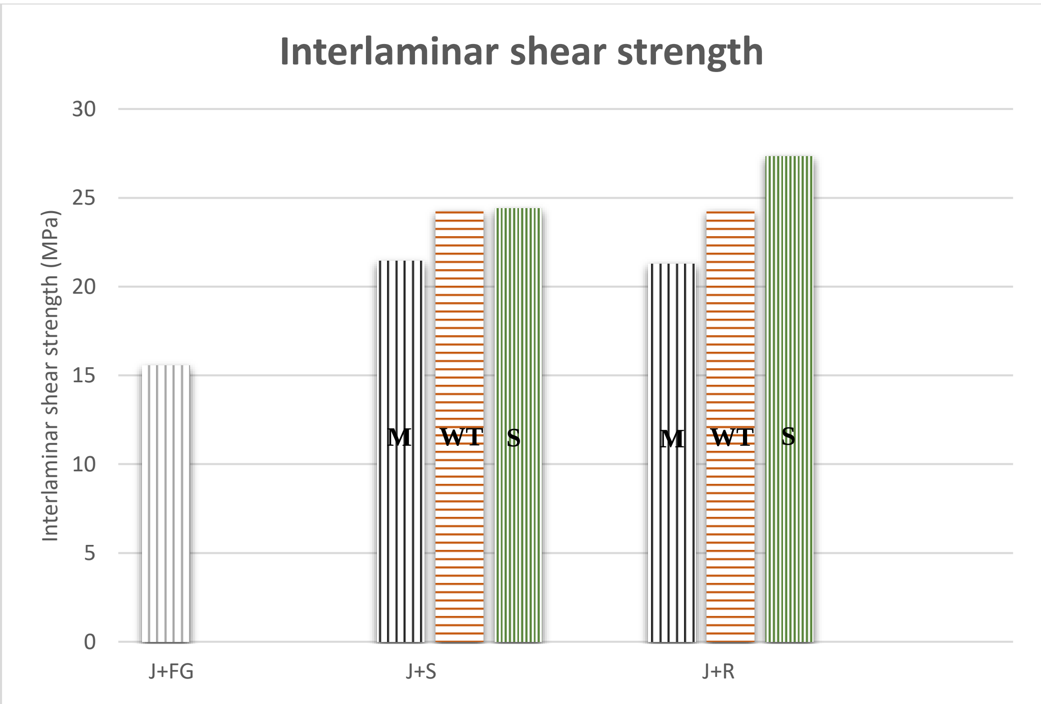


Figure 9: Interlaminar shear strength as a function of the type of composite

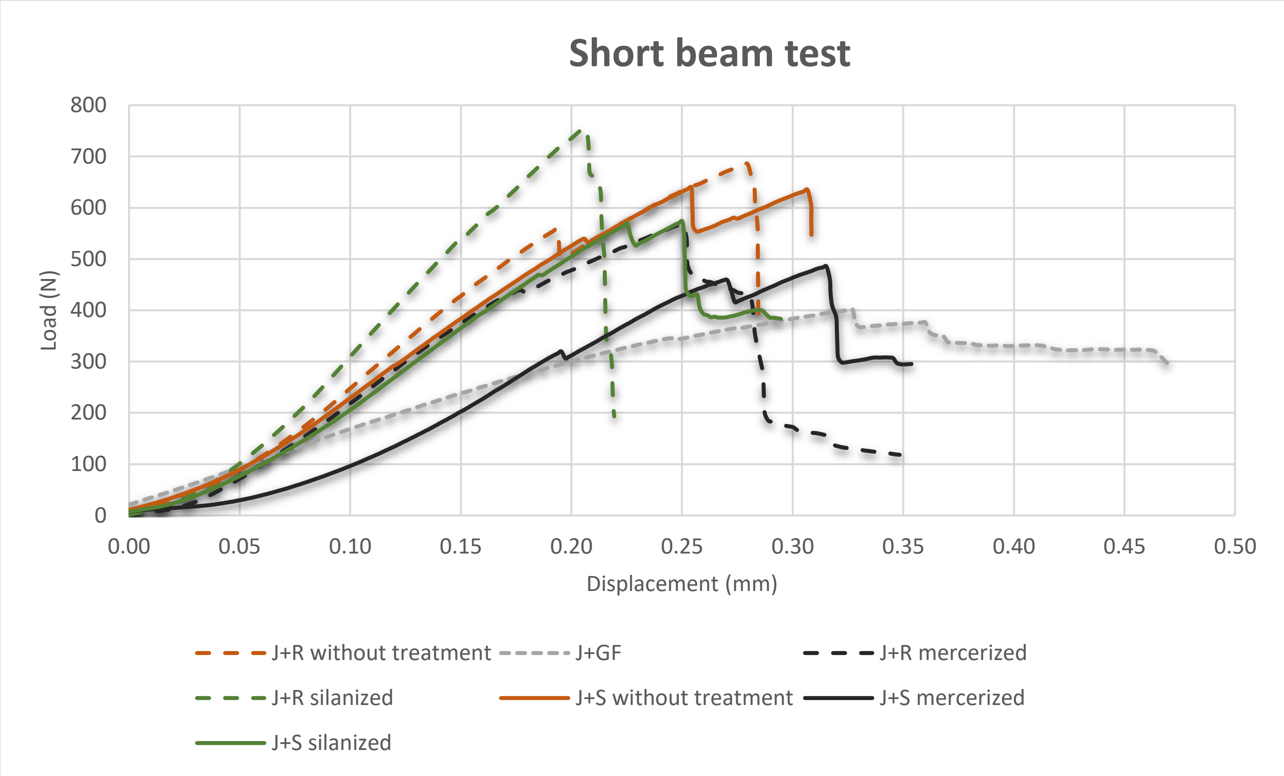


Figure 10: Short beam test – Load x displacement

After the short beam test, the influence of the surface treatments was also verified on the interlaminar adhesion of the hybrid composites produced. The composites with 100% natural fibre fabric presented, in general, better results than the hybrid jute + fiberglass fabric. As can be seen in Tab. 1 and Fig. 9, the jute + ramie silanized composite presented the best result in terms of both interlaminar shear stress and flexural strength. Fig. 10 shows the force displacement graph of the composites produced and from it is possible to observe that the curves characterized by a plateau (J + FG) show lower results when the resistance to delamination. These results are quite consistent with the study by da Silva *et. al*., 2013, on the short beam test in curauá, sisal and fiberglass fibres.

In Fig. 11, it is possible to see the macro of the fracture surface of the composite proving the delamination failure, thus validating the data of the interlaminar shear stress.

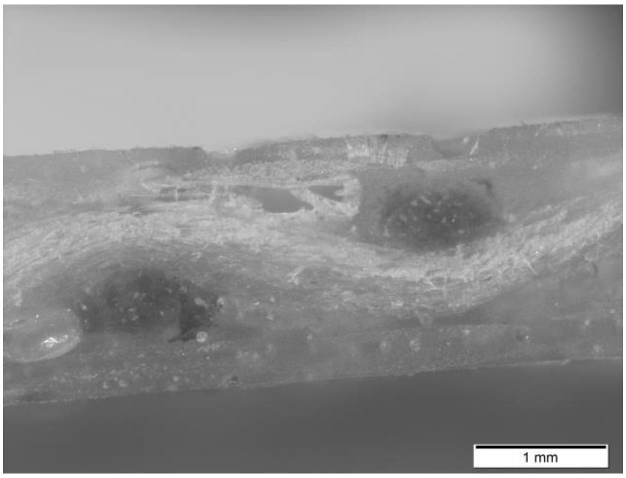


Figure 11: Delamination failure

4. CONCLUSION

The present work relates the influence of surface treatment on natural fibres and the production of intralaminares hybrid composites in the increase of interfacial adhesion between matrix / fibre, in the reduction of the volume of voids and consequently in the increase of the mechanical properties of the composites. The surface chemical treatments showed a clear influence on the mechanical strength of the fibres as in the interlaminar adhesion of the laminated hybrid composite. However, it has been observed that not always the treatment that increases the mechanical strength of the fibre will result in the improvement of the adhesion properties between the fibre / matrix interface. Among all the hybrid composites studied, the one that presented the best result was jute + ramie silanized, followed by the jute + sisal silanized, both in terms of interlaminar adhesion and flexural strength.

5. ACKNOWLEDGEMENTS

The authors acknowledge the financial support of the National Research and Teaching Council (CNPq) and the Coordination for the Improvement of Higher Level Personnel, CAPES for their financial support and the Federal Center for Technological Education Celso Suckow da Fonseca for technical and institutional support.

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