Original Article

Critical length and interfacial strength of PALF and coir fiber incorporated in epoxy resin matrix


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ABSTRACT

The several advantages of natural lignocellulosic fibers (NLFs), such as, economical, technical, environmental and social, make these fibers an alternative to replace synthetic fibers in composite materials. The application of NLFF as reinforcements in polymeric composites has increased in many industrial sectors from civil construction to automobiles. This demands the characterization of promising fibers, such as those extracted from leaves of pineapple (PALF) and the mesocarp of coconut fruit (coir fiber), for possible application in composites. In the present work, pullout tests were performed to compare the interfacial adhesion with epoxy resin of these two fibers that have greatly different characteristics. Results showed a critical length 70% higher for the coir fiber in comparison to PALF and a interfacial strength 3.5 times smaller, which indicates stronger adhesion of PALF with epoxy resin. This may be justified by the distinct morphological aspects, particularly the rougher surface of PALF. Mechanical tests were also performed in both coir fiber and PALF composites. In these tests, it was observed the superiority of mechanical properties for the composite reinforced with 30 vol% of PALF. Additionally, ballistic tests were carried out. In this evaluation, composites were used in a MAS type III against the 7.62 mm ammunition. The results revealed a relatively low depth of penetration (18.2 mm) for the MAS with PALF composite as well as a depth of penetration (31.6 mm) for MAS with coir composite, both considered efficient according to the personal body armor standard. Therefore, all these results highlight the potential of these fibers as polymer composites reinforcement in ballistic armors.

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1. Introduction

Over the last couple of decades, natural lignocellulosic fibers (NLFs) are increasing their market share in many sectors, such as furniture, packaging, building and automotive industries [1–5]. This growing trend is due to their low environmental impact, low cost and technical advantages in comparison to synthetic fibers [2]. However, NLFs have hydroxyl groups in their composition which makes them hydrophilic [6,7]. Indeed, the existing moisture on the surface of the fiber might reduce its interfacial adhesion inside the composite, because polymers commonly used as composite matrix have hydrophobic character. Hence, chemical surface modification or other pretreatment could be applied in NLFs to improve the interfacial adhesion [2,6,8,9].

Initially, Kelly and Tyson [9] proposed the pullout test to evaluate the interfacial adhesion between the fiber and matrix, and later adapted by Monteiro and D’Almeida [10] for lignocellulosic fibers. In this test, it is possible to determine the fiber critical length and the interfacial strength that are associated with the bonding efficiency of the fiber/matrix interface [10].

In the present work, the fibers extracted from the leaves of pineapple (Ananas comosus) known as PALF and the coir fibers extracted from the mesocarp of the green coconut (Cocos nucifera L.) were evaluated without pretreatment in pullout tests embedded in epoxy matrix. These fibers are byproducts of their fruits and, consequently, are inexpensive and abundantly available. The physical characteristics of these fibers differ greatly. The PALF has a high content of α-cellulose (up to 82 wt%), lignin content less than 12 wt% and low microfibrillar angle (14°), whereas the coir fiber is lignin-rich (>41 wt%) and has around 40 wt% of cellulose and microfibril angle greater than 40° [8]. Moreover, the PALF has an equivalent diameter less than 300 μm [11] and the coir fiber has a higher diameter (up to 600 μm) [12]. As a consequence the mechanical properties of PALF are superior than coir fiber and its behavior is quite different [2,13].

In order to compare the adhesion behavior of these two distinct fibers, pullout tests were performed to determine the critical length and interfacial adhesion of PALF and coir fiber with respect to the epoxy matrix, varying the embedded lengths of the fibers as proposed by Kelly and Tyson [9] and adapted by Monteiro and D’Almeida [10].

In addition, mechanical and ballistic tests were carried out to evaluate the influence of interfacial adhesion of PALF/epoxy and coir fiber/epoxy on the properties of their composites.

2. Materials and methods

The coir fibers were supplied by the Brazilian firm “Coco Verde Reciclado” and PALF was provided by Desigan Natural Fibers, Brazil. Fig. 1 shows these fibers as received. Both fibers were dried at 60 °C in an air-oven for 24 h. The epoxy resin of diglycidyl ether bisphenol-A (DGEBA) and hardener triethylenetetramine (TETA) were used as matrix. These two components were mixed with a stoichiometric ratio of phr 13.

Fig. 2 illustrates a specimen used for pullout tests. Epoxy cylindrical blocks with 8 mm diameter were prepared by varying the single fiber embedded length from 2 mm to 43 mm for both the PALF and coir fibers. The length and diameter of the fibers were measured with a Zeiss Stemi 2000C stereoscope.

The pullout tests were performed by means of an Instron universal model 3365 machine with a speed of 0.75 mm/min. Tensile tests were also carried out in 10 single fibers for both PALF and coir fiber.

The correlation between the interfacial adhesion and mechanical properties for both PALF and coir fiber in epoxy matrix was investigated through Izod and tensile tests. Epoxy composites with 30 vol% of PALF as well as 30 vol% coir fiber were produced, both with continuous and aligned fibers.

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**Fig. 1 – Natural lignocellulosic fibers as received: (a) coir fiber; (b) PALF.**

**Fig. 2 – Specimen used for pullout test.**
Izod impact tests were carried out according to the ASTM D256 standard using an instrumented XC-50 model Pantec pendulum. Composites specimens were cut with standard dimensions of $62.5 \times 12.7 \times 10$ mm and notched with a $45^\circ$ of angle, transversely to fibers and the direction of compression molding. Tensile tests were conducted in a model 3365 Instron machine, according to ASTM D638 standard, with a cell load of 25 kN. In order to evaluate the influence of interfacial adhesion in the dynamic response, ballistic tests were performed to compare the absorption capacity of the impact energy caused by a 7.62 mm caliber high velocity ammunition in both 30 vol% PALF and 30 vol% coir fiber composites used as a second layer in a multilayered armor system (MAS). The manufacturing of MAS has been described elsewhere [14]. These tests were conducted at the Brazilian Army Evaluation Center (CAEx), in Rio de Janeiro, Brazil, and the ballistic performance was assessed by the depth of penetration caused in the clay witness as per NIJ 0101.06 standard [15].

Analysis by scanning electron microscopy (SEM) were obtained in a model Quanta FEG 250, Fei microscope operating with secondary electrons at 5 kV.

3. Results and discussion

Fig. 3 shows the results of the pullout tests for the different embedded lengths ($l$) for both PALF and coir fibers with respect to the epoxy matrix. In the first stage, the tensile strength increases linearly with the embedded length of the fiber in the matrix. When this tensile strength reaches the fiber limit stress, then rupture occurs. The embedded length for which the fiber fails is known as critical length ($l_c$) i.e. for lengths below $l_c$, the complete interfacial debonding occurs while at higher $l$ the fiber failure occurs without debonding of the fiber/matrix interface. The interfacial shear strength directly influence the mechanical behavior of the composite [8]. The value of critical length defines if the fiber is long enough to act as reinforcement or it is only an incorporated load. In other words, whether or not there is stress transfer from matrix to the fiber [10].

Fig. 3a presents the linear adjustments of the coir fibers pullout tensile stress vs. embedded length, which correspond to Eqs. (1)–(3):

$$\sigma_l = 8.05 + 8.39 \cdot L$$

(1)

$$\sigma_l = 84.38 + 2.25 \cdot L$$

(2)

$$\sigma_l = 156.05 - 0.24 \cdot L$$

(3)

Eqs. (4) and (5) represent the linear adjustment for PALF (Fig. 3b):

$$\sigma_l = 41.40 \cdot L$$

(4)

$$\sigma_l = 297.05 + 0.53 \cdot L$$

(5)

The intersection of Eqs. (1) and (2) defines the critical length ($l_c$) for coir fiber. The same calculation could be done for PALF through Eqs. (4) and (5). The critical length is reached at $L$ equal 7.3 mm and 12.4 mm for PALF and coir fiber, respectively. Since small values of $l_c$ indicate greater interfacial fiber/matrix adhesion [10], these results of 70% higher $l_c$ for coir fibers reveal better adhesion between PALF and epoxy in comparison with that of coir fiber.

As proposed by Monteiro and D’Almeida [10] it is possible to define a second critical length, $L_c$, for which the fiber does not detach completely from the matrix. This could be observed for coir fiber (Fig. 3a). The value of $L_c$ is given by the intersection of Eqs. (2) and (3), resulting in a length equal 28.8 mm.

It is important to note that the error bars of the tensile strength associated with $L > l_c$ are in the range of tensile strength reported in literature [2,11,16,17] and in the tensile strength of the present work. This behavior is expected and therefore corroborates the experimental data obtained.

The interfacial strength ($\tau_c$) of PALF and coir fiber with respect to the epoxy matrix were evaluated by the equation of Kelly and Tyson [9]:

$$l_c = \frac{d \cdot \sigma_l}{2 \cdot \tau_c}$$

(6)
where $\sigma_f$ is the tensile strength of the fiber and $d$ is the equivalent diameter of the fiber.

For each tested fiber the equivalent diameter was measured as described in the work of Luz et al. [12]. This can be a source of dispersion for NLFs diameters because of heterogeneity in their cross sections, as shown in Fig. 4. The averages of equivalent diameters were 238 $\mu$m and 314 $\mu$m for PALF and coir fiber, respectively. The values obtained for interfacial strength were 1.42 MPa (coir fiber) and 4.93 MPa (PALF).

Therefore, the PALF/epoxy interfacial adhesion was stronger than coir fiber/epoxy and PALF could more efficiently transfer the mechanical strength to the composite matrix. This higher adhesion could be attributed to the naturally rougher surface of PALF [16–20], as illustrated in Fig. 5. Indeed, a rougher surface allows an efficient penetration and anchoring of the epoxy matrix.

Furthermore, the coir fiber has a thin aliphatic surface layer (wax layer) which consists of a long chain of fatty acids that are incompatible with polar matrix composites, resulting in a weak interfacial adhesion, as reported in some studies [4,5,10,21,22]. As coir fibers were used in as-received condition, the presence of some protrusions with silicon-rich particles was observed on their surface (Fig. 6). This may have also contributed to low interfacial epoxy/coir fiber adhesion as noticed by Prasad et al. [21]. They found that the removal of these particles and the wax layer from the coir fiber surface through the NaOH mercerization process promoted a more rugged surface resulting in a 90% increase in interfacial strength over untreated fiber in the matrix polyester [21]. Fig. 7 reveals the better impregnation of the epoxy resin in PALF than coir fiber, which may contribute to the higher interfacial adhesion of PALF/epoxy. Moreover, higher tensile strengths obtained for single PALF fibers corroborate the trend reported in several studies indicating higher mechanical strength for NLFs with smaller diameters that could be explained by the lower probability of internal defects [2].

Table 1 shows the mechanical properties obtained in tensile tests and the impact energy absorbed in Izod tests for both epoxy composites reinforced with 30 vol% PALF and 30 vol% coir fiber, in the longitudinal direction. It is clear that the PALF composite presented more than twice the value of both tensile strength and elastic modulus when compared to the coir fiber composite. This can be explained by the predominance of the brittle fracture mechanism for coir fiber composites, which is typical of the epoxy resin. It also indicates that coir fibers acted only as composite load, and no effective reinforcement occurred in relation to the tensile properties. By contrast, a higher pullout incidence was observed in PALF composites in comparison to coir fiber composite. In PALF composites, the brittle fracture mechanisms of the epoxy matrix together with the debonding and rupture of the fiber justify a significant increase in tensile strength and stiffness.

Although the heaviest hammer available (22 J) was used in the Izod impact test, samples of composites reinforced with 30 vol% PALF did not break completely. Therefore, the impact energy value for the PALF composite could not be compared with the result of coir fiber composite, in which a complete rupture occurred. However, the fact that PALF composite sam-

![Fig. 4 – SEM of the heterogeneities of the NLFs cross sections: (a) coir fiber; (b) PALF.](image1)

![Fig. 5 – NLFs surfaces: (a) coir fiber; (b) PALF.](image2)
Fig. 6 – SEM showing the presence of some protrusions with silicon-rich particles in the surface and the debonding interface.

Fig. 7 – SEM of the interface of composites: (a) coir fiber/epoxy; (b) PALF/epoxy.

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<th>Table 1 – Mechanical properties of epoxy composites reinforced with 30 vol% PALF and 30 vol% coir fiber.</th>
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<td>Properties</td>
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* Did not occur complete breakage.

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<th>Table 2 – Depth of penetration (DOP) for MAS with different composites as second layer.</th>
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<td>MAS second layer</td>
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Fibers did not break completely is an indicative of the high toughness of this composite, provided by PALF reinforcement. In fact, if there were total rupture of this composite, the energy absorbed would be even higher. In addition, the early failure and low values of impact strength of coir fiber composite can be attributed to the weak interfacial interaction between coir fiber and epoxy resin.

DMA results are presented elsewhere [23], and support the mechanical testing results. The curves of storage modulus also indicated a greater reinforcement of epoxy composite with 30 vol% PALF in comparison to 30 vol% coir fiber. This trend is given by the raising of the thermal mechanical stability at high temperature denoted by a higher rubbery plateau for PALF composite. These results also revealed an increase of loss modulus and glass transition temperature values, which means an efficient stress transfer through PALF/epoxy interface.

The depth of penetration (DOP) in ballistic tests, corresponding to the residual impact energy dissipated by the MAS, was measured by means of a laser sensor and is shown in...
4. Conclusions

- The coir fiber presented in pullout tests a critical length 70% higher than PALF, which indicates weaker interfacial adhesion between this fiber and epoxy matrix. Also, the value of interfacial strength for the PALF was 3.5 times greater than coir fiber. This result could be justified by the rougher surface of PALF and the presence of wax layer in the coir fiber;
- A second critical length was found for the coir fiber. In the embedded length between 12.4 mm and 28.8 mm the coir fiber did not detach completely from the epoxy matrix;
- It was observed that the statistic dispersion of the tensile strength associated with higher length is in the range of tensile strength for single fiber found in literature and in the present work for both PALF and coir fiber. This high dispersion is a consequence of their cross section heterogeneities;
- Coir fiber presented an equivalent diameter 30% higher than PALF, which explains the higher strength associated with PALF and corroborate the trend reported in several related works;
- Tensile tests showed superior mechanical properties of PALF composites as well as higher capacity of PALF composites to absorb energy in Izod test in comparison to coir fiber composite. This trend is supported by results of DMA tests already published;
- In ballistic tests, the PALF composite exhibited the lowest depth of penetration (18.2 mm), which represents a higher ballistic performance as compared to MAS with coir fiber composite. Combined, all these results highlight the potential of the PALF composite in ballistic armors.

Conflicts of interest

The authors declare no conflicts of interest.
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REFERENCES