Hybridization effect on the mechanical properties of curaua/glass fiber composites

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1. Introduction

Glass fiber reinforced polyester composites are largely used mainly due to a combination of low cost and good mechanical properties. However, in the last years, the growing environmental awareness and new legislation boosted the use of bio-fiber reinforced polymer composites. These composites have some advantages in relation to glass or other synthetic/mineral fibers, since they are environmentally friendly, and yield low abrasion in processing, together with low cost and specific gravity.

Sisal, flax, hemp, jute, banana and curaua are all natural fibers used to reinforce polymer composites (thermosets and thermoplastics) [1]. Curaua is a natural fiber still poorly explored in relation to sisal or jute, even though it shows excellent properties (in relation to other natural fibers) such as elongation at break, tensile strength and low density, providing good specific properties. The cost of curaua fiber is similar to other vegetable fibers but curaua is considered odorless as opposed to some other fibers [2]. In applications with less strict mechanical loads such as car interior, furniture and acoustic insulation components, the curaua/polyester composites can be considered as an alternative to glass fiber composites. However, curaua fiber has also disadvantages, such as high moisture absorption (like all vegetable fibers), inferior mechanical properties (in relation to synthetic fibers), highly polar surfaces [3] and poor adhesion to most polymeric matrices. Thus, hybridization of vegetable and glass fibers appears promising.

The aim of this paper was to evaluate the effect of hybridizing glass and curaua fibers on the mechanical properties of their composites. These composites were produced by hot compression molding, with distinct overall fiber volume fraction, being either pure curaua fiber, pure glass fiber or hybrid. The mechanical characterization was performed by tensile, flexural, short beam, losipescu and also nondestructive testing. From the obtained results, it was observed that the tensile strength and modulus increased with glass fiber incorporation and for higher overall fiber volume fraction (3Vf). The short beam strength increased up to 3Vf of 30 vol%, evidencing a maximum in terms of overall fiber/matrix interface and composite quality. Hybridization has been successfully applied to vegetable/synthetic fiber reinforced polyester composites in a way that the various properties responded satisfactorily to the incorporation of a third component.

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with intralaminate hybrid curaua/glass fiber composites with distinct fiber curaua/glass content and overall fiber content. Theoretical predictions based on micromechanical analysis of hybrids were included for comparison.

2. Materials and methods

2.1. Materials

The following materials were used:

- Curaua fiber (interlaced rope), from a cooperative located at the Para State of Brazil. Curaua composition is, in weight, around 74% cellulose, 7.5% lignin, 10% hemicellulose, 1% ashes and 9% moisture [11].
- Glass fiber roving ME 3050 (areal density of 4000 tex), from Owens Corning.
- Commercial unsaturated and accelerated orthophallic polyester resin ARAZYNE® 13.0 (supplied by Ashland).
- Acetyl acetone peroxide (AAP), as initiator, and degassing agent A515 (BYK).

2.2. Composite molding

The curaua fiber rope was disentangled and classified. Both fibers (curaua and glass) were cut to 50 ± 2 mm in length. This is around 10 times the curaua fiber critical length (in polyester) [12]. The chopped fibers were randomly distributed in a pre-mold (with the same dimensions of the mold) and compacted using hydraulic press (Marconi – MA 098/A3030) for 30 min at 3 ton and room temperature. All mats, except the pure glass one, were dried in an oven at 105 °C for 30 min prior to molding.

The resin system was prepared by adding 2 wt.% of curing agent and 2 wt.% of degassing agent, in relation to the polyester. Both were followed by 2 min of manually stirring for homogenization. The resin was then submitted to ultrasonic bath at room temperature for 5 min. The composites were produced by compression molding under 6 ton, at 90 °C for 75 min, followed by post-curing at 80 °C for 120 min in an oven with air circulation. The overall fiber volume fraction (%Vf) was varied within 20–40% and the curaua/glass fiber ratio also was varied. Table 1 shows the sample codes and fiber contents.

2.3. Characterization

The rule of mixtures (Eq. (1)) was used to estimate the density of the composites:

\[ \rho_c = \left[ (C_{cu} \times \rho_{cu}) + (C_g \times \rho_g) \right] \times V_f + (\rho_m \times V_m) \]

where \( \rho_{cu} \) and \( \rho_m \) are the density of curaua (1.38 g cm\(^{-3}\)), glass (2.54 g cm\(^{-3}\)) and matrix (1.18 g cm\(^{-3}\)), respectively [9]; \( C_{cu} \) and \( C_g \)

are curaua and glass fibers relative content; \( V_f \) and \( V_m \) are the overall volume fraction of fiber and matrix, respectively.

Tensile tests were carried out using an Instron 3382 universal testing machine with a 100 kN load cell. Seven samples (dimensions: 170 × 25 × 3 mm) were tested at constant speed (2 mm min\(^{-1}\)), according to ASTM D3039-08 standard. Three-point bending flexural tests were carried out in the same equipment using eight samples (127 × 12.7 × 3 mm) with span-to-depth ratio of 16:1, at 2 mm min\(^{-1}\), following ASTM D790-10 standard. The losipescu shear test was performed in a Shimadzu (AG-X model) testing machine, with load cell of 50 kN and test speed of 0.5 mm min\(^{-1}\). Three rectangular specimens (76 × 20 × 3 mm) with two symmetrical centrally located V-notches were tested, according to ASTM D5379-05 standard.

The short-beam shear tests (ILSS) was performed according to ASTM D2344-00 standard, using span-to-depth ratio of 4:1 and test speed of 1 mm min\(^{-1}\), in an EMIC DL3000 (load cell of 200 kN) testing machine. Twenty samples were tested for each family and, as per this ASTM standard, sample length is 6 × thickness and width is 2 × thickness. The fractured samples were observed using scanning electron microscopy (SEM) in a JEOL (Model 6060) equipment.

The elastic properties of non-used losipescu specimens were also evaluated using non-destructive testing based on impulse excitation of vibration, carried out in Sonelastic® (ATCP Engenharia Fisica) equipment. In this technique, the specimen under testing is subjected to a mechanical tap and reacts emitting an acoustic response. The elastic moduli are calculated from the natural frequencies of vibration extracted from the acoustic response by Fast Fourier Analyses. The specimen emits an acoustic response which depends on its elastic properties, dimensions and mass, and the elastic (E-modulus) [13] can then be obtained using Eq. (2), according to ASTM E1876-09 standard:

\[ E = 0.9465 \left( \frac{m f^2}{W} \right) \left( \frac{L}{T} \right)^3 \quad \text{and} \quad T = 1 + 6.858 \left( \frac{L}{T} \right)^2 \]

where \( m \) is the mass, \( f \) the fundamental resonance frequency in flexure, \( L \) the length, \( w \) the width, \( r \) the thickness and \( T \) the correction factor.

3. Results and discussion

3.1. Tensile properties

The stress–strain curves of the 20% %Vf samples are presented in Fig. 1. It can be seen that the hybrid samples presented intermediate behavior in relation to the pure composites. The glass fiber composite (sample 20/0/100) shows higher tensile strength, modulus and elongation at break than the curaua one.

Fig. 2 shows the tensile properties found for all studied composites. The tensile strength of the neat polyester resin was of 45.7 MPa and the addition of 20 vol.% of fiber yielded a significant increase in strength but only if at least some of this fraction was comprised of glass fiber. This is due to its higher strength and modulus and better adhesion to the matrix in relation to the curaua fiber [14].

The composites with 20 vol.% showed high standard deviation in general, probably since the low fiber content leads to resin-rich regions in the mats. The samples with 30 vol.% presented an intermediate behavior, and samples with 40 vol.% yielded better results, showing lower standard deviation and higher strength, mainly for the hybrid and the pure glass fiber composites. It is also interesting to see that the 30/30/70 and 30/0/100 samples were in the same range of strength (considering the deviation), and that the higher the overall amount of fiber, the more difficult it is for the hybrid
to match the pure glass composite since this is a fiber-dominated property.

In Fig. 2, the specific strength shows an increasing trend for higher glass fiber content, even though this fiber has higher density (meaning specific gravity) than curaua. Indeed, density increases with the overall fiber content, and with the relative glass content. In addition, it is interesting to notice that specific strength (i.e. strength/density) increased more clearly with more effective reinforcement (glass fiber) in each %\( V_f \). Among the hybrid specimens, the 30/30/70 presented higher specific strength.

It is hard to find any papers in the literature on the theoretical properties of natural/synthetic random short fiber hybrid composites. For random in-plane short fiber composites, the micromechanical model developed by Cox and Krenchel[15,16] is one of the most commonly used. This model includes a limiting stress transfer efficiency factor named \( g_L \), based on fiber length and other factors, and a parameter to account for the type of fiber orientation, named \( g_o \)[17]. Cox[15] introduced the \( g_L \) factor in the rule of mixture aiming to account for a relatively ineffective loading of fibers along its stress transfer length in short fiber composites (compared to continuous fibers)[17]. Krenchel[16] introduced \( g_o \) to adjust the model when the fiber orientation is not exclusively that of the loading, being equal to 0.375 for random in-plane composites.

In this work, the Cox and Krenchel model was adapted to intralaminate hybrid composites, as follows:

\[
E_c = \eta_L V_f (\eta_L C_g E_g + \eta_o C_{cu} E_{cu}) + (1 - V_f) E_m
\]

where \( E_c, E_g, E_{cu} \) and \( E_m \) are the modulus of composite, glass fiber (73 GPa [2]), curaua fiber (36 GPa [3]) and matrix (2.1 GPa [this work]), respectively. The parameters \( \eta_L \) and \( \eta_o \) are the length efficiency factor of the glass and curaua fibers, respectively.

Input data for this model include \( l \) (fiber length), \( d \) (fiber diameter, 16 µm and 72.4 µm for glass and curaua, respectively [this work]), \( G_m \) (shear modulus of the matrix, 789 MPa, for Poisson’s coefficient of 0.33 [this work]) and \( X_i \), which depends on the geometrical arrangement of fibers. The \( X_i \) factor was considered equal to 4.0 (the same for square packing), based on Thomason and Vlug [18] and Robinson and Robinson [19]. Full description of this model can be found in the literature [17].

The Halpin–Tsai model, widely used to predict the elastic modulus of composites, was also employed for comparison. The elastic modulus can be estimated by:

\[
E_c = \frac{3}{8} E_1 + \frac{5}{8} E_2
\]

where \( E_1 \) and \( E_2 \) are the predicted modulus for short unidirectional fibers in the longitudinal and transverse directions. The full description can be found in the literature [20].

The theoretical modulus predictions based on Cox–Krenchel and Halpin–Tsai model for isotropic in-plane composites and the obtained experimental results are shown in Fig. 3. It can be observed that the same trend was found for all of them, i.e. stiffness increases with glass fiber incorporation. Also, the predictions were generally close to the experimental results. It is also worth noting that incorporation of curaua (100% in weight) led to 221% increase in tensile modulus (in relation to the pure resin) and that stiffness of the 20/30/70 and 30/30/70 hybrid composites were similar to the respective pure glass fiber composites. Both theoretical models yielded the same trend, but experimental data adjusted better to the Cox–Krenchel theory.

In polymer composites, adding fibers does not always improve the mechanical properties, because there should be sufficient matrix to transfer load to the fibers. Besides, quality of the composites varies with %\( V_f \) due to experimental conditions (mat production and composite molding). In the case of this work, the elastic modulus of the composites with 30% overall fiber content showed optimized behavior. The elastic modulus \( E \) of the hybrid composites were also evaluated with nondestructive testing (NDT), and the results
are included in Fig. 3. The elastic modulus presented the same
general trend obtained with tensile testing, i.e. the elastic modu-
lus increased for higher overall fiber content and for higher glass
fiber content. Modulus increased up to $30\% V_f$ of 30 vol.%, and the
hybrid 30/30/70 presented the highest modulus among the
hybrids. It is important to mention that the values obtained with
impulse excitation technique may be somewhat different from
those obtained from tensile tests, since they are based on differ-
ent principles.

3.2. Flexural and shear properties

Flexural strength of the composites is shown in Fig. 4a. In the
flexural test, some mechanisms act simultaneously such as tension,
compression and shearing. In general, it can be seen that the hybrid
composites showed intermediate behavior between the pure cur-
aua fiber and the pure glass fiber composites. Some fluctuations
in strength can be credited among others to thickness variations
in the samples.

Flexural modulus (Fig. 4b) showed a similar trend, i.e. an in-
crease with glass fiber content. But, in this case, the increase with
the overall fiber content was clearer. The increase in flexural mod-
ulus for higher $V_f$ may be related to good matrix–fiber stress
transfer [14], indicating good interaction between them. It can also
be noticed that the 40/30/70 hybrid composite showed similar
modulus than the pure glass fiber composite with 30% overall fiber
content. These results are in accordance to those obtained by Idicu-
la et al. [1] who obtained higher flexural modulus for higher $V_f$
and for higher content of the more rigid fiber in the hybrid com-
posite (with sisal and banana fibers).

The short beam test, formerly known as interlaminar shear
strength (ILSS), may be used to evaluate interfacial strength and
may also indirectly address fiber/matrix adhesion and void content
[21]. Fig. 5 shows that short beam strength (SBS) of the composites
increased with glass fiber incorporation within each group. This
behavior is due to the more complete fiber wetting and stronger fi-
ber/matrix adhesion obtained with the synthetic fiber in relation to
the curaua fiber, which has hydrophilic character, yielding lower
compatibility.

SBS also increased with overall fiber content but only up to
30 vol.%, decreasing afterwards. SBS is primarily dependent on
the matrix properties and the fiber/matrix interfacial properties
rather than the fiber properties [22]. The decrease in SBS for
40 vol.% specimens may be due to a large amount of fiber–fiber
contact and because poorer quality composites are obtained with
the molding technique used (i.e. compression molding) when high
fiber content is employed, which is likely to lead to larger void con-
tent. This is expected to have a more pronounced detrimental ef-
fect in properties like SBS and in-plane shear.

The failure in short beam testing occurred suddenly, with hardly
no prior damage. In most specimens, small interlaminar longitudi-
nal cracks were noticed in the mid-plane, differently from a single
major crack through the specimen length, as reported for lami-
nates [23], as shown in the micrographs of Fig. 6. Fiber length influ-
ences SBS and the 50 mm length is adequate for good matrix/cur-
aua fiber stress transfer perhaps due to the extent of fiber bridg-
ing effect during crack propagation [24,25]. This is in the same
range of length (45–60 mm) reported by Silva et al. [25] for maxi-
mum SBS.

The Iosipescu shear strength of the composites is presented in
Fig. 7. These results did not show a clear trend, and most of the
composites presented shear strength within the 20–24 MPa range,
with relatively large deviation. Variations in specimen thickness
and experimental difficulties (e.g. related to machining the sym-
metrical notches of specific dimensions when vegetable fibers are
used) may have negatively influenced these results.
the two fibers and the resin. Whereas the in-plane shear strength increased for higher fiber volume fraction, evidencing results also favored the hybrid composites. The specific tensile behavior is mainly due to the higher strength and stiffness of the glass fiber in relation to curaua and the better adhesion of the former with the polyester resin.

The theoretical predictions based on the micromechanical Cox–Krenchel and Halpin–Tsaï models for stiffness of in-plane isotropic composites ratified the experimental results. Hybridization was considered successful since replacing 30 vol.% of the glass by curaua fiber produced mostly similar results. The specific stiffness results also favored the hybrid composites. The specific tensile strength increased for higher fiber volume fraction, evidencing good fiber–matrix load transfer within the studied range of $V_f$. The elastic modulus was also evaluated with a nondestructive test based on vibration excitation technique. The obtained elastic modulus was similar to that of the tensile test, increasing upon incorporation of the more rigid fiber and for higher $V_f$ (up to 30 vol.%).

Regarding short-beam shear behavior, strength increased up to 30 vol.%. This was considered the optimum fiber content, showing greater fiber/matrix interface and an optimum balance between the two fibers and the resin. Whereas the in-plane shear strength did not show a clear trend probably due to experimental difficulties related to the notching of the specimens.

In all, hybridization was successfully carried out and the mechanical properties presented intermediate behavior between the pure glass and pure curaua composites, sometimes close to the pure glass fiber composites. Among the studied composites, the 30 vol.% overall content with 30% of curaua and 70% glass fiber showed optimum overall performance. Also, the specific properties highlighted the fiber hybridization advantage, which is able to combine lower weight and high mechanical performance, being also more environmentally friendly in this case.

4. Conclusions

The scope of this study was to evaluate the mechanical properties of natural (curaua) and synthetic (glass) fibers reinforced polyester composites with distinct overall fiber volume fraction and curaua/glass fiber ratio. The tensile strength and elastic modulus increased upon glass fiber incorporation and for higher $V_f$. This behavior is mainly due to the higher strength and stiffness of the glass fiber in relation to curaua and the better adhesion of the former with the polyester resin.

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References


