Original Article

JOURNAL OF REINFORCED PLASTICS & COMPOSITES

Journal of Reinforced Plastics and Composites
2014, Vol. 33(5) 472–478

DOI: 10.1177/0731684413517519
jrp.sagepub.com
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Analysis of curaua/glass hybrid interlayer laminates

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Abstract

This study compares the mechanical properties of curaua, glass and hybrid curaua/glass laminates evaluated experimentally with those obtained theoretically using commercial software for composites. Hybrids laminates were produced varying the fiber layer stacking sequence with the aim of optimizing the combination of vegetable and synthetic fibers. Composites were compression molded using eight layers of glass and/or curaua fiber mats and mechanically tested as per ASTM standards. The combination of curaua and glass fibers produced a material with intermediate properties and, depending on the type of mechanical loading and the stacking sequence, some hybrids showed properties close to the pure glass fiber composites. Furthermore, hybrid composites with glass layers closer to the surfaces showed the best overall results.

Keywords

Hybrid, vegetable fiber, curaua, micromechanics, composites

Introduction

Development of alternative materials from renewable resources is an interesting field due to its environmental appeal. Among these materials there are vegetable fibers (such as curaua and sisal) which may be applied in the reinforcing of polymeric materials to produce composites. Vegetable fibers are lignocellulosic materials and their mechanical properties are said to improve for higher cellulose content, higher degree of polymerization and lower microfibril angle in relation to the longitudinal fiber axis. These properties show a significant influence on the composite tensile strength and stiffness.[1] Curaua, in particular, is cultivated in the Amazon region of Brazil, under semi-arid conditions, and its chemical composition has been reported as 73.6% of cellulose, 9.9% of hemicellulose, 7.5% of lignin and 0.9% of ashes.[2]

Vegetable fibers offer advantages such as low density, low toxicity and low cost compared to synthetic ones. Furthermore, curaua and sisal fibers, for example, exhibit specific modulus and mechanical strength comparable to glass fibers for non-structural applications despite their inferior absolute tensile properties.[1,3] Due to that, natural fibers appear as an alternative for the partial or full replacement of glass fibers in some composites.

Another interesting possibility regards the combined use of glass and vegetable fibers to reinforce a single matrix, producing a hybrid composite. This way, one could take full advantage of suitable characteristics of each constituent, yielding optimal overall performance. It is generally accepted that the mechanical properties of hybrid composites are controlled by factors such as aspect ratio, content and length, orientation and grade of fibers intermingling. Fiber/matrix interface bonding, arrangement of both fibers, i.e. hybrid design[4]

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(e.g. individual fiber ply or mixed fiber ply), and failure strain of both fibers also have significant influence on properties, and maximum hybrid results are usually obtained when fibers are highly strain compatible.[5]

Amico et al.[6] observed that tensile strength of sisal/glass fiber mats laminate composites is not much dependent on the sequence of laminas, but glass mats on the surfaces of the composite improves its flexural properties. Pavithran et al.[7] also verified that sisal/glass laminates with glass fibers in the stacking ends improve mechanical properties and help protecting the vegetable fibers from the moisture. Khalil et al.[8] verified that acai/glass fiber composites with only a glass lamina in the mid-plane showed higher tensile strength compared to hybrid composites which have glass laminas at the surfaces, which was justified by the stronger fiber/matrix interface adhesion improving the load transfer process.

In this context, the aim of this work is to evaluate the mechanical properties of curaua, glass and various curaua/glass hybrid composite laminates under tensile, flexural, Iosipescu and short-beam tests, investigating experimental and theoretical properties of these laminates as a function of the layer stacking sequence.

Experimental section

The following materials were used for the composite molding:

- Polyester resin Arazyn 13.0 from Ara Ashland;
- Initiator acetyl-acetone peroxide (AAP);
- E-glass fiber CSM mats (aerial density = 300 g/m^2) from Owens Corning cut from the roll;
- Curaua fiber (obtained from the North region of Brazil). Fibers were untangled, cut to 50 mm length and subsequently immersed in distilled water for 1 h to remove impurities. Then, fibers were dried for 1 h at 100°C in air-circulation oven. Mats were subsequently produced with the curaua short fibers following manual and random homogeneous distribution.

Composites were molded using: curaua fiber mats only, glass fiber mats only and a stacking of glass and curaua fiber mats, keeping in all cases constant overall fiber volume fraction (%Vf) of 30%. In the hybrid composites, a 50/50 (v/v) ratio between curaua and glass was used. Polyester resin was mixed with 1.5 wt% of the peroxide and the resin mixture was degassed with sonication for 5 min and cast on the steel mold with the fibers. The composite was then produced by hot compression molding. To monitor fiber content, density of the composites, the individual fibers and the resin were evaluated by picnometry.

Six different types of composites were produced, two of them single fiber composites and four of them symmetric hybrid laminates in which the stacking sequence of mat layers was varied. Composites used in this work were named: curaua, $[c_2/g_2]_s$, $[c/g]_2s$, $[g/c]_2s$, $[g_2/c_2]_s$, in which c and g mean curaua and glass, respectively. Specimens were prepared according to the ASTM standards following a cutting and polishing protocol prior to testing. The cross-section of the laminates was analyzed to determine actual thickness of each lamina using a Carl Zeiss model AX10 optical microscope.

Flexural tests were performed according to ASTM D790 using a universal testing machine EMIC DL3000 with a 200 kN load cell. Eight specimens (dimensions: $170 \times 12.7 \times 3$ mm) were used for each composite family. Tensile tests were performed according to ASTM D3039, in a universal testing machine INSTRON-8801 with 100 kN load cell, using 2 mm/min and an extensometer. Eight specimens (dimensions: $170 \times 25 \times 3$ mm) were used for each family.

Short-beam tests (formerly known as ILSS) were performed according to ASTM D2344 using the same equipment and load cell described above for the flexural tests. Fourteen specimens (length = 6 × thickness; width = 2 × thickness) were used for each family. Iosipescu tests were performed according to ASTM D5379 using a Shimadzu universal testing machine (model AG-X, 50 kN load-cell) with speed of 1 mm/min. Eight specimens were used for each family.

Results of mechanical properties were analyzed with a single-factor ANOVA in commercial software. A confidence level of 95% was considered, so that if $p < 0.05$, the families were considered to differ.[9] The ANOVA results are presented as lower-case letters in graphs. In addition, commercial software called Laminator (version 3.6) was used to estimate the theoretical hybrid laminate properties. This software was used to estimate tensile and shear modulus of each hybrid laminate based on their construction and the properties of individual laminas.

Results and discussion

Mechanical properties

Figure 1 shows images of laminates cross sections obtained by optical microscopy. The main characteristics that can be visualized include good dispersion of vegetable fibers and irregularity in thickness of the various laminas (justified by the mat architecture). Thickness of each layer of the hybrid laminates has been obtained from the micrographs, as illustrated in Figure 1(b). The mean curaua/glass thickness ratio in the hybrids was 1.21:0.93; 1.3:0.49; 1.26:0.48 and

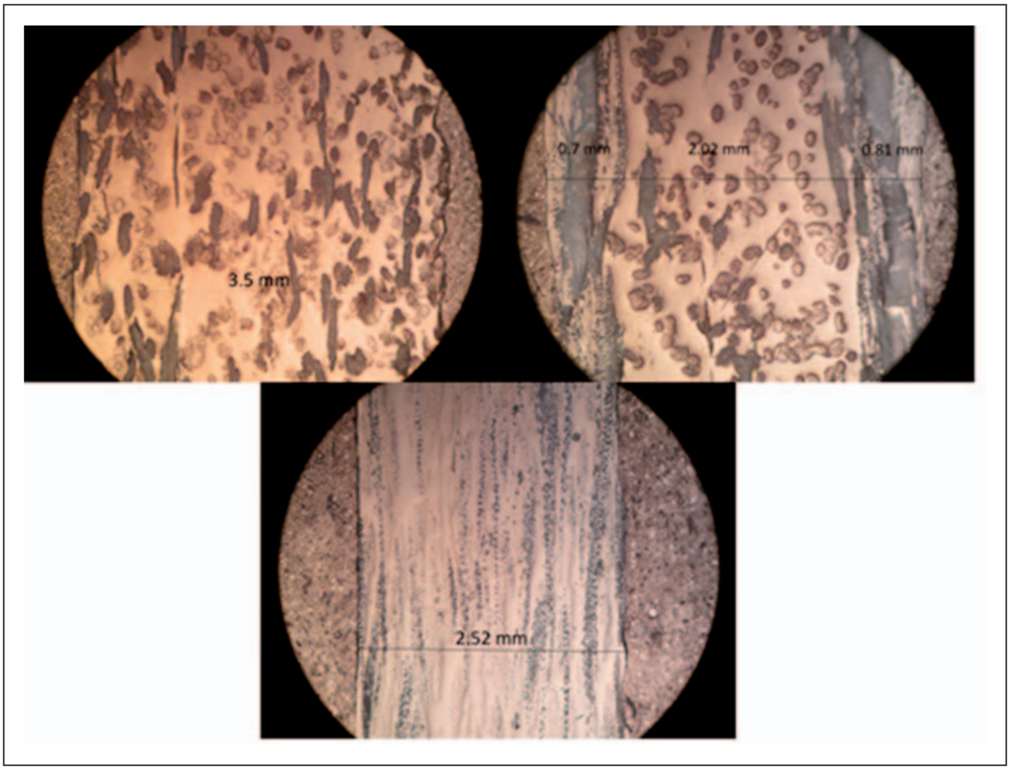

Figure 1. Optical microscopy of the laminates: (a) curaua, (b) $[g_2/c_2]_s$ and (c) glass.

1.01:0.75 for laminates $[c_2/g_2]_s$, $[c/g]_{2s}$, $[g/c]_{2s}$ and $[g_2/c_2]_s$, respectively. The actual overall thickness of the laminates, the overall volumetric fraction and weight fraction are presented in Table 1.

Figure 2(a) and (b) shows the mean results of flexural properties of the laminates. Flexural tests are easy to carry out but difficult to interpret because the stress state changes through the thickness of sample.[10] Nevertheless, the results showed, in general, higher mechanical strength and stiffness for the pure glass composite, intermediate values for the hybrid laminates and lower for pure curaua composite. Besides, hybrids with vegetable fibers on the surface yielded lower strength and stiffness than the other hybrids. The $[g/c]_{2s}$ sample showed higher strength than $[g_2/c_2]_s$ due to the higher fiber content of the former. This is in agreement with the findings of Gowda et al.,[11] who reported that flexural strength and modulus of composites are controlled by the strength of the outer reinforcement layer, which in this case, is the synthetic fiber (with superior mechanical properties than the vegetable fiber).

Cracks initiated in the tensile side of the beam and slowly propagated in an upward direction, showing little or no fiber pull-out. It is important to stress out that better adhesion is expected between glass/polymer than curaua/polymer, which is more flexible.[12] The results of maximum strain were somewhat scattered and no particular trend was identified except that the glass composite fractured at lower strain than the curaua composite, around 5% and 17%.

Table 1. Actual thickness, volumetric fraction and weight fraction of laminates.

Samples	Actual overall thickness (mm)	Overall fiber volumetric fraction (%)	Overall fiber weight fraction (%)
Curaua	3.50	32	36
$[c_2/g_2]_s$	4.28	35	48
$[c/g]_{2s}$	3.59	35	47
$[g/c]_{2s}$	3.48	40	46
$[g_2/c_2]_s$	3.53	34	46
glass	2.52	37	57

Figure 3(a) and (b) shows the mean results of tensile properties of the laminates. Hybrid composites displayed similar strength values, in between those of pure glass and pure curaua composites. The hybrids tended to show the same strength because the major variation is in the laminate stacking only. Under tensile loading, all laminas are subject to the same strain and the properties will be mainly dependent on fiber strength and modulus, fiber length and orientation,

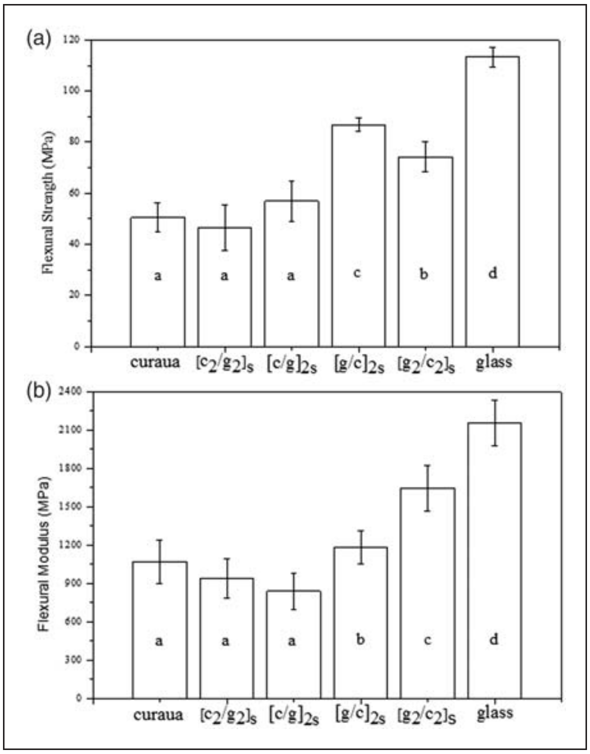

Figure 2. Mean values of flexural strength (a) and modulus (b) of the laminates.

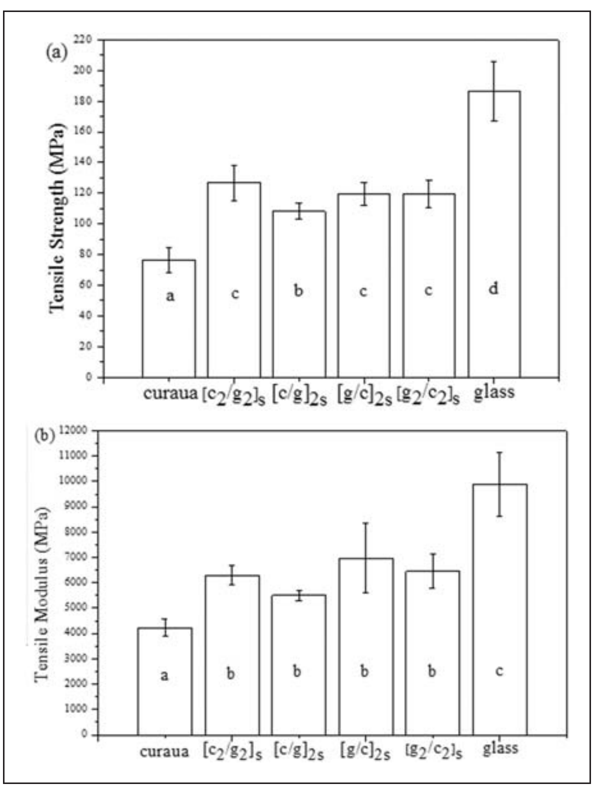

Figure 3. Mean values of tensile strength (a) and modulus (b) of the laminates.

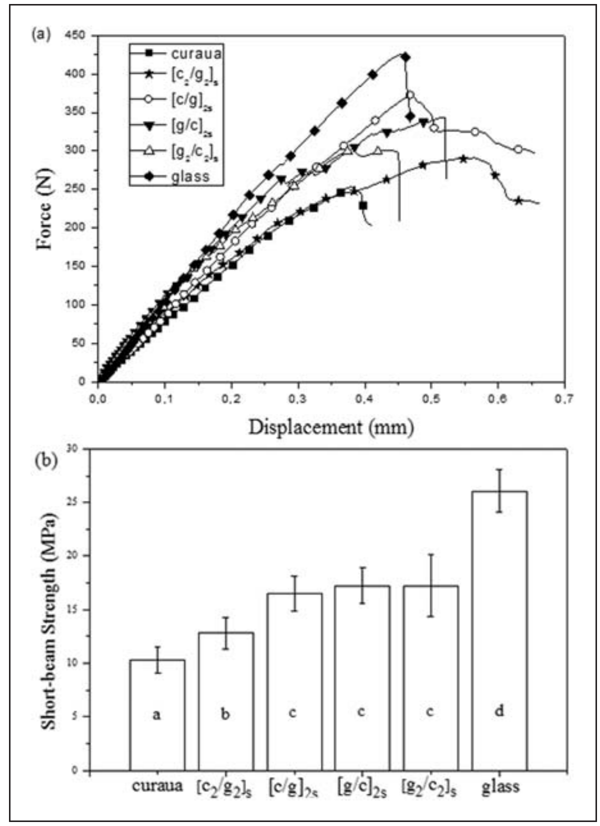

Figure 4. Typical short-beam curves of the laminates (a) and mean inter-laminar shear strength of the composites (b).

fiber/matrix interfacial bonding and fiber content,[10] and these factors did not vary significantly within the hybrids.

The lower strength obtained for the $[c/g]_{2s}$ laminate could be a consequence of the clamping of the samples, since the grip may induce early damage to the more sensitive curaua layers located at the surfaces of the lay-up during testing.[13] Results for modulus followed by the same general trends, i.e. hybrid composites showed intermediate values between curaua and glass composites, with no significant differences within the hybrids, whereas all composites yielded similar strain at break values, around 1.93%.

Figure 4(a) shows representative curves of force versus displacement obtained during short-beam testing of composites. There was some variation in curves profile for different specimens and even within the same composite family. Sample failure occurred due to inter-laminar shear, with some influence from breaking of fibers and pull-out in the results, somewhat similar to the work of Gowda et al.[11] Figure 4(b) shows that hybrid composites yielded higher short-beam strength (SBS) than pure curaua and lower than pure glass. Considering that SBS is primarily dependent on

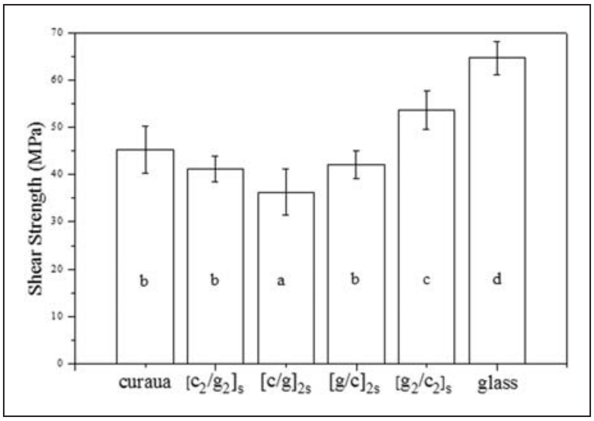

Figure 5. Mean values of Iosipescu shear strength of the laminates.

matrix properties and fiber/matrix interfacial adhesion instead of fiber properties,[14] it can be concluded that quality of the fiber/matrix adhesion or of the composite itself is poorer when curaua is used substituting glass fibers.

Classical beam theory states that shear stress distribution along the thickness of the sample is a parabolic function, symmetrical about the neutral axis (maximum shear stress), decreasing to zero on the compressive and tensile surfaces. However, the actual stress field during testing is often dominated by stress concentrations around the sample area near the loading nose. In this context, it is important to mention that premature failure before overall inter-laminar failure was noticed at the compressive surface of the $[c_2/g_2]_s$ sample, due to the influence of the weak external curaua layers.

Regarding Iosipescu in-plane shear testing, microcracks were observed in the composites during initial loading, eventually leading to overall failure, with consequent large and visible cracks. The mean shear strength values are displayed in Figure 5. Considering that in-plane shear is also dominated by matrix properties and fiber/matrix interfacial adhesion, it appears that the heterogeneity and overall quality of some hybrid composites were poorer than that of pure curaua composite. Indeed, only one of the hybrids showed higher Iosipescu strength than the pure curaua composite, which is justified by the more labor-intensive molding when using various layers instead of a single material. Also, the pure curaua was produced based on a single thick layer instead of eight thin layers and therefore resin-rich inter-laminar effects may be occurring.

Selmy et al.[15] measured in-plane shear strength of glass-fiber laminates (overall fiber content of 37% (v/v), with a combination of random and unidirectional orientations) and obtained up to 35 MPa. This is below the values reported here, but this may be a consequence of the hand lay-up process used, which can lead to greater void content in the composite.

The shear modulus of the composites could not be experimentally measured. Instead, it was obtained using equation (1) ($G_c = E_c/2(1 + \nu_c)$),[16] since all layers were composed of randomly distributed short fibers and therefore isotropic behavior is expected. The calculated shear modulus is displayed in Table 2 for comparison with the purely theoretical values. The Poisson's coefficient of every layer was considered equal to 0.33, according to Cox (1962).[16]

Mechanical analysis

The micro- and macro-mechanical analyses were carried out through three distinct procedures, with increasing degree of complexity, as detailed below:

Procedure 1: All layers were considered to have the same thickness (1/8 of the overall thickness) and the same $\%V_f$ (fiber volume content). For each layer (glass or curaua fiber), E_c (tensile) and ν_c (equal to 0.33) were used to calculate G_c, according to equation (1) ($G_c = E_c/2(1 + \nu_c)$).

Procedure 2: The actual thicknesses of each layer were measured using optical microscopy (see Figure 1) but considering an invariable $\%V_f$ in each layer. For each layer of material (glass or curaua fiber), E_c (tensile) and ν_c (equal to 0.33) were used to calculate G_c, as above.

Procedure 3: In this case, the actual thickness and the actual $\%V_f$ of each layer were used, as follows:

- By knowing the mass of fiber used, fiber density, layer thickness and area of the composite, the $\%V_f$ of the pure curaua and of the pure glass composites were calculated. This revealed that the actual fiber volume fraction was 30% of curaua and 37% of glass, respectively.
- Using the Tsai-Pagano equation[17] for the pure glass composite (considering $l_f = 50\,mm$, $D_f = 14\,\mu m$, $E_m = 2.8\,GPa$ and the measured E_c), effective E_f of the glass fiber was calculated as 65 GPa, which is in the same range of the literature.[18]
- Using the Tsai-Pagano equation[17] for the pure curaua composite (considering $l_f = 50\,mm$, $D_f = 41\,\mu m$, $E_m = 2.8\,GPa$ and the measured E_c), effective E_f of the curaua fiber was calculated as 8.8 GPa, which is in the same range of the literature.[19]
- Then, $\%V_f$, E_c and G_c were calculated for each layer taking into account its actual thickness measured by

Table 2. Comparison between measured and calculated elastic and shear modulus.

	Experimental		Procedure 1 – constant thickness		Procedure 2 – actual thickness (theoretical $\%V_f$)		Procedure 3 – actual thickness (and actual $\%V_f$)	
	E (MPa)	*G* (MPa)	*E* (MPa)	*G* (MPa)	*E* (MPa)	*G* (MPa)	*E* (MPa)	*G* (MPa)
$[c_2/g_2]_s$	6299	2368	7957	2864	7457	2693	8444	3041
$[c/g]_{2s}$	5500	2068			6184	2265	6699	2450
$[g/c]_{2s}$	6971	2621			6273	2288	6897	2508
$[g_2/c_2]_s$	6455	2427			7420	2680	8398	3024

Obs. Pure curaua: E = 4230 MPa; G = 1590 MPa; Pure glass: E = 9879 MPa; G = 3714 MPa, where shear modulus was obtained using $G = E/2(1 + \nu)$.

Table 3. Deviation of each procedure in relation to the experimental values.

	Procedure 1 – Constant thickness		Procedure 2 – actual thickness (theoretical $\%V_f$)		Procedure 3 – actual thickness (and actual $\%V_f$)	
	E (MPa)	*G* (MPa)	*E* (MPa)	*G* (MPa)	*E* (MPa)	*G* (MPa)
$[c_2/g_2]_s$	26%	20%	18%	13%	34%	28%
$[c/g]_{2s}$	44%	38%	12%	9%	22%	18%
$[g/c]_{2s}$	14%	9%	−10%	−13%	−1%	−4%
$[g_2/c_2]_s$	23%	18%	15%	10%	30%	25%
Mean absolute deviation	24%		12.5%		20.25%	

optical microscopy, and then ν_c was estimated. It was found that the mean actual $\%V_f$ of the curaua layers was 34% and of the glass layers was 39%.

- In the laminator software, E_c, G_c, ν_c, the thickness of each layer and the lay-up of a particular laminate were used as input to estimate the engineering elastic properties of that laminate.

Table 2 shows the results obtained using these three procedures for the hybrid laminates. Table 3 shows the deviation of each procedure in relation to the experimental values to facilitate the comparison. It can be seen that even though all procedures yielded a particular set of data, they showed deviation 12.5−24% in relation to the experimental values. Interestingly, for Procedure II (actual thickness) the calculated values showed less deviation what means that the properties composite are influenced by manufacturing process and the curaua fiber properties which are not uniform when compared to glass fiber. In addition, it can be said even though it is not common to use these software to analyze composites with randomly arranged fibers, this can be carried out applying micromechanics as an auxiliary tool. Table 3 also suggests poor manufacturing and/or testing of the $[c/g]_{2s}$ laminate since deviation was invariably high.

Conclusions

From the combination of curaua and glass fibers layers originated a hybrid laminate of generally intermediate properties. Depending on the type of mechanical loading and the stacking sequence, some hybrids showed properties close to the pure glass fiber composites. Hybrid composites with glass layers at surfaces showed the best results regarding flexural strength and modulus, SBS and Iosipescu shear strength. On the other hand, tensile strength and modulus were less sensitive to the laminate lay-up.

Mechanical analysis was carried out using different procedures, with increasing degree of complexity, which included the use of corrected thicknesses and fiber volume content for each layer. The difference between the deviations of procedures could be credited to experimental molding and testing difficulties and also to the variable mechanical properties of the curaua fiber, which is inherent to any vegetable fiber.

Funding

This research received no specific grant from any funding agency in the public, commercial, or not-for-profit sectors.

Acknowledgments

The authors would like to thank CAPES, CNPq and FAPERGS for the financial support.

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