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SISAL-GLASS HYBRID COMPOSITES REINFORCED WITH SILICA MICROPARTICLES

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Abstract

In recent years, studies of composite materials have focused on the use of natural fibres as an alternative to synthetic ones. Their attractive mechanical properties, sustainability, low cost and low weight have leveraged research in this area, with the potential of a variety of applications in the engineering field. In this paper, a Full Factorial Design (2¹6¹) experiment was performed to identify the effect of silica microparticle inclusions and the stacking sequence of glass fibre cross-ply fabric and short sisal fibre layers on the apparent density, tensile and flexural strength and modulus of hybrid epoxy composites. In general, the hybrid composites with higher number of glass fibre layers achieve higher values of tensile and flexural strength (348 MPa and 663.28 MPa), tensile and flexural modulus (22 GPa and 2.50 GPa) and higher apparent density (2.02 g/cm³). It is, however, noteworthy that the incorporation of silica particles improves the mechanical performance of composites containing larger amounts of sisal fibres.

Keywords: hybrid composite; natural fibre; glass fibre; silica particles; mechanical properties; design of experiment (DoE).

INTRODUCTION

In recent decades, the growing demand for lightweight materials, particularly important in aerospace, wind and automotive applications, has boosted the development of synthetic or natural fibre-reinforced composites [1-4]. Despite the better mechanical reinforcement provided by synthetic relative to natural fibres, synthetic fibres present higher manufacturing
costs and pose important environmental issues such as renewability and disposal. On the other hand, natural fibres are highly hygroscopic due to their hydrophilic character, which is a durability limiting factor, and also present large variation of their physical properties owing to the maturation level of the plant, soil and humidity conditions during growth and the part of the harvested specimens from which fibres are extracted (leaf or stem), processing and production methods [5]. Despite these drawbacks, natural fibres may play an important role in the development of sustainable materials owing to their low density, low cost, greater availability and biodegradability [5-6].

In an effort to reduce the use of synthetic fibres and yet maintain attractive mechanical properties, recent research has focused on hybrid reinforced composites using both synthetic and natural fibres [7-9], which provide a significant increase in the mechanical properties and reduction of moisture absorption [10,11].

Among the various types of natural fibres, sisal is the most commercially used due to its good mechanical properties combined with ease of production. The annual worldwide production of sisal fibres is approximately 600 million tons, of which about 200 million tons are produced in Brazil [12]. Regarding synthetic fibres, glass fibres are the most commonly used owing to their high strength and stiffness, low density, low cost, high ductility and practically negligible water absorption. Hybridization of sisal and glass fibres has been proposed in the literature, especially to reduce final costs of composite materials with sustainable feature [13,14].

Studies reveal effective improvement of the tensile and flexural strength of sisal-glass fibre-reinforced composites with increase of the glass fibre weight fraction [15-17]. Analysing the stacking sequence of the hybridization of sisal and glass fibres, the best flexural properties were found using glass fibres in the outer layers and sisal in the composite core [18].

The mechanical performance of the hybrid composites varies according to the type of reinforcement, stacking sequence of fibre layers, fibre orientation, fibre-matrix compatibility, fibre weight fraction and manufacturing process [19, 20].

Another way to improve the properties of the hybrid composites is to use nano or micro scale particulate reinforcement as an alternative to increase the stiffness of the matrix phase, in addition to promoting an interlocking effect between laminates [21-23]. According to Silva et al. [24], the particles also intercept the propagation of cracks, retarding their growth and preventing catastrophic failure. The type of fibres and particles and their arrangement affect the compatibility and interaction of these elements with the polymeric matrix phase, being determinants of the final mechanical properties of the composite material [25].
The novelty of this paper is the analysis of three-phase composite composed of sisal fibres, glass fibres and silica particles. The natural fibres were replaced with glass fibres from bottom to the top of the beam side in order to obtain a greater tensile stress of the synthetic fibres under three-point bending loads. Silica particles have also been incorporated to stiffen the matrix phase and enhance the mechanical performance of the composites, especially when short and random sisal fibres are used. A full factorial design was performed to identify the effects of fibre hybridization and silica particle inclusions on density, tensile and flexural properties.

MATERIALS AND METHODS

Materials

The hybrid composites were fabricated with five layers of sisal/glass fibres via compression moulding. Sisal short fibres were supplied by Sisalsul Company (Brazil). A cross-ply glass fibre fabric (200 g/cm²) was supplied by Texiglass-Brazil. Renlam M epoxy resin and HY Aradur 951 hardener were supplied by Hunstman Brazil. The silica micro particles were sourced by Omega Mining Company (Brazil).

Design of experiment

A Full Factorial Design (2¹6¹) was established to identify the effect of silica inclusions (0 and 5 wt%) and five layer stacking sequence, combining short sisal fibres (S) and glass fibre fabric (G), on the physical and mechanical properties of hybrid composites (see Table 1). The responses investigated in the experiment were apparent density, mechanical strength and modulus under tensile and flexural loads. Fifteen hybrid composite specimens (5 for density, 5 for tensile, 5 for bending tests) were fabricated for each of the 12 experimental conditions. Two replicates were considered, running a total of 360 specimens. The replicate consisted of repetition of the experimental condition to give indication of the variability of the individual response. A randomization procedure was adopted during sample manufacturing and experimental testing, avoiding uncontrolled factors from affecting responses [26]. Minitab 17 software was used to perform statistical analysis based on Design of Experiment (DoE) and Analysis of Variance (ANOVA).

Table 1. Full Factorial Design (2¹6¹).

<table>
<thead>
<tr>
<th>Conditions</th>
<th>Layers Sisal/Glass</th>
<th>Silica inclusion (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>5S</td>
<td>0</td>
</tr>
</tbody>
</table>
Manufacturing process

The epoxy polymer was prepared by mixing the resin and hardener at a ratio of 10:1 for 5 minutes. Silica microparticles (classified by sieving in monomodal size of 0.037 mm) were subsequently added to the epoxy polymer and hand-mixed for 5 minutes at room temperature (~21°C). The fibre volume fraction (20%) was kept constant, being determined based on preliminary tests, in order to avoid resin leakage after compaction.

The composites were fabricated using a wooden mould (200 x 200 mm) under cold uniaxial pressure of 0.75MPa for 12 hours. After 7 days of curing, the plates were cut according to ASTM standards [27,28] (Figure 1). The sisal fibres are replaced by glass fibres from the bottom to the top surfaces, as shown in Figure 2. This experimental design was proposed since glass fibres support higher tensile stress compared to sisal fibres, being at the bottom beam side when subjected to bending loads. Each layer of glass fibre fabric weighed 7.0 g, while each layer of random sisal fibre weighed 3.4 g.

<table>
<thead>
<tr>
<th>Layer</th>
<th>Fibre Type</th>
<th>Volume Fraction</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>4S/1G</td>
<td>0</td>
</tr>
<tr>
<td>2</td>
<td>3S/2G</td>
<td>0</td>
</tr>
<tr>
<td>3</td>
<td>2S/3G</td>
<td>0</td>
</tr>
<tr>
<td>4</td>
<td>1S/4G</td>
<td>0</td>
</tr>
<tr>
<td>5</td>
<td>5G</td>
<td>0</td>
</tr>
<tr>
<td>6</td>
<td>5S</td>
<td>5</td>
</tr>
<tr>
<td>7</td>
<td>4S/1G</td>
<td>5</td>
</tr>
<tr>
<td>8</td>
<td>3S/2G</td>
<td>5</td>
</tr>
<tr>
<td>9</td>
<td>2S/3G</td>
<td>5</td>
</tr>
<tr>
<td>10</td>
<td>1S/4G</td>
<td>5</td>
</tr>
<tr>
<td>11</td>
<td>5G</td>
<td>5</td>
</tr>
<tr>
<td>12</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Figure 1. Manufacturing process of the hybrid composites: (a) lamination, (b) wooden mould, (c) cold pressing, (d) material after compaction.
Testing

Tensile and flexural tests were carried out based on the recommendations of ASTM D3039-14 [27] and ASTM D790-15 [28], respectively. A Zwick /Roell Z020 test machine with a 20 kN load cell was used to perform both tests, conducted at a crosshead speed of 2 mm/min. The apparent density was determined based on Archimedes principle using a precision balance (0.001g) and distilled water, as recommended by ASTM D792 [29].

RESULTS

Table 2 shows the analysis of variance (ANOVA) for the physical and mechanical responses investigated. P-values less or equal to 0.05 indicate the individual factor or the interaction that significantly affects the response. When one or more interaction effects of superior order are significant, the interacting factors should be considered together [30]. The underlined P-values relate to factors or interaction of factors that significantly affected the responses. P-values in bold correspond to main effects or interaction that will be analysed via effect plots. Values of R²-adj close to 100% indicate that the model has higher predictive capacity [30]. To validate the ANOVA, a normality test (Anderson-Darling) was conducted based on the residual analysis for each response-variable. In this case, P-values greater than 0.05 indicate that the data follow a normal distribution, as shown in Table 2.

Table 2. Analysis of Variance (ANOVA)

<table>
<thead>
<tr>
<th>Experimental Factors</th>
<th>Apparent density (g/cm³)</th>
<th>Tensile Strength (MPa)</th>
<th>Tensile Modulus (GPa)</th>
<th>Flexural Strength (MPa)</th>
<th>Flexural Modulus (GPa)</th>
</tr>
</thead>
</table>

Figure 2. Stacking sequence of the fibres.
### Table 1: Main Factors and Interactions

<table>
<thead>
<tr>
<th>Main Factors</th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Silica Inclusion (SI)</td>
<td>0.442</td>
<td>0.000</td>
<td>0.951</td>
<td>0.000</td>
<td>0.115</td>
</tr>
<tr>
<td>Glass fibre inclusion (GFI)</td>
<td><strong>0.000</strong></td>
<td><strong>0.000</strong></td>
<td><strong>0.000</strong></td>
<td>0.000</td>
<td>0.000</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Interactions</th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>SI x GFI</td>
<td>0.13</td>
<td><strong>0.000</strong></td>
<td>0.389</td>
<td><strong>0.000</strong></td>
<td><strong>0.029</strong></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>R² – adj</th>
<th>99.13%</th>
<th>98.63%</th>
<th>98.49%</th>
<th>98.68%</th>
<th>98.28%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Anderson-Darling (P-value ≥ 0.05)</td>
<td>0.133</td>
<td>0.132</td>
<td>0.989</td>
<td>0.313</td>
<td>0.057</td>
</tr>
</tbody>
</table>

### Apparent Density

The apparent density of the composites ranges from 1.15 g/cm³ to 2.02 g/cm³. The apparent density of the composites is not affected by silica particles in the range up to 5 wt% used (P-value of 0.442, see Table 2). The glass fibres promote an increase in mean apparent density, as shown in Figure 3, due to their higher density relative to the sisal fibres.

![Figure 3. Main effect of glass fibre inclusions for mean apparent density of the composites](image)

### Tensile Strength

Figure 4 shows the interaction effect plot for the mean tensile strength of the composites. The presence of silica reduces the tensile strength. This fact can be attributed to the inefficiency of the particle-reinforced matrix under tensile loads, and possible reduction of interlaminar adhesiveness due to silica incorporation. The tensile strength values are increased
when the amount of glass fibre layers was increased. It is noteworthy that the replacement of a single sisal fibre layer by glass (4S1G) without silica inclusions substantially increases the tensile strength by 84% when compared to composites made with only natural fibres (5S). In contrast, significant reduction is achieved when a single glass fibre layer was replaced by sisal fibres (1S4G), especially when no silica particles were added. Similar findings are in fact observed for both tensile and flexural mechanical properties.

![Figure 4. Interaction effect plot for mean tensile strength.](image)

**Tensile Modulus**

Modulus of elasticity ranges from 3.50 GPa to 22.00 GPa. Figure 5 shows the main effect plot for glass fibre inclusion. The treatments containing larger numbers of glass fibre layers result in higher tensile modulus, leading to a substantial increase of 477% from 5S to 5G. This behaviour is expected, considering that glass fibres exhibit superior mechanical performance compared to natural random fibres. The silica inclusions do not affect the tensile stiffness of the composites, as shown in Table 2.

![Tensile Modulus Graph](image)
Figure 5. Main effect plot for mean modulus of elasticity.

Flexural Strength

Flexural strength ranges from 51.36 MPa to 663.28 MPa. Figure 6 shows the interaction effect plot for mean flexural strength. The inclusion of particles reduces the strength, except for composites made with 100% sisal fibres. The presence of silica particles substantially reduces (~30%) the flexural strength of glass fibre reinforced composites (5G). Significant reductions are also observed when glass fibres were replaced by one or more layers of sisal fibres. It is noteworthy that 2S3G composites without particles reveal an increase of 407% when compared to composites reinforced with sisal fibres (5S). When only one layer of sisal fibres without silica inclusion (1S4G/0% silica) was used, a strength reduction of 25% is observed. This behaviour can be attributed to the reduction of the area moment of inertia when a thinner layer of sisal is combined with glass fibres. This effect is not evident when the silica particles are in the system, avoiding the reduction in laminate thickness.
Figure 6. Interaction effect plot for mean flexural strength.

Figure 7 shows the mechanical behaviour of 3S2G condition with and without silica inclusions under three-point bending test. The incorporation of silica particles leads to increased modulus and reduced strength. The displacement at maximum load is substantially decreased (~190%) by the presence of silica, indicating a reduction in ductility. The other hybrid composites achieve similar behaviour with the presence of silica particles.

Figure 7. Flexural behaviour of 3S2G composite with and without silica particles.

**Flexural Modulus**

The flexural modulus varies between 1.61 GPa and 2.50 GPa. Figure 8 shows the interaction effect plot for mean flexural modulus. The highest flexural modulus is achieved when the composites were fabricated using five layers of glass fibre (5G) without silica.
particles. Silica inclusions promote a positive effect only when a greater amount of sisal fibres is considered (5S and 4S1G). This behaviour suggests that the particles contribute more effectively to increase the matrix phase stiffness rather than the interlocking effect. A system composed of random short fibres is largely dominated by the properties of the matrix phase. In addition, the use of short random sisal fibres can lead to internal regions without fibre reinforcement. The particles may, consequently, provide a mutually beneficial effect, increasing matrix phase stiffness and filling voids. A significant reduction (~70%) in flexural modulus is evidenced when only a single layer of glass fibres (1S4G) was replaced by sisal fibres. It is noteworthy, however, that a significant increase of approximately 324% in stiffness is achieved between 5S and 2S3G composites, the latter reinforced with 3 glass fibre layers.

![Interaction effect plot for mean flexural modulus.](image)

**Fracture Surface Analysis**

A microstructural analysis of the fractured cross-section of composites subject to flexural tests was performed using Hitachi T-3000 scanning electron microscope. Figure 9 shows the fracture surface of the glass fibre composites after a bending test. Extensive regions of fibre pull-out are observed in Fig 9a. A typical mode of brittle fibre fracture for glass fibre composites is shown in Fig. 9b.
Figure 9. Fracture surface of the flexural test in glass fibre composites (5G).

Figure 10 shows the fracture section of composites reinforced with sisal fibres (5S). The random distribution of fibres of different sizes is observed. Fibre pull-out is widely evidenced due to the weak adhesion between the natural fibre and the matrix phase (Fig. 10a). Micro pores are also observed, being attributed to the random and hydrophilic features of the natural fibres that hinder the homogenization of the system (Fig. 10b).

Figure 10. Fracture surface of the flexural test in sisal fibre composites (5S).

The delamination effect is more present in the hybrid composites due mainly to distinct topography and fibre orientation between the natural and the synthetic fibres (Figure 11a). The structure of the sisal mat comprises short and random fibres which provide a significant amount of fibre pull-out (Figure 10). In contrast, glass fibre fabric has a uniform cross-ply distribution and increased stiffness leading to a brittle fracture, as shown in Figure 11b. This variation along with the difference in the properties of each material lead to combined behaviour at the time of fracture. Figure 11 shows the fractured section of 3S2G composite, revealing sisal the fibre pull-out characteristic and the brittleness characteristic of the glass fibres.
CONCLUSIONS

Hybridization of sisal/glass fibres and silica microparticles is assessed in this work. The main conclusions are described below:

- Replacement of sisal fibres by glass fibres leads to significant effects on tensile strength, flexural strength, tensile modulus, flexural modulus and apparent density of the composites. Hybrid composites with higher number of glass fibre layers achieve higher mean values of tensile and flexural strength (348 MPa and 663.28 MPa, respectively), tensile and flexural modulus (22 GPa and 2.5 GPa, respectively) and apparent density (2.02 g/cm³).

- It is noteworthy that the replacement of a single sisal fibre layer by glass (4S1G) substantially increases the tensile and flexural strength and modulus (approx. 80%) when compared to composites made with only natural fibres (5S), with a small increase in density. It is, therefore, possible to combine sustainability and cost reduction and yet achieve a remarkable enhancement in the flexural and tensile mechanical properties.

- Silica microparticles significantly affect the flexural strength, tensile modulus and flexural modulus. The presence of silica particles improves the mechanical performance of the composites, especially when considering a larger amount of sisal fibres, i.e. 5S and 4S1G conditions.

- Fracture analysis of composites consisting of sisal (5S) and glass fibres (5G) subjected to flexural loads reveal the sisal fibre pull-out and the typical brittle mode of glass fibres.
• Delamination effects under three-point bending were more evident in the hybrid structure.

• Finally, 2S3G composites without silica particles achieve moderate strength and stiffness, being a promising alternative for secondary structural parts in projects with sustainable requirements.

ACKNOWLEDGMENTS

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