Materials Research. 2023; 26:e20230188
DOI: https://doi.org/10.1590/1980-5373-MR-2023-0188

Physical and Mechanical Evaluation of Sisal/Glass Fiber Hybrid Polyester Composites Obtained by the Vacuum Infusion Process (VARTM)

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Received: March 30, 2023; Revised: July 13, 2023; Accepted: August 17, 2023

The research interest in developing alternative solutions to replace conventional synthetic fibers with natural fibers in composites is due to the several possible benefits of using these fibers, such as low cost, low density, high specific strength and high availability. The objective of this study was to produce hybrid composites from a commercial ortho-terephthalic unsaturated polyester resin obtained from the recycled PET (polyethylene terephthalate) glycolysis reinforced with sisal/glass fibers obtained by the vacuum infusion process. The composites were produced with 3 layers of fibers, the volume of each layer was fixed, and the amount of resin used was the same. For the results, the relative density decreased up to 23% and the moisture absorption reached up to 10.41%, being possible to observe the influence of the layer arrangement on the results. From the mechanical tests of tensile, flexural and impact strength it was observed that the properties decreased as sisal fibers were added, except in the results of maximum deflection in the flexural test of formulations. From the SEM (scanning electron microscope) analyses showed some defects in the direction of the thickness of all formulations produced, possibly from the process conditions adopted and the moisture present in the sisal fibers, which negatively affected the results of mechanical properties and moisture absorption. According to the results of this study, it was possible to obtain some proposed formulations of hybrid composites, such as the (F1), composed of glass, sisal and glass layers (|GSG), which reached properties close or equivalent to those of the reference composite (F0), composed of glass, glass and glass layers (|GGG) and which showed to be potential alternatives to replace conventional glass fiber composites.

Keywords: *Vacuum infusion, hybrid composites, unsaturated polyester resin, glass fiber, sisal fiber.*

1. Introduction

Over the past few decades, research interest in developing alternative solutions to replace conventional synthetic fibers present in composites with natural fibers has been growing due to the high demand for ecologically sustainable materials, the desire to reduce the impact on the environment, and the ambition to reduce the costs of polymer composites[1-5].

Natural fibers in composites have several advantages compared to synthetic ones, such as low cost, low density, high specific strength, biodegradability, and abundant availability. Currently, there are several natural fibers that are used in polymeric composites, such as sisal, jute, ramie, coconut and loofah fibers[1,3,6,7]. Among these, sisal fiber stands out for its physical and chemical characteristics[3,8], besides its availability in the Brazilian scenario, due to the fact that of the annual world production of 600 million tons, 200 million tons are produced in Brazil[9,10].

Some disadvantages associated with natural fibers hinder their use, such as the difficulty in obtaining homogeneous fibers, low dimensional stability, the difficulty of interaction between the fiber and the polymeric matrix in some cases, besides the high sensitivity to variations in environmental conditions, temperature and humidity[1,4,6]. Thus, some physical and chemical treatments are proposed in order to improve the interaction of fiber with the matrix, maintaining its mechanical and thermal properties[1,6,9,11-13].

In addition, the combination of natural fibers with synthetic fibers is an excellent option to mitigate the impacts of high moisture absorption and lower mechanical properties of composites with natural fibers[1,4,11].

Among the types of polymeric matrices used in composites, unsaturated polyester resin has been used in large proportion due to its characteristics, such as low viscosity, which favors the impregnation of the reinforcement; good interaction with fibers; low processing energy; low cost; high mechanical strength, and chemical and thermal resistance[14,15].

Another relevant aspect for obtaining composites of thermosetting polymeric matrix refers to the types of processing of these materials, highlighting the vacuum resin infusion technique (VARTM) because it is a closed process, which aims to reduce the emission of volatile organic compounds (VOC), makes it possible to obtain high fiber contents, has good repeatability and ease of scaling production and has advantages over the open-mold hand lay-up processing[16].

The objective of the present study was to develop and characterize in terms of physical and mechanical properties the polymer matrix hybrid composites. These composites were formed by unsaturated ortho-terephthalic polyester resin obtained from the glycolysis of recycled PET, reinforced with sisal/glass fiber, due to the properties and

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availability of the sisal fiber and glass fiber in the Brazilian market, obtained from the resin vacuum infusion process. The following analyses were performed on the composites: relative density, moisture absorption, tensile test, flexural test, impact test and observation of the interfaces through scanning electron microscope.

2. Experimental Procedure

2.1. Materials

The sisal fibers in chopped strand mat form were supplied by the company Grupo Hamilton Rios, Bahia, Brazil, with a basis weight of 1030g/m^2, without chemical or physical treatments and were used as received (Figure 1a). The glass fibers type M821B, recommended for vacuum processes, in the form of chopped strand mat was supplied by the company Owens Corning, São Paulo, Brazil, with a basis weight of 450g/m^2 and were used as received (Figure 1b). The unsaturated polyester resin (UPR) CRISTALAN INF-512, of the ortho-therephthalic type, with an average viscosity of 175mPa.s, monomer content of 44% and average gel time of 50 min with 1% peroxide, was supplied by the company Novapol Plásticos Ltda, Espírito Santo, Brazil. The methyl-ethyl-ketone peroxide (MEKP) Butanox M50 manufactured by the company Nouryon was used as a curing initiator.

Table 1 shows data available in the literature[6] of the characteristics and mechanical properties of sisal fiber and glass fiber.

2.2. Preparation of hybrid composites

The hybrid composites were produced using the vacuum resin infusion technique (VARTM) and a glass plate used as a rigid mold. The fiber mats, in the dimension of 45x30cm, were positioned on the glass plate, followed by the release agent fabric (peel ply) and the flow mesh. To facilitate the resin flow and establish a consistent vacuum throughout the mold evenly, two spiral tubes were positioned over the edges of the top layer, one tube being used as the resin inlet and the other used as the outlet, connected to the vacuum pump. The system was closed with the vacuum bag and sealing tape (Figure 2). The resin entered the system through one of the tubes and traveled to the other tube, so the resin feed was stopped, and the vacuum kept by 1 and a half hour. After that, the composite plate was demolded 24h later.

2.3. Hybrid composites formulation

The composites were produced with 3 fiber layers each and the compositions varied according to the amount of sisal and glass fiber mats, as well as different positions of the fiber layers. The volume of each layer was fixed and for that an equivalence of 1 sisal fiber mat (1030g/m^2) to 4 glass fiber mats (450g/m^2) was adopted due to the different basis weights and densities of the fibers in order to obtain the same thickness among the formulations. In all composites, the amount of resin used was 1500g. The formulations are presented in Table 2.

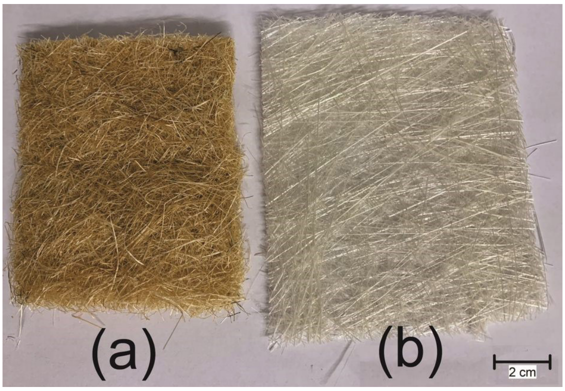

Figure 1. (a) Sisal fiber; (b) Glass fiber.

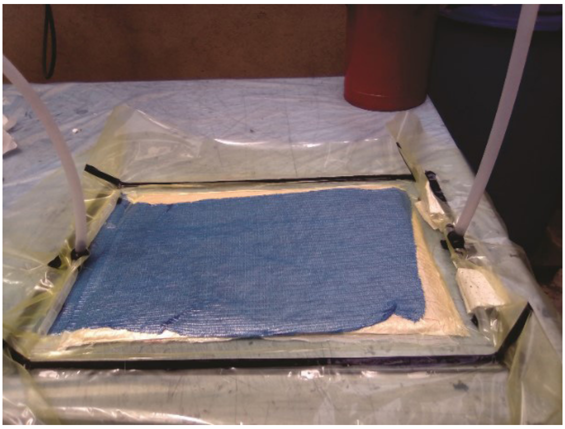

Figure 2. Infusion system used to manufacture the composites with the resin flow from left to right.

Table 1. Mechanical properties of sisal and glass fiber[6].

Properties	Glass fiber (E-glass)	Sisal fiber
Density (g/cm^3)	2.5	1.3 – 1.5
Tensile strength (MPa)	2000 - 3000	507 – 855
Young's modulus (GPa)	70	9.4 – 28
Specific tensile strength (MPa/ g.cm^{-3})	800 – 1400	362 – 610
Specific Young modulus (GPa/ g.cm^{-3})	29	6.7 – 20
Failure strain (%)	2.5	2.0 – 2.5

Table 2. Formulations of the composites.

Comp.	Bottom surface	Layer 1	Layer 2	Layer 3	Top surface
F0 (|GGG)	Glass plate (|)	••••	••••	••••	Flexible mold
F1 (|GSG)		••••	×	••••	
F2 (|SGG)		×	••••	••••	
F3 (|GSS)		••••	×	×	
F4 (|SGS)		×	••••	×	
F5 (|SSS)		×	×	×	

(×) Sisal CSM* - (•) Glass CSM*; *Chopped strand mat.

2.4. Characterization

2.4.1. Relative density

The determination of the relative density of the composites was carried out according to ASTM D 792 at temperature of 25 ºC. Three specimens were tested for each formulation. The dimensions of the specimens were 50 mm x 50 mm x 9.5 mm.

2.4.2. Moisture absorption test

The moisture absorption test was carried out according to an adaptation of ASTM D 570 at temperature of 25 ºC. Three specimens were tested for each formulation. The dimensions of the specimens were 25 mm x 25 mm x 9.5 mm. The specimens were immersed in a water bath at a temperature of 25 ± 1 ºC and the moisture absorption values were recorded after 1, 3, 7, 10, 14, 21 and 28 days.

2.4.3. Tensile test

The tensile test was carried out according to ASTM D 3039 at temperature of 25 ºC in an INSTRON model 3369 machine with a load cell with maximum capacity of 50 kN, distance between grips of 175 mm, and crosshead speed of 2 mm/min. Five specimens were tested for each formulation. The dimensions of the specimens were 250 mm x 25 mm x 9.5 mm.

2.4.4. Flexural (3-point bend) test

The flexural test was carried out according to ASTM D 790 at temperature of 25 ºC in an INSTRON model 3369 machine with a load cell with maximum capacity of 20 kN. Five specimens were tested for each formulation. The dimensions of the specimens were 200 mm x 25 mm x 9.5 mm. The distance between supports (span) was 144 mm and test speed was 3.84 mm/min.

2.4.5. Impact test

The IZOD impact test was carried out according to ASTM D 256 at temperature of 25°C, manually operated on a CEAST 9000 with a maximum capacity of 50J. Five specimens were tested for each formulation. The dimensions of the specimens were 80 mm x 4 mm x 9.5 mm, with a notch depth of 2.5 mm and an opening radius of 45°.

2.4.6. Scanning electron microscopy (SEM)

The cross section of the specimens of the different composites was observed with a JEOL model JSM-IT200 scanning electron microscope at a voltage of 18 kV. All specimens were previously metallized with a thin gold layer on the surface with the Quorum Q150R vacuum metallizer.

2.4.7. Specific properties

The specific properties for the tensile, flexural and impact test were defined as the experimental value of the property divided by the density of the material (Table 3).

3. Results and Discussion

3.1. Relative density

Table 3 presents the results of relative densities of the composites. According to the results, it can be observed that the density decreased with the increment of the sisal fiber layers replacing the glass fiber layers. This behavior can be attributed to the differences in densities between the fibers[4], which can be visualized in Table 1. In the work by Ornaghi et al.[17] this behavior is also observed in the density results of hybrid composites.

However, when analyzing the graph presented in Figure 3, it is noted that considering the deviations, the arrangement of the layers in the hybrid composites did not affect significantly the densities since the ranges of values overlap. If considering the formulation F0 and F5 it is possible to observe the differences and it can be related to the values of each fiber separately (Table 1).

3.2. Moisture absorption

Table 4 presents the results obtained for moisture absorption of the composites.

It is observed that there was a high rate of moisture absorption during the first days for the formulations containing sisal fiber in the composition, with an asymptotic tendency to reach a saturation level in the course of the test after the 28-day period (Figure 4).

Table 3. Relative density of the composites.

Composites	Relative density (g/cm^3)
|GGG (F0)	1.47 ± 0.08
|GSG (F1)	1.29 ± 0.08
|SGG (F2)	1.37 ± 0.07
|GSS (F3)	1.24 ± 0.06
|SGS (F4)	1.25 ± 0.05
|SSS (F5)	1.14 ± 0.07

Table 4. Water absorption of the composites.

Days	|GGG (F0) (%)	|GSG (F1) (%)	|SGG (F2) (%)	|GSS (F3) (%)	|SGS (F4) (%)	|SSS (F5) (%)
Initial	0	0	0	0	0	0
1	0.08 ± 0.01	0.86 ± 0.13	1.05 ± 0.13	1.43 ± 0.20	1.77 ± 0.15	2.86 ± 0.39
2	0.20 ± 0.03	1.15 ± 0.14	1.38 ± 0.14	1.91 ± 0.26	2.35 ± 0.24	3.84 ± 0.55
3	0.21 ± 0.00	1.54 ± 0.21	1.78 ± 0.20	2.44 ± 0.32	2.88 ± 0.30	4.70 ± 0.68
4	0.25 ± 0.04	1.77 ± 0.26	1.94 ± 0.27	2.74 ± 0.39	3.31 ± 0.37	5.27 ± 0.81
7	0.38 ± 0.06	2.27 ± 0.33	2.59 ± 0.35	3.46 ± 0.54	4.18 ± 0.48	6.71 ± 1.00
14	0.54 ± 0.09	3.09 ± 0.47	3.29 ± 0.47	4.59 ± 0.73	5.32 ± 0.66	8.65 ± 1.36
21	0.57 ± 0.10	3.61 ± 0.57	3.63 ± 0.58	5.24 ± 0.87	5.92 ± 0.79	9.70 ± 1.61
28	0.66 ± 0.10	4.03 ± 0.63	3.89 ± 0.61	5.68 ± 0.93	6.35 ± 0.89	10.41 ± 1.80

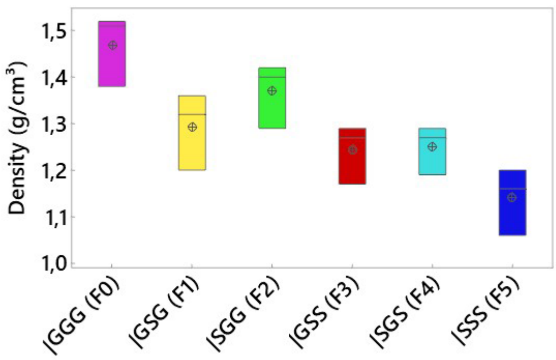

Figure 3. Density of the composites.

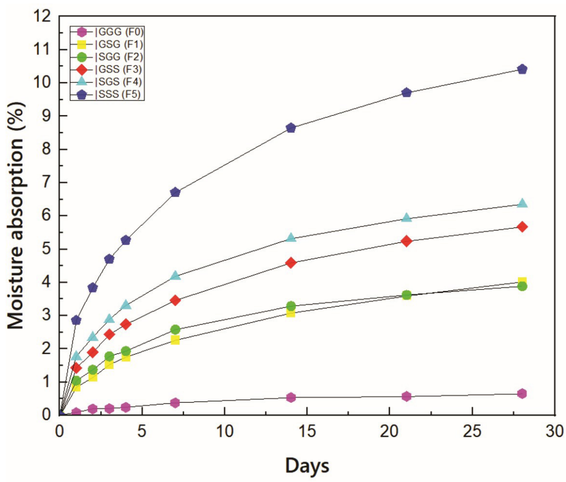

Figure 4. Moisture absorption of the composites.

Figure 4 shows that the maximum moisture absorption of the reference formulation (F0) remained at 0.66%, indicating a tendency to reach saturation faster than the other formulations. For formulations F1 and F2, which contain 1 layer of sisal fiber, regardless of the positioning of the fibers, the results showed no significant differences, reaching saturation at approximately 4%. Moreover, these similar results indicate that, in this composition with 2 layers of glass fibers and 1 of sisal, the positioning of the fibers had no significant influence on moisture absorption. Ford et al.[18] studied the moisture absorption of unsaturated polyester hybrid composites containing 40% sisal fibers and 60% glass fibers in number of layers, a composition like the present study, and presented a saturation of 5%, corroborating the result found here.

Between the formulations that contain two layers of sisal fiber and one layer of glass, i.e. formulations F3 and F4, it can be seen in Figure 4 that formulation F4 presented a tendency of higher absorption than formulation F3, reaching saturation values of 5.7% and 6.3%, respectively. It is believed that this occurred due to the larger contact area of sisal fibers with water, since in this formulation the sisal fibers are at the ends of the composite. In formulation F5, which contains only sisal fiber, it is observed a tendency to reach saturation at approximately 10%.

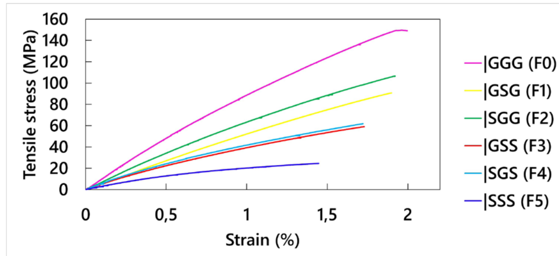

Figure 5. Tensile stress-strain curves for different composite recipes presented on Table 2.

In general, Figure 4 also shows a tendency to increase moisture absorption as natural fibers are added to the composite. This can be justified by their hygroscopicity, since they are mainly composed of cellulose and this polysaccharide has a high number of hydroxyls along the chain and it has affinity with water. Also, the sisal fibers had not undergone any kind of previous treatment[19]. This tendency also was observed in the study of Abd El-baky and Kamel[20], but in their work, the results presented some differences depending on the position of each layer for the same composition.This behavior was associated with the void content and to the hand lay-up process.

3.3. Tensile test

Table 5 presents the results of the tensile test, the following properties were determined: Young's modulus, maximum strain, tensile strength, specific Young's modulus and specific tensile strength. Figure 5 presents the tensile stress-strain curves for the tensile test considering an average curve for each formulation.

From Table 5 and Figure 5, it can be observed a tendency of decrease in Young's modulus and tensile strength as sisal fibers are added to replace the glass fiber in the composite[4,9,11,21]. There was also a tendency of decrease for the results obtained for specific modulus and specific tensile strength, but with less variation between the standard and the hybrid composites. This result refers to the characteristic of lower stiffness and lower strength of sisal fiber compared to glass fiber. As for the maximum strain results of the hybrid composites, the values did not show differences.

Figure 6 graphically presents the results of the tensile test. From Figure 6a and b, in the results of Young's modulus and tensile strength, it is possible to note that there were no significant differences between the pairs that have the same layers in different arrangements, formulations F1; F2 and F3; F4, indicating that the arrangement of the layers was not significant for the results, including for the specific Young's modulus and specific tensile strength, which did not vary with the different arrangements, either. This fact was also observed in the study by Abd El-baky[22]. This behavior is related to the effort applied in the tensile direction to both extremities, so the arrangement of the layers did not have an influence.

Table 6 shows the results of the tensile test arranged in percentage of retention considering the reference formulation as 100%. From the results in Table 6, it can be observed that there is a tendency for the modulus to decrease as sisal fibers are added, and for each layer of glass fiber replaced

Table 5. Tensile mechanical properties of the composites. Specific tensile strength and specific Young's modulus are based on the relative density of the composites.

Composites	Young's modulus (GPa)	Maximum strain (%)	Tensile strength (MPa)	Specific Young's modulus [GPa/(g/cm³)]	Specific tensile strength [MPa/(g/cm³)]
|GGG (F0)	10.1 ± 0.3	1.97 ± 0.08	152.8 ± 3.9	6.9	103.9
|GSG (F1)	7.1 ± 0.5	1.74 ± 0.20	97.5 ± 10.4	5.5	75.4
|SGG (F2)	7.6 ± 0.9	1.87 ± 0.20	106.8 ± 3.5	5.5	77.9
|GSS (F3)	4.9 ± 0.4	1.72 ± 0.28	54.6 ± 5.8	3.9	44.0
|SGS (F4)	5.0 ± 0.3	1.71 ± 0.07	60.8 ± 5.2	4.0	48.6
|SSS (F5)	3.0 ± 0.2	1.44 ± 0.24	24.6 ± 1.0	2.6	21.6

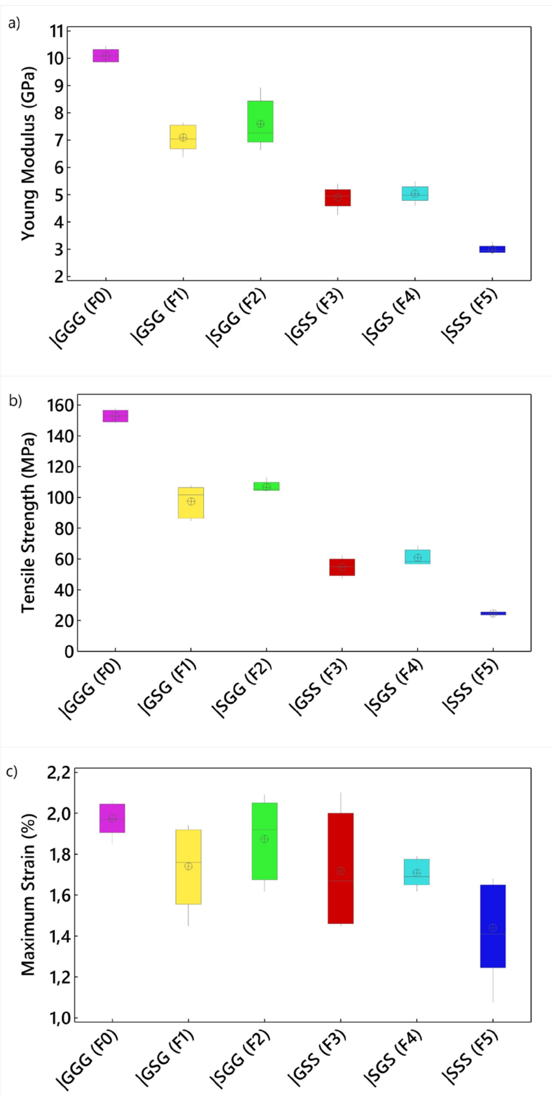

Figure 6. Tensile test results a) Tensile Young's modulus; b) Maximum tensile strength; c) Maximum tensile strain.

with natural fiber, the modulus is reduced by an average percentage between 20 and 30%. It can be seen that the tensile strength decreased between 25% and 35% per layer of sisal fiber added to the composite. The specimens that showed lower tensile strength were those of formulation F5, reaching a minimum value of 16% compared to the reference, which may be related to the low interaction between the sisal

Table 6. Retention percentage of the tensile mechanical properties of the composites, considering fiberglass composite as being 100%.

Composites	Young's modulus	Maximum strain	Tensile strength
|GGG (F0)	100%	100%	100%
|GSG (F1)	70%	88%	64%
|SGG (F2)	75%	95%	70%
|GSS (F3)	49%	87%	36%
|SGS (F4)	50%	87%	40%
|SSS (F5)	30%	73%	16%

fiber and the matrix. And for the maximum strain, it can be observed that only formulation F5 showed lower results than the others, reaching only 73% of the maximum strain of the reference formulation. This fact can be associated with the proximities between the strains of each of the glass and sisal fibers, as shown in Table 1.

3.4. Flexural (3-point-bend) test

Table 7 presents the results of the flexural test, the following properties were determined: flexural modulus, maximum flexural strain, flexural strength, specific flexural modulus and specific flexural strength. Figure 7 presents the flexural stress-strain curves for the flexural test considering an average curve for each formulation.

From Table 7 and Figure 7, it can be seen that the results of flexural modulus and flexural strength showed a decrease as sisal fibers are added, starting from the F0 formulation, a fact also observed in the study by Ornaghi et al.[17].

As for the flexural test, the direction in which the specimen is tested is relevant for the results, the behavior of formulas with the same fiber layers was modified according to the stacking of each one of them[23,24], even considering the standard deviations. This fact is due to the type of stress suffered by each end, being compression in the upper end and tensile in the lower layer. In other words, for the flexural test, the outer layers presents more influence on the results[23-26] and for the present study, if the glass fiber is in the tensile position, the results are higher than those for sisal fiber with the same composition. When comparing the specific modulus without the deviations, the value presented for formulation F1 (6.5 GPa/(g/cm³)) was higher than the reference (6.2 GPa/(g/cm³)), a behavior close to that found by Ford et al.[18]. As well as the result found for formulation F3 (3.5 GPa/(g/cm³)), which was tested in the direction of compression on sisal fibers, was higher than

Table 7. Flexural mechanical properties of the composites. Specific flexural strength and specific flexural modulus are based on the relative density of the composites.

Composites	Flexural modulus (GPa)	Maximum flexural strain (%)	Flexural strength (MPa)	Specific flexural modulus [GPa/(g/cm³)]	Specific flexural strength [MPa/(g/cm³)]
|GGG (F0)	9.1 ± 0.4	2.90 ± 0.07	216.6 ± 6.7	6.2	147.3
|GSG (F1)	8.3 ± 0.5	2.76 ± 0.16	188.5 ± 9.6	6.5	145.8
|SGG (F2)	4.6 ± 0.2	4.34 ± 0.18	134.4 ± 1.5	3.3	98.0
|GSS (F3)	4.3 ± 0.3	4.66 ± 0.31	119.0 ± 7.4	3.5	95.7
|SGS (F4)	3.2 ± 0.4	4.06 ± 0.53	64.3 ± 1.9	2.6	51.5
|SSS (F5)	2.8 ± 0.4	2.73 ± 0.50	43.5 ± 1.8	2.4	38.2

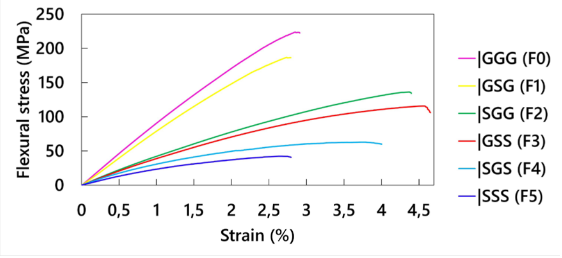

Figure 7. Flexural stress-strain curves for different composite recipes presented on Table 2.

that of formulation F2 (3.3 GPa/(g/cm³)), and the results for formulation F4 (2.6 GPa/(g/cm³)) was higher than the result presented by formulation F5 (2.4 GPa/(g/cm³)). In addition, the study by Abd El-baky[22] observed that for the same recipe with different arrangements, when the natural fiber (jute) composes the extremities of the laminate, the results of flexural strength were lower than when the glass fiber composes the extremities. This behavior is observed in the results from Table 7.

Figure 8 graphically presents the results of the flexural test. Regarding the modulus in Figure 8a, it is observed that, if the deviations are considered, the value presented by formulation F1 (8.3 ± 0.5 GPa) is statistically equivalent to that of the sample with all glass fiber layers (9.1 ± 0.4 GPa). Still analyzing this parameter, it is noted that the result presented for formulation F3 (4.3 ± 0.3 GPa) is statistically equivalent to the value presented for the modulus of formulation F2 (4.6 ± 0.2 GPa), which has one more layer of sisal fiber. Finally, the modulus of formulation F5 (2.8 ± 0.4 GPa) is statistically equivalent to the value presented by formulation F4 (3.2 ± 0.4 GPa).

Finally, for the F0 and F1 recipes, as well as for F2 and F3, the presence of sisal fiber in the inner layer is what makes the formulations differ, i.e. in the comparison, one has one more layer of sisal fiber than its peer. Note that the results are equal considering the standard deviations and in this case it is understood that the tensile and compression forces are acting in the same way on the surfaces of the composites, which have the same types of fiber, indicating that the core is not exerting significant influence on the results.

In Figure 8b, one can observe the decreasing influence of the addition of sisal fiber on the flexural strength results and the significant effect of the different fiber layer arrangements in the composite. Comparing pairs of the same formulation, it can be seen that formulation F1 (188.5 ± 9.6 MPa) presented a higher result than formulation F2 (134.4 ± 9.6 MPa), as well as in the comparison between F3 and F4, formulation F3 (119.0 ± 7.4 MPa) presented a higher result than formulation F4 (64.3 ± 1.9 MPa). Such results are related to the layers of the ends that are suffering the actions in the direction of tensile and compression, since both actions were applied to the glass fiber for formulation F1. For formulation F2,

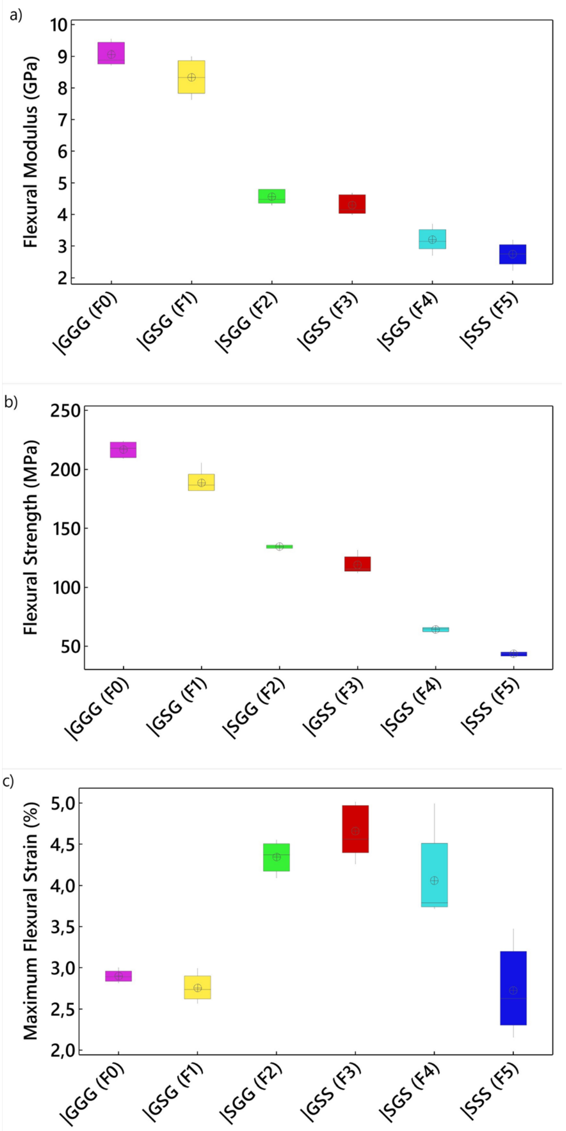

Figure 8. Flexural test results. a) Flexural modulus; b) Maximum flexural strength c) Maximum flexural strain.

compression was applied to the sisal fiber and tensile to the glass fiber. Formulation F3, which was tested with the fiberglass suffering the strain in the tensile direction and sisal in the compression strength direction, presented higher results than F4, which was tested with the sisal fiber at both ends.

Regarding the maximum strains (Figure 8c), it can be observed that there is a positive influence of sisal fibers in the composites, but it is not possible to identify a pattern of additivity as sisal fibers are added in hybrid composites. This result may be related to the proximity between the strains of pure fibers (Table 1).

From the results of flexural modulus in Table 8, it can be observed that the difference between formulations F0 and F1 was only 8%, however, when compared to the other hybrid composites and to formulation F5, the values reach 30% of the value presented by the reference formulation. Likewise, the results for the flexural strength obtained, but reaching a minimum value of 20% in relation to the reference sample for formulation F5. Note also that, for the results of maximum strain, the formulations of hybrid composites F3, F2 and F4 presented the highest values, reaching values 61%, 50% and 40% above the reference, respectively. This fact is in agreement with the results presented in the study of Ford et al.[18].

3.5. IZOD impact test

Table 9 presents the results of the impact test and specific impact strength. Figure 9 represents graphically the results of the impact test. It can be observed that the reference formulation presented the highest impact strength result, on the order of 80kJ/m^2. This result is in accordance with that predicted in the literature, since this formulation is composed exclusively of fiberglass, which has high strength characteristic.

Table 8. Retention percentage of the flexural mechanical properties of the composites, considering fiberglass composite as being 100%.

Composites	Flexural modulus	Maximum flexural strain	Flexural strength
|GGG (F0)	100%	100%	100%
|GSG (F1)	92%	95%	87%
|SGG (F2)	50%	150%	62%
|GSS (F3)	47%	161%	55%
|SGS (F4)	35%	140%	30%
|SSS (F5)	30%	94%	20%

Table 9. Impact mechanical properties of the composites. Specific impact strength is based on the relative density of the composites.

Composites	Impact strength (kJ/m^2)	Specific impact strength [(kJ/m^2) /(g/cm^3)]
|GGG (F0)	80.8 ± 3.4	54.9
|GSG (F1)	59.7 ± 11.2	46.2
|SGG (F2)	68.9 ± 9.7	50.3
|GSS (F3)	61.2 ± 9.1	49.2
|SGS (F4)	56.2 ± 7.7	45.0
|SSS (F5)	49.4 ± 4.5	43.4

The other formulations of hybrid composites (F1, F2, F3 and F4) presented statistically equivalent results when considering the deviations and similarities in the specific strengths presented. The formulation F5, which contains only sisal fiber, presented the lowest value for impact strength, however, when compared the values of specific strength the values are even closer to the hybrid composites, especially formulation F4. And because of the high deviations, also the characteristic of the test with the notched section, it was not possible to identify the influence of the layer order on the results in this test.In opposition to the trend presented in the work of Pavithran et al.[27], who studied hybrid composites of sisal-glass fiber laminates with unsaturated polyester resin, without description the processing parameters and process, they observed by the Charpy method that when the sisal was in the outer layers, the results of impact strength were higher than when the glass fiber was in the outer layers for the same composite composition.

3.6. Scanning electron microscopy (SEM)

Although the micrographs of all formulations are not in the paper, Figure 10 represents the interaction between the glass fiber and the matrix in all composites containing glass fiber in the composition. Similarly, Figure 11 represents the interaction between the sisal fiber and the matrix in all composites containing sisal fiber.

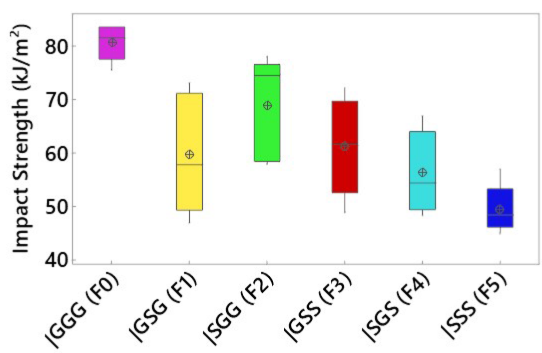

Figure 9. Impact IZOD test results of the composites.

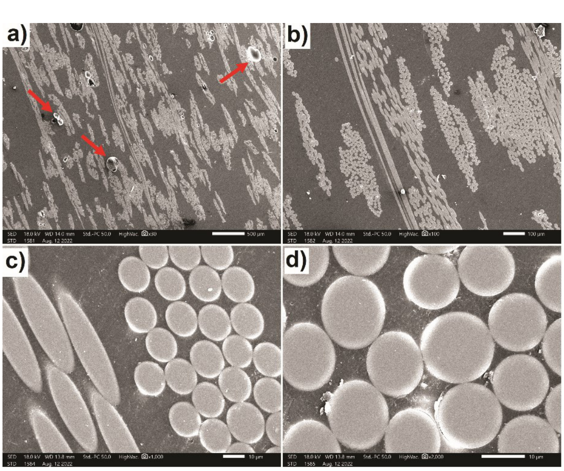

Figure 10. SEM image of the reference composite GGG (F0) with different magnitudes (red arrows indicate voids).

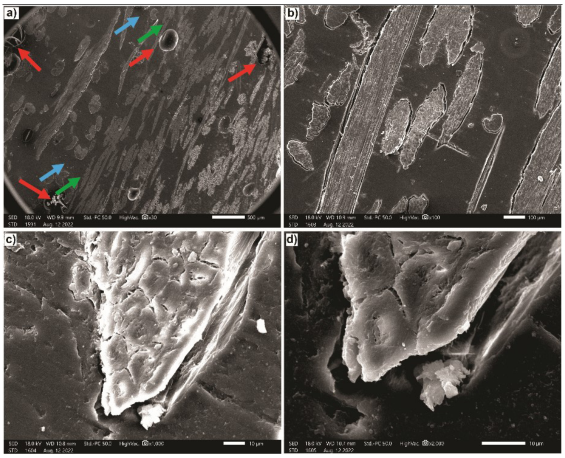

Figure 11. SEM image of GSG composite (F1) (Red arrows indicate voids, blue arrows indicate sisal fibers, and green arrows indicate glass fibers).

In Figure 10a), defects are seen in the form of voids, highlighted by red arrows. As in the resin vacuum infusion process low pressure gradient conditions are used, compared to other forms of higher pressure polymeric matrix processing, such as compression molding, SMC (Sheet Molding Compound), among others, the removal of air bubbles is hindered, and may thus lead to the appearance of defects in the form of voids. Unfortunately, this type of defect was inherent to the process conditions adopted in the infusion process, as it can also be seen in the sequence in all formulations.

The fiber stacking shown in Figure 10a) and b) indicates that there are regions with clusters of fibers, while there is little presence of fibers in other regions. In Figure 8c) and d), it is observed in detail that in some cases there is direct fiber-to-fiber contact, and in Figure 8d), there are also some points of non-coating of the fiber by the resin. Such facts may be the effect of poor percolation of resin in the composite and may have generated fragility points when the material was subjected to the mechanical stresses presented above, even for the evaluated F0 formulation, which presented the highest results in tensile, flexural and impact strength.

In Figure 11a, the presence of large defects on the composite surface in the form of voids is observed, as also verified for the F0 formulation. Although not presented here, it was verified that the aspect of the glass fibers in the formulations of the hybrid composites was equal to the aspect observed for formulation F0, so that the highlight in the micrographs was given to the sisal fibers (Figure 11b), c) and d)). However, what is evident in the micrographs is the non-coating of the sisal fibers by the polyester resin, due to the presence of voids in the matrix-fiber interface. It is known that sisal fiber is composed of a high percentage of cellulose fibers, which have very polar hydroxyl groups, and it is also known that the polymeric matrix used has ester functional groups, which are also polar. Thus, one could expect a certain level of interaction between the matrix and the sisal fiber, which would be inferred by the coating of the fiber by the matrix. However this effect was not verified due to the hygroscopic characteristic of sisal fibers and the fact that they were not previously treated since such treatments can remove some of the moisture and lignin present in the fibers and improve fiber coverage, as shown in the work of Lima et al.[3] and Huang et al.[13].

Similar to the behavior presented in this study, in the work by Silva et al.[4] were also observed bubbles and voids in the formulations of hybrid composites with natural fibers and glass fiber in unsaturated polyester resin. As well as in the work of Gupta and Deep[28], who studied hybrid composites with glass and sisal fiber totaling 8 layers, that related defects in the microstructures of hybrid composites with the worsening of the results of mechanical properties.

4. Conclusions

According to the results obtained, proposed formulations with density reduced by up to 23% and some proposed formulations that approached or exceeded the specific mechanical results of the reference composite with fiberglass were obtained, evidencing the possibility of using proposed composites with a more environmentally friendly UPR, mainly the formulation |GSG (F1) is recommended for future applications. The merits of hybridization process in this study are low cost and lightweight with comparable mechanical properties.

- As sisal fibers were added to the composite, it was noted that the density results decreased, and water absorption increased.
- From the tensile tests, it could be concluded that as layers of sisal fiber were added to the composite, the Young's modulus, maximum strain, and maximum stress obtained decreased, and the order of layering did not significantly affect the results.
- From the flexural tests, it was observed that there is a tendency in decreasing the flexural modulus and strength as sisal fibers are added. However, for the maximum deformation, the performance of formulations |SGG (F2), |GSS (F3) and |SGS (F4) was up to 60% higher than the reference. It was possible to observe that the layer arrangement order significantly affected the properties, and the internal layers did not significantly interfere in the results.
- For the IZOD impact strength tests, it can be observed that the reference formulation presented the highest impact strength result and the other formulations of the hybrid composites ((F1), (F2), (F3) and (F4)) presented statistically similar results if the deviations and the specific strengths are considered.
- By evaluating the micrographs obtained via SEM, it was possible to observe that all the formulations presented defects, in the form of voids, inherent to the processing conditions adopted, which made it difficult to remove the air bubbles from the system, but usually with the adjusted process conditions and recipes, the defects (deviations) obtained with VARTM process are fewer than in other processes.

5. Acknowledgments

The authors are grateful for the financial support of the Federal Institute of Espírito Santo (Ifes).

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