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Mechanical Behaviour of a Green Composite from Biopolymers Reinforced with Sisal Fibres

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ABSTRACT

In recent years, green composites based on thermoplastic matrices from renewable sources, and reinforced with natural fibres, have gained significant importance in different industrial applications, due to lower environmental impacts and production costs than traditional composites. This work investigates the manufacturing process, fibre/matrix integration and mechanical properties of a novel environmentally friendly green composite with a recyclable biobased polymer from a renewable source and a biodegradable natural fibre. Untreated woven sisal fibres reinforced post-consumer green polyethylene composites were evaluated in terms of flexural, tensile and impact properties. Traditional and green high-density polyethylene (HDPE), originated from sugarcane ethanol, were utilised as matrices of the investigated composites. Woven sisal fibres were arranged in two different stacking sequences, i.e. $[0^\circ/90^\circ]$ and $[\pm 45^\circ]$, being incorporated into the HDPE with a mass percentage proportion of 30/70 (fibre/matrix). A low-cost manufacturing process based on the hot compression moulding was used to produce the composites. The results were analysed by a factorial design to identify the effects of polyethylene type and the use of woven sisal fibres, considering the $[0^\circ/90^\circ]$ and $[\pm 45^\circ]$ orientations. Thermal gravimetric analysis was used to verify the thermal stability of the sisal fibre. The topographic surface of sisal fibres was observed by scanning electron microscopy. The results showed that the use of green polyethylene reinforced with untreated woven sisal fibres achieved higher flexural modulus (35%), flexural strength (13%), tensile strength (39%) and ultimate strain (68%) than traditional polyethylene without reinforcement. The green composite presented promising mechanical results to replace materials from non-renewable sources and can reduce manufacturing costs of the final product. These composite materials can be efficient for structural applications such as insulated panels, drywall and partitions for furniture.

33 **Keywords:** Green polyethylene, Woven sisal fibres, Green composites

34

35 **1. Introduction**

36 Natural fibre reinforced polymer composites have received great attention in recent years
37 due to the considerations of developing environmentally friendly materials. These composites have
38 been used in many applications by civil construction and automotive industries, where the weight
39 and density are influential factors in the performance of the final products and there is a great
40 concern about their life cycle [1].

41 The use of natural fibres as reinforcement in polymeric based composites have been
42 widely investigated. These natural fibres have many advantages such as low density, low cost,
43 carbon neutrality, recyclability and sustainability. Replacing synthetic fibres with natural fibres,
44 as the reinforcing element in composites, has been investigated in recent years to produce green
45 composites. In developing countries, the production of natural fibres can improve the life quality
46 and survivability of many communities, due to their environmentally friendly properties [2].

47 Green plastics, originated from renewable sources, are being studied as composite
48 matrices to reduce the environmental pollution generated by the extraction and processing of
49 petroleum [3]. A recently developed green polyethylenecan contribute to the reduction of
50 greenhouse gases, being one of the solutions to complex sustainability problems. It is produced
51 from ethanol that is derived from sugarcane, which is a renewable raw material that captures and
52 fixes carbon dioxide during its production. In addition, green plastics can also be reinforced with
53 fibres that can result in materials with higher mechanical strength [4].

54 The life cycle of a green composite (see [Fig. 1](#)) begins with the use of raw materials from
55 renewable sources, such as sugarcane and sisal, to produce the constituent materials. The ethanol
56 from sugarcane is dehydrated, obtaining the green ethene, which is polymerized and transformed
57 into a green PE, widely used in plastic bags [5]. In the same sustainable context, sisal fibres are
58 extracted from the plant leaf and can be woven in various orientations, resulting in a natural and
59 biodegradable constituent [6]. The resulting green composite is an environmentally friendly
60 material, produced by a rapid and low-cost manufacturing process, which contributes to the
61 reduction of carbon dioxide in the atmosphere.

62

63



Fig. 1. The life cycle of a green composite.

The mechanical behavior of natural fibres reinforced polymer composites can be influenced by several physical and chemical factors, such as properties of constituent materials, fibre and matrix content, fibre length and orientation, physical profile of the contact surface, interlaminar shear strength and interfacial chemical bonding [7]. Consequently, the failure mode of these materials depends on the combination of these factors and the overall system response when a given load is applied to a specific region of the composites.

Many studies have evaluated the mechanical behaviour of green composites by flexural, tensile and impact tests [8, 9]. Robledo-Ortiz et al. [1] studied rotomoulded composites with green polyethylene and natural fibres of agave tequilana and coir treated with maleated polyethylene, as sustainable materials to replace petroleum-based polymer composites, and found improvements in tensile modulus (34%) and tensile strength (12%), at 30% wt fibre content, highlighting also the possibility of reducing production costs with the use of fibres. Guilhen et al. [10] investigated the mechanical properties of bleached cellulose fibres reinforced green polyethylene composites produced by a co-rotating intermeshing twin-screw extruder, obtaining improvement in tensile and flexural properties in case of 30% wt of fibres. Garcia-Garcia et al. [5] used different compatibilisers on green composites, manufactured with green polyethylene and peanut shell, finding good results in tensile and flexural modulus using a polypropylene-graft-maleic anhydride compatibiliser.

Mechanical tests allow the understanding of the mechanical properties of the material and can be the basis of the study of the failure mode resulting from the stress imposed. According to Hughes [11], failures in fibres may occur due to the damage caused in their cellulose molecules,

88 which are essential in the mechanical strength of these materials. Besides, plastic deformation and
89 cracks propagation in various directions in polyethylene may also occur, depending on the applied
90 stress and the chemical bonds between phases [12].

91 The matrix-fibre interface influences the mechanical properties of composites, and so it
92 has been widely studied by researchers. The use of compatibilisers has been investigated to
93 improve the interfacial bonding between the fibres and matrix, showing significant effects on the
94 mechanical properties of these composites [13]. Fávvaro et al. [14] investigated the mechanical
95 properties of post-consumer polyethylene with mercerised and acetylated sisal fibres (5–10 mm in
96 length), mixed in a lab-made mono-screw extruder, founding improvements on flexural and impact
97 properties.

98 On the other hand, chemical treatments in lignocellulosic fibres, such as sisal, fax and
99 coir, can cause strong damage to the structure of these fibres, reducing their mechanical properties.
100 In addition, the use of these chemical agents does not contribute to the sustainable concept of these
101 materials, due to the complexity of degradation of the chemical structure of these agents [15].

102 This study investigates the mechanical behaviour of a novel untreated woven sisal fibre-
103 reinforced post-consumer green polyethylene composite, combining two specific environmentally
104 friendly materials: a post-consumer polyethylene matrix made of sugarcane (green HDPE), which
105 is a recyclable biobased polymer made from renewable source, and untreated natural woven sisal
106 fibres. No chemical treatments were used on the fibres, minimising the potential impacts of these
107 chemical agents to the environment. A low-cost and simple composite manufacturing process was
108 detailed and a factorial design with six treatments was performed. The results indicate that the
109 investigated green composite can be used as an efficient and sustainable alternative to replace the
110 traditional composites in different types of structural applications.

111

112 **2. Materials and methods**

113 [Table 1](#) summarises the designed and conducted experiments in this paper. Six treatments
114 were investigated based on two factors. Two types of PE were defined: the traditional PE and the
115 green PE. The traditional PE represents the plastic generated from the use of petroleum (non-
116 renewable source) and the green PE represents the plastic generated by a renewable source, from
117 sugarcane ethanol. The reinforcement factor had three levels: without reinforcement and
118 reinforced by woven sisal fibres with two different configurations, i.e. $[0^\circ/90^\circ]$ and $[\pm 45^\circ]$.

119 Flexural, tensile and Charpy impact tests were carried out in a room temperature of 25 °C and an
 120 average relative humidity level was at 65 %. Five specimens for each experimental condition plus
 121 a replica were evaluated.

122
 123

Table 1
 Experimental conditions

Experimental conditions	Reinforcement type
Traditional HDPE	Without reinforcement
Traditional/90	Woven sisal fibres in [0° / 90°]
Traditional/45	Woven sisal fibres in [± 45°]
Green HDPE	Without reinforcement
Green/90	Woven sisal fibres in [0° / 90°]
Green/45	Woven sisal fibres in [± 45°]

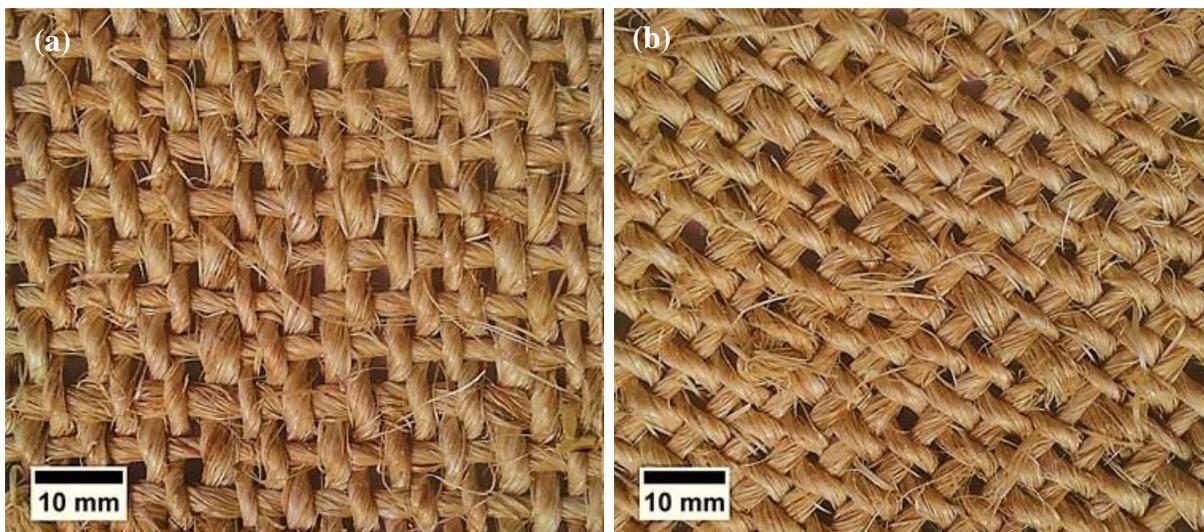
124

125 2.1. Materials

126 Traditional high-density PE (HDPE) films, supplied by the PACK FORT, and green PE
 127 biopolymer films, supplied by the UNISOLD and produced by the BRASKEM petrochemical
 128 company, were used in this work. This biopolymer was industrially prepared from ethene produced
 129 from ethanol, originated a result of fermentation and distillation processes of sugarcane [9].

130 Woven sisal fibres were used to manufacture the composites. Woven sisal fibres (Fig. 2),
 131 with yarns type 530/1, were supplied by APAEB Sisal (Brazil). It should be noted that all the raw
 132 materials were used as supplied, with some simple cleaning for removal of dirt and surface
 133 impurities. No further treatment or modification was done in their molecular structure.

134



135

136 **Fig. 2.** Woven sisal fibres (a) $[0^\circ/90^\circ]$ and (b) $[\pm 45^\circ]$.

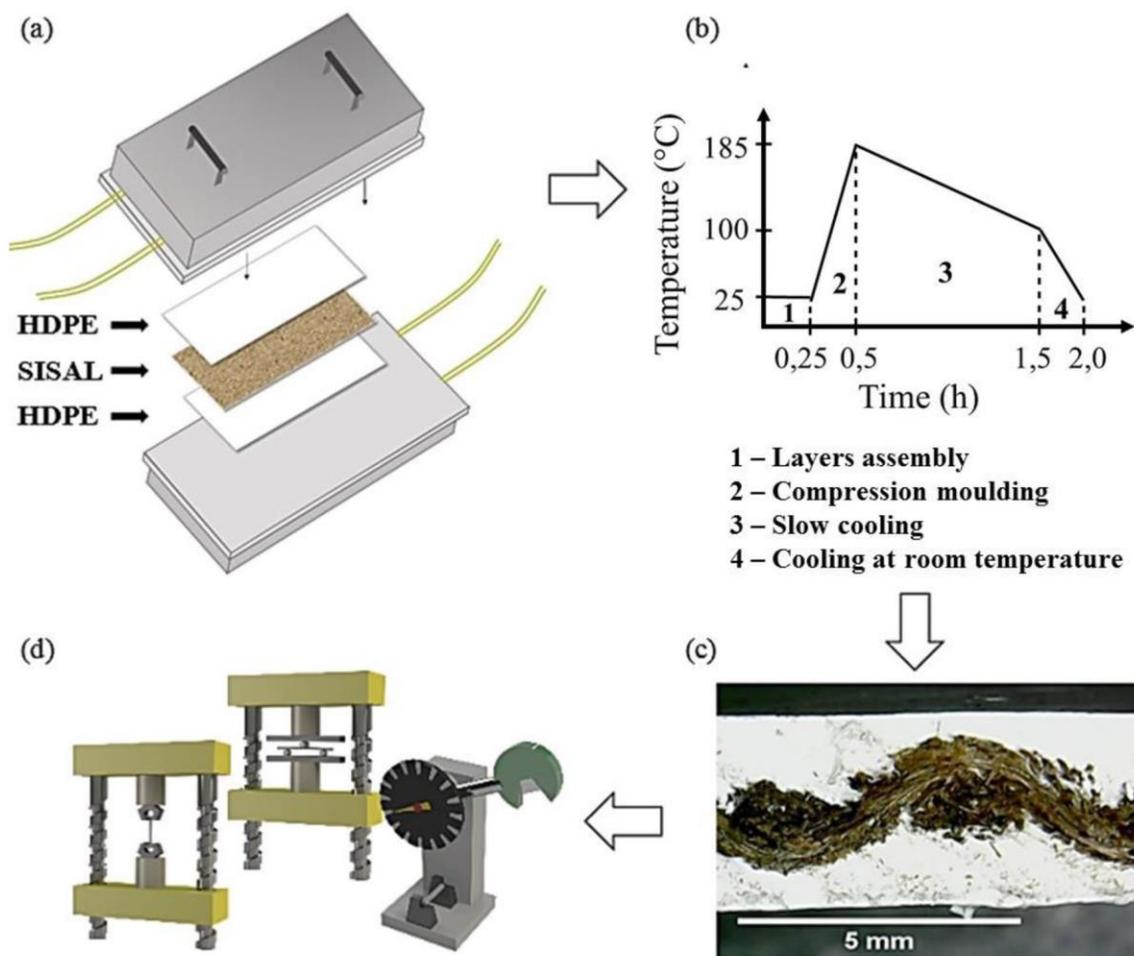
137

138 *2.2. Composites Manufacturing*

139 The composite samples were manufactured in the Laboratory of the Innovation and
140 Technology in Materials (GITEM), Federal University of Minas Gerais. A single layer of cleaned
141 woven sisal fibres was stacked by the PE films and the laminate was cured in a compression mould
142 with a controlled heating (185°C). The woven sisal fibres had a biaxial configuration and they
143 were investigated in two orientations in the composites, i.e. $[0^\circ/90^\circ]$ and $[\pm 45^\circ]$. The woven sisal
144 fibres had a length of 190 mm and a width of 140 mm, weighing approximately 34g. The PE
145 weight was 79g for each plate.

146 A compression machine (Fig. 3) was constructed for the compression and compaction of
147 the composites in a proportion of 30% fibre / 70% matrix [13]. The pressure used was 12.39 KPa.
148 A k-type thermocouple, which was attached to a multimeter Icel model MD-5660C, was used to
149 measure the mean temperature near the plates. The temperature was controlled to reach
150 approximately $185 \pm 5^\circ\text{C}$. This temperature was based on the results of thermal gravimetric
151 analysis (TGA) in the sisal fibres and in experiments conducted by other researchers at different
152 temperatures [13]. The average heating rate measured in the experiments was $12.5^\circ\text{C}/\text{min}$. Once
153 the temperature was set for the heating of the machine, the resistors were switched off and the
154 plates were kept inside the machine to cool down until 100°C . The specimens were in a solid form
155 at 100°C . After that, the specimens were removed from the machine and kept at room temperature
156 (25°C). The curing cycle is shown in Fig. 3.

157



158
159 **Fig. 3.** Manufacturing procedures of the composites: (a) materials and compression machine, (b)
160 the curing process, (c) manufactured composites and (d) mechanical tests.

161 *2.3. Thermal, Morphological and Mechanical Characterization*

162 The surface of the fibre was examined using a scanning electron microscope (SEM),
163 Hitachi TM-3000, with an accelerating voltage of 15 kV. Thermal gravimetric analysis (TGA) was
164 performed in a sisal fibre using a Perkin Elmer DTG-60H Thermal Analyzer. The test occurred
165 under controlled atmosphere (Nitrogen), with a heating rate of 3°C/min, ranging from 25 °C to
166 360 °C. Three-point flexural tests were performed using a universal test machine of Shimadzu,
167 model AG-X Plus, in accordance with [17], with specimens' dimensions as 78 mm in length and
168 12.7 mm in width, and 3 mm/min cross-head feed rate. The flexural modulus, flexural strength and
169 displacement were measured for each experimental condition. The tensile tests were also carried
170 out using a Shimadzu test machine, model AG-X Plus, and in accordance with [18], specimens'

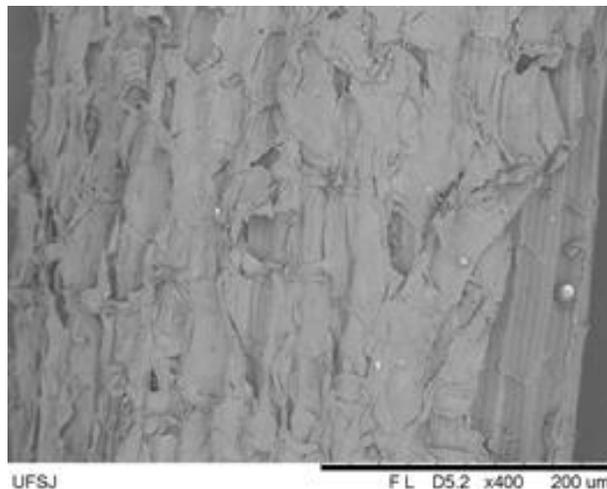
171 dimensions as 160 mm in length, 19 mm in width and cross-head feed rate of 2 mm/min. The
172 tensile modulus, tensile strength and ultimate strain were evaluated. Charpy impact tests were
173 performed using an XJJ-50 Series machine, with a pendulum of 15 J and low-velocity speed at 3.8
174 m/s, to measure the energy absorbed. The impact was performed according to [19] in the lateral
175 section of the specimens, which had dimensions of 80 mm in length and 10 mm in width.

176

177 **3. Results and discussion**

178 *3.1. Scanning Electron Microscopy (SEM)*

179 For this study, the sisal woven passed through a washing process in order to reduce
180 impurities on its surface to improve the fibre/matrix interface. After drying at room temperature,
181 the washed fibres were used to manufacture the composite. Fig. 4 shows optical images of the pure
182 sisal fibre, the fibre's surface presents impurities composed by parenchyma, lignin, hemicellulose
183 and waxes, these impurities influence adhesion between the fibre and polymer. It was only done
184 by washing the fibres to avoid toxic chemical agent commonly used in treatment of natural fibres
185 [14]. No treatment with chemical agents was performed, due to degradation in the fibres and
186 especially the environment caused by these agents [16, 20].



187

188 **Fig. 4.** Optical microscopy of the neat sisal fibres.

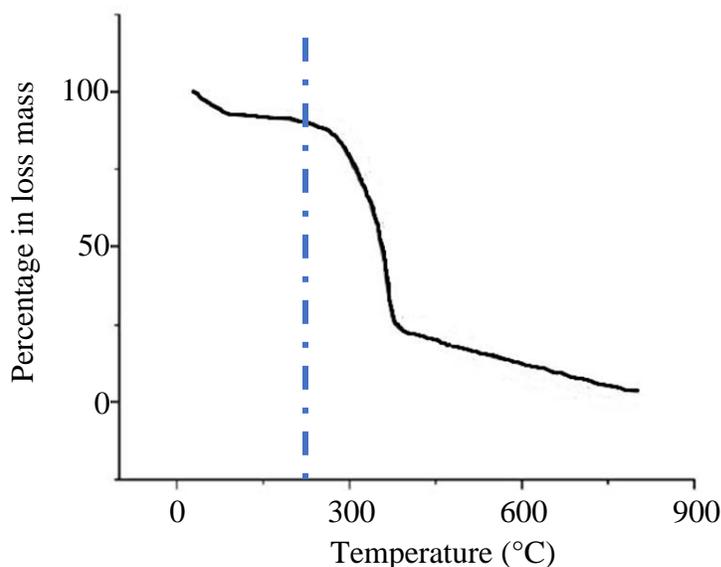
189

190 *3.2. Thermal Gravimetric Analysis (TGA)*

191 Fig. 5 shows the TGA results of the utilised sisal fibre. An initial weight loss around the
192 temperature range 20-225 °C occurred that is related to the surface water evaporation. In

193 temperature range 230-350 °C, the fibres began to degrade, implying a significant loss in their
194 weight and mechanical properties [21].

195



196

197

198

Fig. 5. Thermal gravimetric analysis of the utilised sisal fibre.

199

200 From the TGA, it was possible to find the working temperature range for the sisal fibres,
201 which is before they begin to degrade (the blue dashed line in Fig. 5, approximately 200 °C). This
202 is an important manufacturing parameter, especially for high temperature applications. A
203 temperature selection is, therefore, a necessity for the manufacturing of composites to avoid
204 degradation by the high temperature in the sisal fibre.

205

206 3.3. Mechanical Tests

207 Analysis of Variance (ANOVA) was adopted to verify if the effect of the main factors
208 and interaction of factors are statistically significant. A p-value lower or equal to 0.05 indicates a
209 significant effect provided by the main factors, or the presence of the interaction of factors on the
210 response variables, considering a confidence interval of 95%. The main factor is only interpreted
211 individually when there is no evidence that it interacts with the other factors [22]. Table 2 presents
212 the results of ANOVA for the mean values of the mechanical properties for each experimental
213 condition. The assumptions of the normal probability of residuals and the homogeneity of
214 variances were checked and confirmed. ANOVA was used to verify the influence of the resin and

215 fibre types and the interaction between them on the response variable, as a support to the
 216 conclusions found in the descriptive statistics, verifying whether the means of each experimental
 217 condition are different or not. Values of adjusted R^2 are greater than 90%, showing that the models
 218 explain a considerable variation of data. Minitab 17 software was used to perform statistical
 219 analysis based on ANOVA.

220 **Table 2**
 221 Analysis of Variance (ANOVA)

Factors	P-value						
	Flexural Modulus (GPa)	Flexural Strength (MPa)	Displacement (mm)	Tensile Modulus (GPa)	Tensile Strength (MPa)	Ultimate Strain (%)	Energy absorbed (J)
Polyethylene type	<u>0.000</u>	<u>0.000</u>	<u>0.000</u>	0.115	<u>0.000</u>	<u>0.000</u>	0.957
Reinforcement type	<u>0.000</u>	<u>0.000</u>	<u>0.000</u>	<u>0.000</u>	<u>0.000</u>	<u>0.000</u>	<u>0.000</u>
Polyethylene* Reinforcement type	0.132	<u>0.000</u>	<u>0.030</u>	<u>0.000</u>	0.219	<u>0.000</u>	<u>0.000</u>
R² (Adjusted)	92.06 %	93.31 %	93.25 %	92.39 %	92.74 %	91.56 %	91.02 %

222

223 3.3.1. Flexural Tests

224 The type of matrix and the reinforcement orientation significantly influenced the flexural
 225 properties presented from the tests, with p-value lower than 0.05, including the interaction between
 226 these factors in the case of flexural strength and displacement (Table 2). Mechanical properties
 227 obtained from the flexural tests are presented in Table 3. In general, green composites presented
 228 higher values for the flexural properties than composites with traditional HDPE, and composites
 229 with woven sisal fibres in orientation $[\pm 45^\circ]$ showed better results than those with orientation
 230 $[0^\circ/90^\circ]$. The heterogeneity of the composite structure, considering the gaps between the yarns,
 231 the wettability of the fibres and slight variations of the temperature in heat zones with polymers in
 232 melting process, may have influenced these results. In addition, flexural involve a complex stress
 233 state, with tensile, compression and shear stresses present simultaneously in the specimen. When
 234 the fibres are oriented at $[\pm 45^\circ]$, the applied load is resolved into longitudinal and transverse
 235 components, thus developing shear flow in the matrix [23]. According to [24], shear stresses are
 236 common in these materials under flexural tests and they influence the results. When the interfacial
 237 bonding is poor the delamination size can increase linearly, and it is only affected by the applied

238 stress. When the ultimate limit is exceeded, the layers are debonded and the global stiffness is
 239 reduced [25].

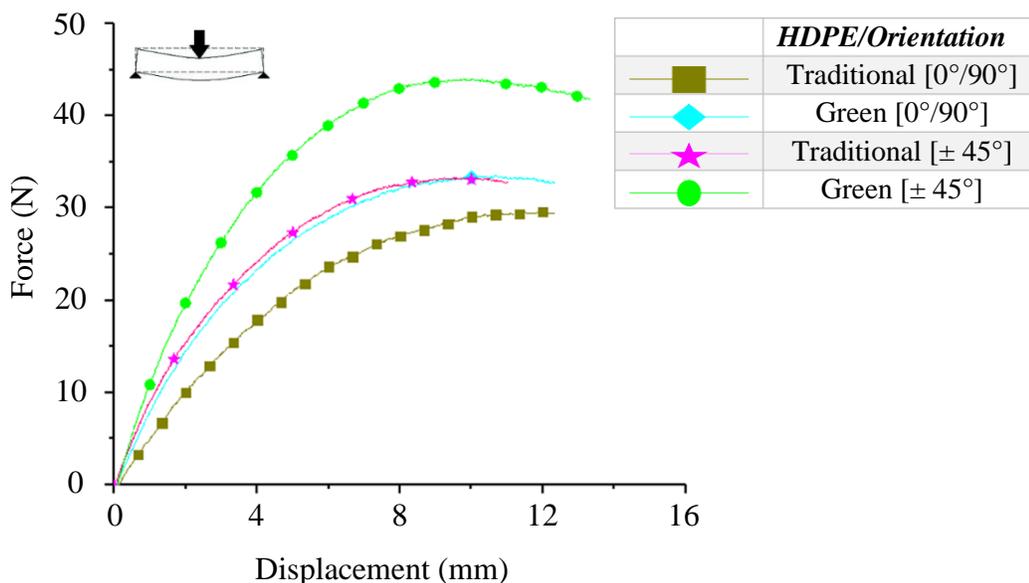
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 241

Table 3
 Flexural tests

Experimental conditions	Flexural Modulus (GPa)	Flexural Strength (MPa)	Displacement (mm)
Traditional HDPE	0.63 ± 0.03	16.27 ± 1.11	9.81 ± 0.85
Traditional/90	0.53 ± 0.02	14.20 ± 0.35	11.94 ± 0.35
Traditional/45	0.72 ± 0.02	14.45 ± 0.80	11.66 ± 0.42
Green HDPE	0.72 ± 0.04	22.67 ± 0.45	11.46 ± 0.19
Green/90	0.65 ± 0.02	14.62 ± 0.92	12.36 ± 0.46
Green/45	0.85 ± 0.03	18.44 ± 0.61	13.27 ± 0.73

242

243 **Fig. 6** shows examples of force-displacement curves for the composites, that evidence the
 244 elastic region of these materials, with lines more inclined in case of reinforcements in the $[\pm 45^\circ]$
 245 orientation. In these tests, shear failure can be assumed as the predominant mode, when the load-
 246 displacement curve decreases gradually to zero [24]. Considering the shear modulus of this type
 247 of reinforcement, when fibres are at $+45^\circ$ and -45° , there is a higher incidence of fibre performance
 248 than at 0° and 90° , due to the fact that fibres oriented to 90° (upright to the test) do not behave in
 249 the same proportion as a 45° , which tends to increase its rigidity under flexural stress. Chianelli-
 250 Junior et al. [26] found a similar result in his study of the flexural behaviour of composites with
 251 post-consumer HDPE and sisal fibres, attributing it to the weak interfacial adhesion of these
 252 composites.



253

254

Fig. 6. Flexural tests: force-displacement curves of composites.

256

257 3.3.2. Tensile Tests

258 The statistical ANOVA showed a significant influence of the tensile modulus and
 259 ultimate strain in the tensile tests, with p-value less than 0.05 (Table 2). Table 4 shows the tensile
 260 modulus, tensile strength and tensile ultimate strain values. The tensile tests results showed that
 261 woven sisal fibres in orientation $[\pm 45^\circ]$ reinforced composites had lower tensile modulus than the
 262 non-reinforced HDPE in both types of HDPE. These composites presented a high level of
 263 interlaminar tension in tensile stresses, reducing stiffness in the tensile stress. This mechanism can
 264 be explained by interfibre / interlaminar failure theory proposed by Daniel et al. [27], which is
 265 based on the assumption that the failure of composite materials is governed by the limiting
 266 microscopic strain in the interfibre/interlaminar region of the layer. A compression dominated
 267 failure is governed by the microscopic shear strain, a shear dominated failure is governed by the
 268 tensile strain, and failure by combined tension / shear is dominated by the tensile strains [28].

269

Table 4

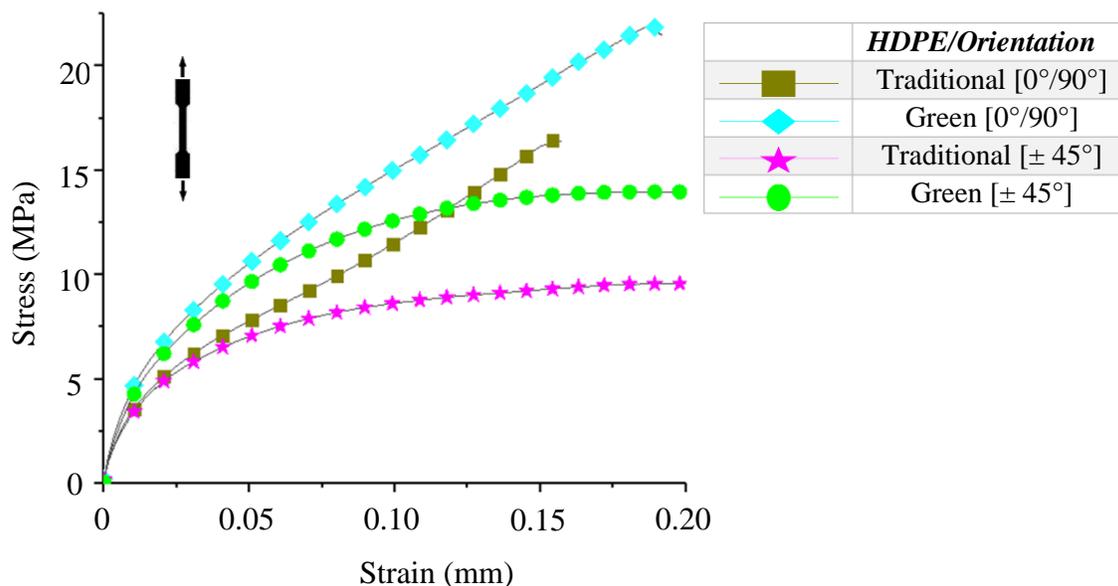
Tensile tests

Experimental conditions	Tensile Modulus (GPa)	Tensile Strength (MPa)	Ultimate Strain (%)
Traditional HDPE	0.72 ± 0.02	14.43 ± 0.40	11.73 ± 0.75
Traditional/90	0.63 ± 0.02	16.86 ± 0.75	15.57 ± 0.49
Traditional/45	0.55 ± 0.02	10.26 ± 0.83	19.88 ± 1.27
Green HDPE	0.67 ± 0.01	19.12 ± 0.92	19.83 ± 0.67
Green/90	0.65 ± 0.01	20.12 ± 1.26	19.69 ± 0.94
Green/45	0.63 ± 0.01	13.69 ± 0.88	19.42 ± 1.51

271

272 Regarding the reinforcement, composites with woven sisal fibres in orientation $[0^\circ/90^\circ]$
 273 presented higher tensile strength than those with orientation $[\pm 45^\circ]$, as can be seen in Fig. 7, with
 274 examples of stress-strain curves for each composite. Tensile tests were performed in the 0°
 275 direction, so fibres in alignment with that direction provide better results in tensile strength,
 276 improving the performance of composites reinforced with woven sisal fibres in $[0^\circ/90^\circ]$. Jawaid
 277 et al. [29] investigated the tensile properties of woven jute fibres reinforced polymeric composites
 278 and emphasized that the tensile strength of fibres reinforced composites is influenced by the
 279 strength and modulus of fibres. The authors reported that woven fibres exhibit low deformation at

280 the break and hence these composites exhibit high strength. It may be due to the stretching nature
 281 of woven fibres and strands break at different times as each fibre can stretch independently and
 282 break individually when reaching their failure stress. In tensile tests, composites with woven sisal
 283 fibres in orientation $[\pm 45^\circ]$ present multi-axial states of stress, involving combined transverse and
 284 shear loadings, which may have a strong effect on the failure behavior of the material, reducing
 285 the tensile strength of these composites [28]. In addition, Fig. 7 also shows a higher stiffness of
 286 the green PE composites with both types of reinforcement, evidencing the significant influence of
 287 different factors interaction in the tensile modulus as verified by the ANOVA.



288 **Fig. 7.** Typical stress-strain curves for composites.

289

290 3.3.3. Charpy Impact Tests

291 The results from Charpy impact tests showed that the interaction of different factors
 292 affecting the absorbed energy significantly, based on the ANOVA and p-value less than 0.05 as
 293 exhibited in Table 2. The behaviour in impact resistance of the composites with woven sisal fibres
 294 in orientation $[0^\circ/90^\circ]$ has been more effective than the orientation $[\pm 45^\circ]$ (Table 5). This was
 295 observed in both the green and traditional composites. Some internal defects inside the composites
 296 emerged from the manufacturing process, such as voids, delamination, and resin-poor or resin-rich
 297 regions, which had notable effects on the mechanical properties of the composites [30].

298

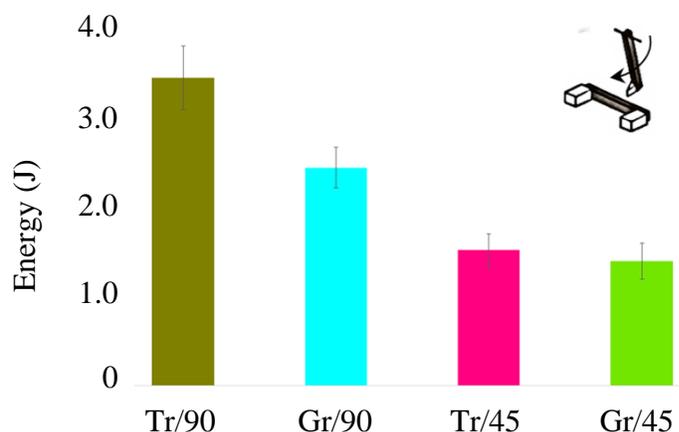
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Table 5
Charpy impact tests

Experimental conditions	Energy absorbed (J)
Traditional HDPE	2.57 ± 0.29
Traditional/90	3.45 ± 0.36
Traditional/45	1.52 ± 0.18
Green HDPE	3.99 ± 0.16
Green/90	2.44 ± 0.23
Green/45	1.39 ± 0.16

300

301 Considering the Charpy impact tests, the applied load transferred by shear to fibres was
 302 greater than the interfacial bond strength, causing debonding. In this context, the fibre length did
 303 influence the energy dissipation. Long fibres had a larger absorption capacity by avoiding damages
 304 like fibre pull-out and fibre breakage [31]. The woven sisal fibres in orientation $[0^\circ/90^\circ]$ had larger
 305 yarns in the transverse direction of the impact than those in orientation $[\pm 45^\circ]$, which provided
 306 higher energy absorption and higher impact resistance (Fig. 8).



307

308 **Fig. 8.** Absorbed Charpy impact energy for each condition.

309

310 3.4. Fracture analysis of the composites

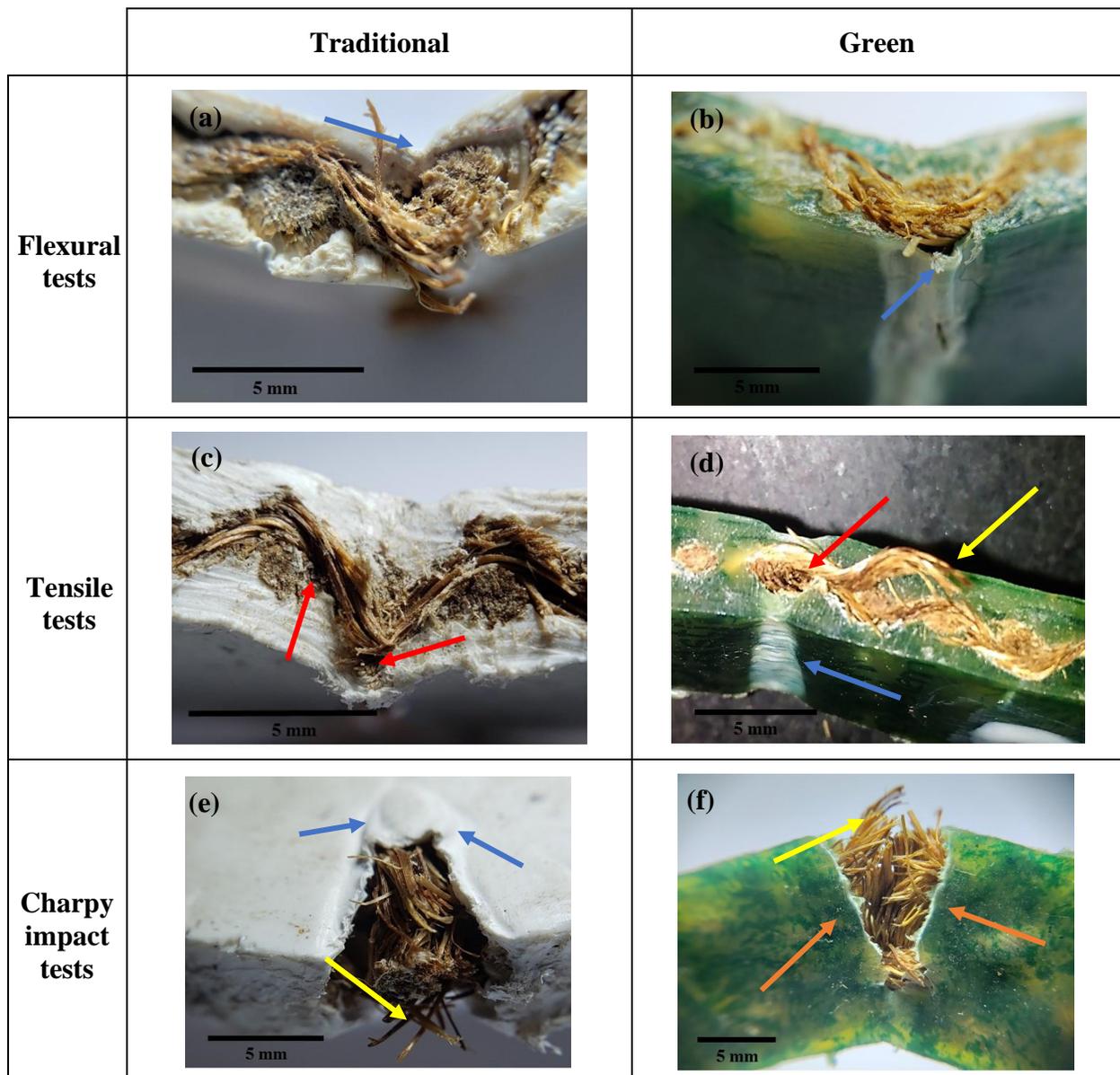
311 Fig. 9 and Fig. 10 show the failures of the composites, with woven sisal fibres in
 312 orientation $[0^\circ/90^\circ]$ and $[\pm 45^\circ]$, respectively. In the flexural tests, fibre-matrix debonding were
 313 evident in the specimens, being the predominant failure mechanism. This failure is a typical
 314 problem of poor fibre-matrix adhesion, which may be due to the hydrophilic nature of sisal fibres
 315 and the hydrophobic nature of PE, not allowing the wetting of the fibres by the matrix and avoiding
 316 coupling of the phases. In addition, delamination (red arrow) and fibre pull-outs (yellow arrow)
 317 were also detected, as shown in composites with woven sisal fibres in orientation $[\pm 45^\circ]$ (Fig. 10a

318 and 10b), indicating the weak interfacial bond. Chaudhary et al. [32] investigated the flexural
319 properties of sunnhemp fibre-reinforced waste PE bag composites, and reported a similar behavior,
320 attributing it to the poor fibre-matrix adhesion that caused microcrack formation at the interface
321 and a non-uniform stress transfer between the fibre and the matrix. Moreover, there are stretching
322 zones in the composites with woven sisal fibres in orientation $[0^\circ/90^\circ]$ and $[\pm 45^\circ]$ (Fig. 9a, 9b and
323 Fig. 10b), with an evident plastic deformation (blue arrow) of the matrix phase. Thermoplastics,
324 such as PE, can be reprocessed through melting and molding processes. According to the heating
325 and cooling conditions, the polymer chains can be reconfigured, which modifies their strength and
326 stiffness. In addition, when plastic deformation (blue arrow) of these materials occurred, a change
327 in their color was observed in the composites with woven sisal fibres in orientation $[0^\circ/90^\circ]$ (Fig.
328 9b) due to the molecular interactions or molecular conformations [33].

329 Similar to the flexural tests, the presence of plastic deformation was also be observed in
330 the composites with woven sisal fibres in orientation $[0^\circ/90^\circ]$ and $[\pm 45^\circ]$, under the tensile tests
331 (Fig. 9c, 9d and Fig. 10d). They also presented delamination and fibre pull-outs, as can be seen in
332 Fig. 10c and 10d. The problems in the interfacial adhesion did not allow the transfer of loads
333 between different phases of the composites. Thus, after the interfacial shear strength was exceeded,
334 the tension resulting from the tensile stress was concentrated on the PE plastic deformation and on
335 the tensile strength of the oriented fibres in the test direction (90°), which explains the better
336 performance of the composites reinforced with woven sisal fibres in $[0^\circ/90^\circ]$, compared to
337 orientation $[\pm 45^\circ]$. Habibi et al. [34] also reported a similar behaviour, i.e. observing delamination
338 in tensile tests for lignocellulosic fibres reinforced with PE, indicating that lignocellulosic fibres
339 were not well wetted by the matrix, creating a gap around the fibres.

340 In Charpy impact tests, the woven fibres reinforced with traditional PE, and orientation
341 $[0^\circ/90^\circ]$, failed with a notable plastic deformation (Fig. 9e). Composites with green PE with woven
342 sisal fibres in orientation $[0^\circ/90^\circ]$ and $[\pm 45^\circ]$ (Fig. 9f and Fig. 10f) showed a matrix breakage
343 (orange arrow) without the evident zone of stretching. This difference in the plastic deformation
344 in the failure of the PE can be explained by the different conformations and molecular interactions
345 resulting from the reprocessing of the material and additives already existing in these plastic
346 materials. In addition, breakages and pull-outs of fibres (Fig. 9e, 9f and Fig. 10e, 10f) could be
347 observed, reaffirming the weak interfacial bond presented in the composites. The occurrence of
348 fibre pull-out and fracture may be related to the energy dissipated in the impact, as the fibres of

349 greater length in the $[0^\circ/90^\circ]$, compared with the $[\pm 45^\circ]$, may allow a better energy propagation.
 350 These longitudinal fibres of the composites, oriented transversely to the direction of the impact
 351 pendulum, may realize friction in the matrix and dampen a greater amount of energy. According
 352 to Mulinari et al. [35], brittle behavior of fibres can limit the plasticity behavior of the composites
 353 because of mechanical friction process, resulting in the fibre pull-out from the matrix. Zhao et al.
 354 [36] measured impact properties of sisal fibres reinforced PE composites, and reported that in the
 355 impact event, fibre pull-out, fracture of fibres and the matrix are the most common modes of energy
 356 dissipation when the interfacial bond is weak.



357 **Fig. 9.** Fractures analysis of the composites with woven sisal fibres at $[0^\circ/90^\circ]$.

358

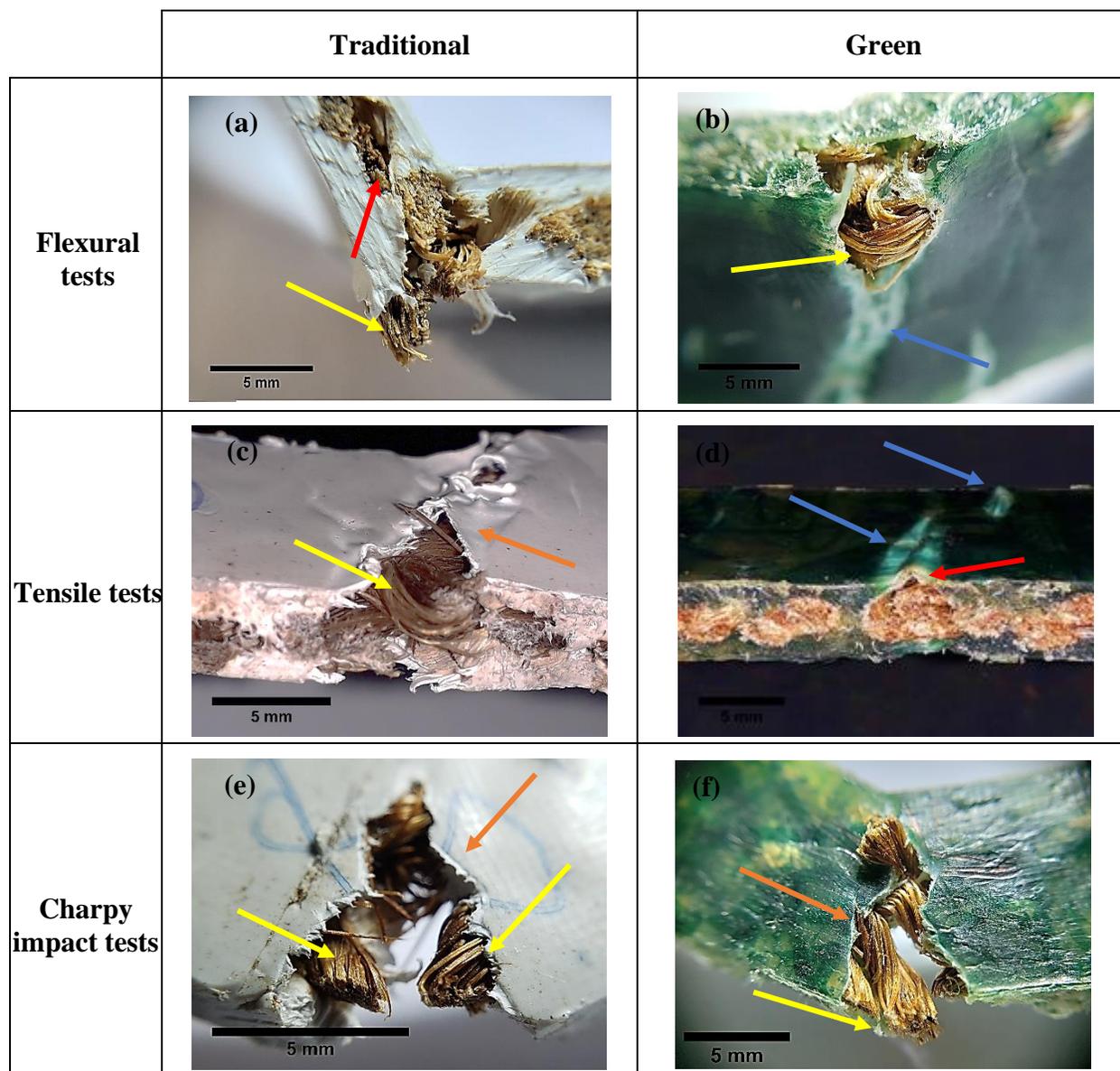


Fig. 10. Fractures analysis of the composites with woven sisal fibres at $[\pm 45^\circ]$.

359

360

361 4. Conclusions

362 This work investigated the mechanical properties of green/traditional post-consumer polyethylene
 363 composites reinforced of untreated woven sisal fibres oriented in the $[0^\circ/90^\circ]$ and $[\pm 45^\circ]$. The main
 364 conclusions of this study were:

- 365 • The low-cost manufacturing process based on the hot compression moulding with controlled
366 pressure (12.4 kPa) and temperature (185 °C) was able to manufacture the composites with
367 homogeneity and repeatability.
- 368 • Based on ANOVA, the use and orientation of woven sisal fibres into the polyethylene matrix had
369 a significant effect on all the mechanical properties evaluated, and values of adjusted R2 were
370 greater than 90%, showing that the models explained a considerable variation of data.
- 371 • The green composites presented higher mean values for flexural modulus (35%) and flexural
372 strength (13%) than traditional polyethylene without any reinforcement.
- 373 • The mean flexural modulus and flexural strength were higher for green composites with woven
374 sisal fibres in $[\pm 45^\circ]$ than composites with fibres in $[0^\circ/90^\circ]$, presenting increases of 30% and 26%,
375 respectively.
- 376 • The green composites had higher mean values for tensile strength (39%) and ultimate strain (68%)
377 than traditional polyethylene without any reinforcement, but lower mean tensile modulus (10%)
378 due load transmission problems between matrix and fibre.
- 379 • Tensile tests showed higher mean tensile strength for the composites with woven sisal fibres in
380 $[0^\circ/90^\circ]$ than those with fibres in orientation $[\pm 45^\circ]$, presenting increase of 64% with the traditional
381 polyethylene matrix and increase of 47% with the green polyethylene matrix.
- 382 • The energy absorbed in the Charpy impact tests of the green composites was similar to traditional
383 polyethylene without reinforcement and composites with woven sisal fibres in orientation $[0^\circ/90^\circ]$
384 presented higher energy absorbed than the composites with fibres in $[\pm 45^\circ]$, with 126% increase
385 with the traditional polyethylene matrix and 75% increase with the green polyethylene matrix.
- 386 • The reinforcement of woven sisal fibres into composite materials, besides improving their
387 mechanical behaviour, it can decrease manufacturing costs because less plastic can be used, and
388 also enables products with more environmentally friendly composite materials.

389 This research showed that the untreated woven sisal fibre reinforced post-consumer green
390 polyethylene composites can be used efficiently in structural engineering applications, such as
391 insulated panels, drywall, furniture, among others. Moreover, physical treatments on woven sisal
392 fibres will also be considered in future investigations, to improve interfacial adhesion between
393 phases and to avoid fibre pull-out and fibre-matrix debonding failure modes.

394

395

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