



Characterization of hybrid biocomposite Poly-Butyl-Succinate/Carbon fibers/Flax fibers

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ABSTRACT

The investigation of renewable and recyclable materials becomes more critical every day due to the high levels of waste and carbon emissions and their impact on the environment. The use of eco-friendly materials, such as natural fibers and bioplastics, is increasingly important, and their use is always more popular. The aim of this research is to evaluate the changes in properties of composite materials made of Poly-Butyl-Succinate (PBS), a biodegradable thermoplastic matrix, and carbon fiber when some layers of carbon fiber are replaced by some layers of flax fiber to create a hybrid composite; in order to obtain a material more environmentally friendly with similar mechanical properties. To modify the flax fiber's surface energy and improve the wettability with the PBS matrix, an Atmospheric Pressure Plasma Torch (APPT) treatment was performed. The fibers' surfaces were characterized by measuring the contact angle; the contact angle values confirmed the wettability and accordingly, adhesion increased after plasma treatment. Different experiments were performed after substituting carbon fibers to evaluate the changes in the composite material's mechanical and thermal properties: tensile test, three-point bending test, impact tests and differential scanning calorimetry. Replacing the carbon fiber core layer with one or two flax fiber layers did not compromise the thermal stability. It led to the manufacturing of a hybrid composite with improved mechanical properties and higher impact resistance.

1. Introduction

Composite materials play an essential role in many industrial sectors as automotive, energy [1], marine and aerospace industry [2]. These materials are attractive for different applications, not only for the lightweight but also for their endurance skills against the corrosion, chemical, and water impact [3]. In the recent decade, due to the increasing environmental and renewable green resources awareness, natural fiber reinforced polymer (NFRP) composites gained lots of attention from scientists for the next generation of composite products [4–6]. Despite the benefits of NFRP composites compared to synthetic fiber composites, such as better ecological performance, lower density, recyclability and renewability, they have some challenging issues such as poor mechanical performance and low compatibility of a matrix and natural reinforcement [7]. These weaknesses limit natural fiber composites' applications to structural components with minor mechanical requirements [8]. In recent years several modifications were introduced

to enhance the capability of natural fiber composites. One of the most promising solutions is combining natural and synthetic fibers in one composite. With this technique, it is possible to combine each fiber type's benefits in one hybrid composite. Over the years, glass fibers were used as a dominant synthetic fiber in hybrid composites owing to their high mechanical performance, high density, and low cost. Thus, hybridization of glass fibers with natural fibers resulted in cost-effective composites with improved mechanical performance-to-weight ratio [9–11].

On the other hand, hybridized composites with carbon fibers (CFs) are mostly used in applications where mechanical performance and low weight are more important than the cost. The focus of this research is on the hybridizing of carbon fiber composite with flax fiber (FF). Very few works regarding carbon/flax hybrid composites can be found within the last ten years. Among the natural fibers, flax fibers (*Linum usitatissimum*) have aroused great interest of many researchers due to their lower environmental impact, low cost, and better mechanical properties such

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as tensile strength (450–1500 MPa) and Young's modulus (27.6–38 GPa) [12–14]. In the year 2000, the FF was the most used natural fiber in the European automotive industry, with 71% of natural fibers consumption [15].

To cite some examples of research on carbon/flax hybrid composites, Fiore et al. [13] reported improving the tensile and bending properties of flax-epoxy composites by introducing carbon fabric layers. Assarar et al. [16] studied the effect of the stacking sequence of carbon/flax fiber reinforced epoxy hybrid composite on bending and damping properties. The effect of carbon/flax hybridization on mechanical and thermal properties as well as water absorption was explored by Dhakal et al. [17]. They reported significant improvement of tensile and flexural strength, water absorption behavior, thermal stability, and toughness properties. Al-Hajaj et al. demonstrated an increase in composite materials' energy absorption capacity by hybridizing carbon and flax fibers [18]. In another study, the addition of CF to flax fiber-reinforced polymer enhanced hydrothermal aging behavior [19] because CF has a better hydrothermal aging resistance compared to the natural fibers with hydroxyl groups that make them water-sensitive [20]. Ameer et al. worked on the effect of stacking sequence and fibers ratio of carbon/flax epoxy composites on static and tensile fatigue properties [21]. Kureemun and his team [22] reported significant improvement of flexural properties of flax-epoxy composite by the introduction of recycled short CF. Amiri et al. investigated a bicycle frame made of carbon/flax epoxy composite regarding vibration damping, stiffness, bending strength, and cost [23]. In addition to the last example, carbon/flax hybrid composites have received significant attention in sports applications. For example, Flemish Bicycle Company produced professional bike frames made of flax fibers (Flaxpreg® 2.0) and carbon fibers with minimum vibration [24]. Another recent application of carbon/flax composites is in the ecologic prototype boat named Araldite, which has introduced the smallest offshore racing boat [25].

The high moisture absorption of the FF influences the adhesion with the matrix; for this reason, chemical surface treatments, such as alkaline, silane, peroxide or plasma treatments, are necessary before manufacturing the composite materials [26]. In the present study, before preparing the composite material, the FF was treated by Atmospheric Pressure Plasma Torch (APPT) to modify the surface energy and improve the matrix adhesion. It is noteworthy that plasma treatment also decreases water absorption of flax fibers which was investigated in the previous studies of authors [27,28]. APPT produces dry and clean fibers' surface, just as it does on other polymeric surfaces, even on carbon fiber reinforced polymer (CFRP) [29,30]. Plasma treatment is a fast, nontoxic, dry, and environmentally friendly technique to alter the physicochemical nature of materials' surfaces without affecting bulk properties [31,32]. Plasma source includes energetic ionized gas produced by applying sufficient energy to the species existing in the gas [33]. The investigation on APPT treatment's effect was performed by measuring the contact angle [34].

High relative humidity and temperature concur with the fibers' degradation, which also compromises the material properties [35]. The degradation temperature of flax fibers has been reported around 319 °C [36]. Considering the degradation temperature of the FF, a thermoplastic polymer is required as a matrix. Thermoplastic materials could be melted more than once and recycled, unlike thermoset polymer. Thermoplastics have high impact resistance and are lightweight, easy to manufacture; however, they have lower heat resistance. In this paper, PBS, a thermoplastic polymer, is used as a matrix. It is possible to divide the plastic materials into four categories, depending on their origin and their biodegradation; for example, polylactic acid is biobased and biodegradable, and the bio-polypropylene is biobased but is not biodegradable. Examples of plastic polymer fossil-based and non-biodegradable are green polypropylene and polyethylene. PBS belongs to the fourth category, a plastic created from fossil-based materials but biodegradable [37]. PBS exhibits balanced performance in mechanical and thermal properties and processability from the

environmental and resource point of view compared to frequently used biodegradable polymers [38,39]. PBS which is a synthetic aliphatic polyester, is obtained by combining 1,4-butanediol with succinic acid and due to its tendency to hydrolyse it has a biodegradable characteristic [40].

This research evaluates the changes in composite materials' properties made of PBS matrix and carbon fiber when one carbon fiber layer is replaced by some layers of flax fiber to create a hybrid composite. To evaluate the effects of CF replacement with natural fibers, different characterization could be applied: tensile and flexural tests, which were performed in the previous experiments [41,42], as well as impact test to measure the mechanical properties [18,43]. Even thermal properties changes would be beneficial compared to pure synthetic composite and the hybrid composite [44,45]. Although the hybridization of flax and carbon fibers has been investigated recently, to the best of the authors' knowledge, research dedicated to hybridizing carbon/flax fibers with PBS matrix has not been reported elsewhere. This work's novelty is to employ a biodegradation PBS polymer for manufacturing hybrid composite by carbon fibers and flax fiber without changing thermal properties but improving mechanical properties.

2. Experimental procedure

2.1. Materials

Carbon fibers were produced by Materiales Estructurales Ligeros S.L. (Barcelona, Spain), bidirectional fabric with a mass per unit area of 600 g/m². Flax fibers were a unidirectional woven provided by Easy Composites (Staffordshire, United Kingdom) with a weight of 275 g/m². Flax, like other natural fibers, is not a homogeneous fiber with a constant section diameter and each fiber could vary from 12 to 100 μm. However, in this study, flax woven fabric is made of yarns (each yarn made of individual fibers) with a diameter of 0.1–0.3 mm (Fig. 1). The biodegradable PBS matrix that was synthesized from 1,4-butanediol and succinic acid or its anhydride in the presence of a catalyst [46] was supplied by TNJ Chemical Industry (Hefei, China).

2.2. Plasma treatment

To improve the adhesion between the matrix and the FF, an Atmospheric Pressure Plasma Torch (Plasma treat GmbH, Steinhagen, Germany) treatment was performed on the flax fabrics. As illustrated in Fig. 2, the APPT device consists of a discharge tube which is connected to three coaxial cylinders with decreasing diameters which is ended to the rotating nozzle (1900 rpm). The air gas is fed into the APPT from the gas inlet at the bottom of the discharge tube. Then air plasma was

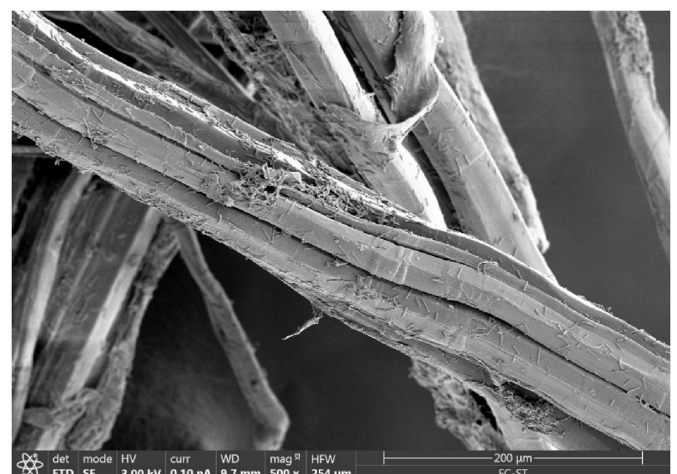


Fig. 1. SEM micrograph of flax fibers.

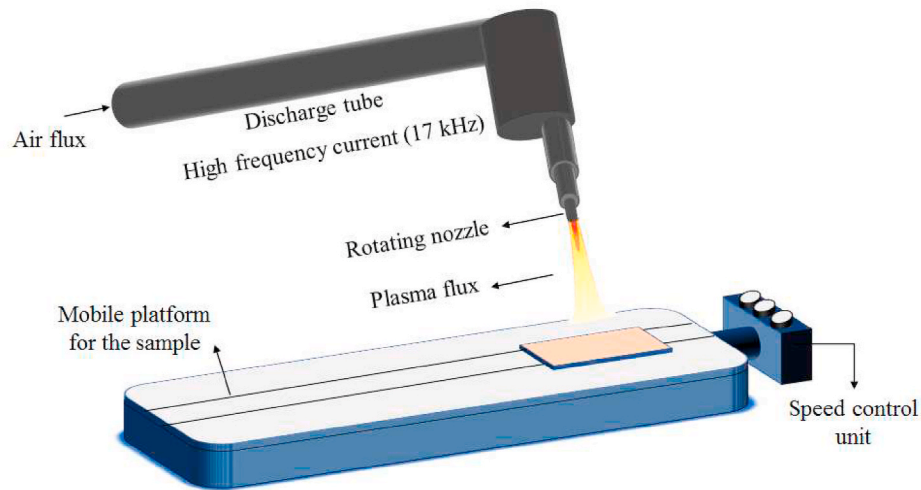


Fig. 2. Scheme of the APPT device.

generated at a working pressure of 2 bar by non-equilibrium discharge inside the rotating nozzle and expelled through a circular aperture onto the sample. The sample is located on the speed controller platform to move under the fixed nozzle. The setup operated at a frequency of 17 kHz and a high-tension discharge of 20 kV. The fiber-to-torch distance and platform speed were set at 20 mm and 2.5 m/min, respectively. The operation setup was according to the previous authors' work [27]. The carbon fibers exhibited an excellent adhesion with the matrix without plasma treatment.

2.3. Fiber-matrix interaction

2.3.1. Fiber-matrix wettability

The wetting behavior of a reinforcement fiber surface by a polymer matrix can be generally described by spreading of the matrix on the fiber surface which can be determined by contact angle [47]. The static contact angle of PBS matrix with flax and carbon fibers were determined with the sessile-drop method [48,49]. According to this technique, a pellet of PBS matrix was put on the surface of CF or FF. Then the temperature rises up in order to spread the polymer pellet on the surface. After obtaining stable spread drop on the fiber, the contact angle was measured. To evaluate CF-PBS wetting, the oven was heated to 115 °C, and the carbon fibers were inserted when the oven reached the right temperature for 40 min with a small PBS pellet on it. The contact angle between the CF and the matrix after cooling were measured by DataPhysics OCA15 plus goniometer and SCA20 software (DataPhysics Instruments GmbH, Filderstadt, Germany). The test was repeated five times. Regarding FF-PBS wetting evaluation, since the molten PBS passed through FF after 40 min, it was impossible to do the same procedure as for the CF. In this regard, Kruss contact-angle system (GmbH, Germany), which was equipped with a camera and a thermal chamber, was chosen to follow contact angle by increasing temperature with a heating rate of 5 °C/min. The different parameters that had to be set up were the final temperature (115 °C) and the heating rate.

2.3.2. FF wettability and surface energy

In order to evaluate the effect of plasma treatment on the flax fibers, their wettability was measured using three liquids with different polarity (water, diiodomethane, and glycerol) before and after APPT treatment. In this regard, four drops of each liquid with a volume of 6 μ l were filed on the fiber's surface. Then, the surface energy (SE) was calculated by the Owens-Wendt-Rable-Kaelble method [50]. With this method, both the dispersive and polar components of the surface energy can be identified. Prior to the surface energy measurement, the contact angle of liquids with FF was measured using the same system for the

CF-PBS adhesion.

2.4. Composite material preparation

PBS sheets were prepared by a hot plates press machine (Fontune Presses TPB374, Barendrecht, Netherlands). PBS pellets (20 g) were placed between the hot press plates with a specific program. The maximum temperature was 130 °C and it was chosen considering the melting point of the PBS matrix (106 °C). Actually, the max temperature of hot-press is higher than the melting point of PBS to increase the impregnation of fibers with matrix. Also, It was experimentally defined that more than this temperature (130 °C), the flax fibers degraded. The second step was inserting the fiber layers ($15 \times 15 \text{ cm}^2$) between the matrix sheets. The thickness of one ply for each carbon and flax fibers is 0.60 and 0.35 mm respectively. There is a PBS layer between every layer of fibers, and always the composite outer layers are PBS, as it can be observed in Fig. 3. The same program applied for preparing the PBS sheets was used to prepare the final composite materials in the last step. Three different composite materials were prepared as follows:

- 1) Three layers of CF (3CF)
- 2) Two layers of CF and one of FF replacing the central layer of CF (2CF/1FF)
- 3) Two layers of CF and two of FF replacing the central layer of CF (2CF/2FF)

Their density, thicknesses and fiber contents are given in Table 1. Moreover, the volume% of voids (V_v) in each composite are calculated according to ASTM D2734, based on Eq. (1) [51] which are also presented in Table 1.

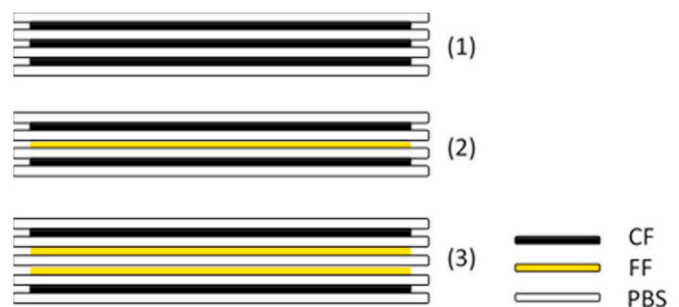


Fig. 3. Composites configurations: (1) PBS-3CF, (2) PBS-2CF/FF, (3) PBS-2CF/2FF

Table 1

Thickness, density, fiber content and void percentage of the PBS-3CF, PBS-2CF/FF and PBS-2CF/2FF composites.

Composite	P (g/cm ³)	CF wt %	FF wt %	PBS layers	Void %	Thickness (mm)
3CF	1.36	64	0	4	27.34	2.01 ± 0.05
2CF/1FF	1.38	41	9	4	14.95	1.91 ± 0.05
2CF/2FF	1.44	41	18	5	12.73	2.14 ± 0.02

$$V_v = 100 - \rho_{composite} \left(\frac{\% \text{ wt. matrix}}{\rho_{matrix}} + \frac{\% \text{ wt. fiber}}{\rho_{fiber}} \right) \quad (1)$$

2.5. Mechanical properties

2.5.1. Tensile test

The tensile test was performed by the Universal Hydraulic Tensile Test Machine (Ibertest IBMT4, Madrid, Spain), according to the ASTM D3039 standard [52]. Five specific samples of each composite material were cut to be tested according to the standard specimen dimensions (Fig. 4). The samples thickness was between 1.7 mm and 2.1 mm, depending on the number of layers. Tests were carried out with a load cell of 20 kN, and the test speed was set at 5 mm/min. Moreover, to prevent slippage of tensile samples at the grip section, they were fixed between the tabs made of glass/epoxy composite with cyanoacrylate as an adhesive. In addition, prior to applying adhesive, the plasma was employed on both the tabs and end parts of the samples.

Tensile strength (σ) and modulus (E) of the hybrid composites were calculated according to Eqs. (2) and (3):

$$\sigma = \frac{F}{A_0} \quad (2)$$

$$E = \frac{\Delta\sigma}{\Delta\epsilon} \quad (3)$$

where F is the applied force and A_0 is the initial cross-section of the test specimen. $\Delta\sigma$ and $\Delta\epsilon$ are the stress and strain increment of the linear part of the tensile curve respectively.

2.5.2. Bending test

Bending tests were performed through a three-point bending device placed on the same electromechanical test machine where tensile tests were performed (Microtest EM2/FR, Madrid, Spain) with a speed of 5 mm/min, grips distance of 50 mm, and a load cell of 20 kN as specified in ASTM D7264 standard [53]. The samples dimension for this experiment were 80 × 10 cm with their respective thickness depending on the number of layers. Bending tests were carried out on five samples. Bending strength and elongation were determined using Eqs. (4) and (5):

$$\sigma_f = \frac{3FL}{2bh^2} \quad (4)$$

$$\epsilon_f = \frac{6\Delta b}{L^2} 100(\%) \quad (5)$$

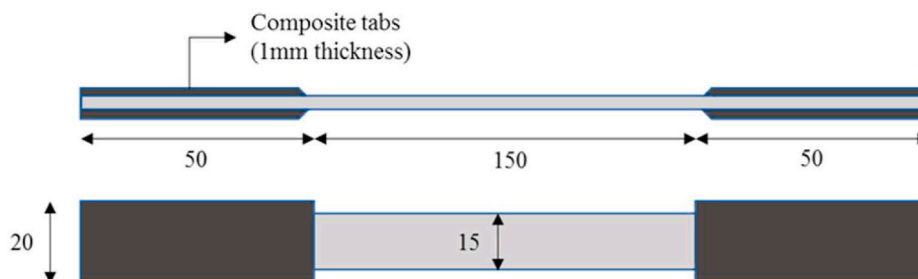


Fig. 4. Tensile specimen test configuration according to the ASTM D3039 (dimensions are in mm).

where F is the applied force, L is the distance between the grips, b is the width, h is the thickness, and s is the vertical displacement.

2.5.3. Impact test

Charpy pendulum CEAST 9050 (Instron, Norwood, MA, US) was used to perform impact tests. A hammer impacts the samples when it reaches the lowest point of its trajectory. It is possible to measure the energy absorbed by the sample during the impact through the pendulum. The Charpy test was carried out according to the ISO 179-1:2011 standard on five samples with the same dimensions as the flexural ones [54]. Although a notch is usually made before the impact test to create a stress concentration point, this was not possible due to the composite's characteristics. The absorbed energy per impact initial cross-section surface (S_0) provides Charpy impact strength according to Eq. 6

$$\text{Charpy impact strength} = \frac{\text{Absorbed energy (kJ)}}{S_0 (m^2)} \quad (6)$$

2.6. Differential scanning calorimetry (DSC)

DSC 822 Mettler Toledo (Greifensee, Switzerland) was used to evaluate composites' thermal properties. The test was performed on samples of 8–12 mg placed in an aluminum crucible with a capacity of 40 μ l and heated from 25 °C to 200 °C with a rate of 10 °C/min. Nitrogen was used with a feeding rate of 50 ml/min as a purge gas. With DSC analysis, it is possible to evaluate the thermal stability, melting point, enthalpy, and crystallinity degree.

The degree of crystallinity was estimated with Eq. (7) [55]:

$$X_c = \frac{\Delta H_m}{\Delta H_m^0 \times w} \times 100\% \quad (7)$$

where X_c is the degree of crystallinity, ΔH_m is the melting enthalpy of composite, ΔH_m^0 is the melting enthalpy of the 100% crystalline PBS (110.3 J/g) [56], and w is the weight fraction of matrix.

3. Results and discussion

3.1. Fiber-matrix wettability

The average contact angle value between CF and PBS was 27.1° (± 0.6); thus, CF has an excellent wettability with the PBS (Fig. 5), no more treatments were required to improve the adhesion. The contact angle between the flax fibers and the PBS matrix was evaluated before and after the plasma treatment. Table 2 shows the average contact angles for untreated and treated flax fibers at three different temperatures (60, 80, and 115 °C). Moreover, Fig. 6 exhibits the configuration of a PBS drop on the flax fibers in different conditions.

At 60 °C, the contact angles of treated and untreated fibers are in the same range because PBS has a high dispersive component of surface energy [57]. Furthermore, even if at 60 °C, the chain started to move, increasing wettability is required a more elevated temperature [58]. At

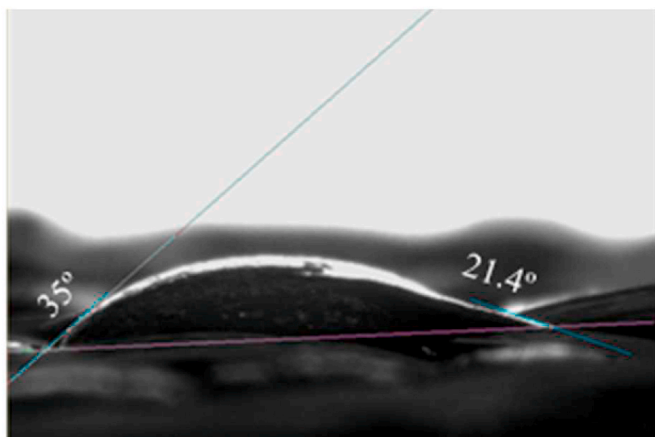


Fig. 5. Drop of PBS on CF fabric.

Table 2
Contact Angles of PBS on untreated and treated FF.

	Temperature (°C)	Contact angle (°)	
		Untreated	APPT treatment
PBS_FF	60°	104.8 ± 2.8	104.6 ± 1.9
	80°	80.6 ± 2.3	50.5 ± 3.4
	115°	67.5 ± 5.6	12.4 ± 0.5

80°, there is a significant difference in contact angle of treated and untreated fibers. This value is lower for treated fiber than the untreated one. At 115 °C, the contact angle value of the treated FF and the droplet picture shows PBS has thoroughly wetted the FF surface, which means that the wettability at 115 °C between PBS and the flax fibers is good [59]. As seen in Table 2, the effect of APPT is more significant at higher temperatures. This refers to the function of plasma. Plasma treatment by non-polymerizing feeding gases like helium, argon, oxygen, air, and nitrogen induces chain scission on the surface of fibers [60]. It has been reported that in the case of flax fibers, these chains consist of oxygen-containing groups that activate the flax fiber surface [60–62]. By increasing the temperature, the interaction of these oxygen groups with the surrounding polymer matrix will be promoted, accordingly effect of APPT is more visible at higher temperatures.

Besides, based on the established correlation between contact angle and interfacial adhesion [63], it is possible to estimate the effect of

wettability improvement on the adhesion of fibers and matrix. This correlation, which has good agreement with the model of Nardin and Schultz [64–66] for prediction of interfacial shear strength, indicates that interfacial adhesion between matrix and fibers increases as the contact angle decreases. Thus, it can be concluded that by decreasing the contact angle of FFs and PBS, their adhesion would be improved.

3.2. Flax fibers SE and wettability

The contact angle of each liquid on flax fibers is presented in Table 3 before and after plasma treatment. After APPT, contact angles decreased for all polar and non-polar liquids. This specifies that the polarity of the treated fibers is successfully reduced, and flax fibers could be wetted and exhibited more hydrophilic behavior after APPT.

The calculated surface energies of flax fibers as well as their dispersive and polar components are presented in Table 4. After APPT treatment, the surface energy decreased, and the polar component of the energy increased, which improves the adhesion between the fiber and the matrix.

The primary wall or the outer layer in the flax fiber cell structure is wax. This layer with 0.2 μm thickness makes up 1.7 wt% of flax fiber [67,68]. The Wax layer, which affects the flax fiber wettability and adhesion characteristics, is degraded by APPT treatment [69–71]. Removal of non-cellulose compounds like wax and also contaminant substances such as dyes, ashes, and fatty oils that cover the fiber surface and hinder the adhesive process, decrease surface energy and facilitate the mechanical interlocking and adhesion between fiber and matrix [62]. Since the wax is a weak cohesive layer and is less polar compared to the underneath layers in the flax structure, by removing it, the surface polarity and wettability would be improved, which can be proven by higher polar surface energy in Table 4.

Moreover, the X-ray photoelectron spectroscopy (XPS) analysis conducted in the previous study of authors [34] on treated flax fibers confirms these obtained results. Based on the percentage of the elements

Table 3
Contact angles of untreated and treated FF with water, diiodomethane and glycerol.

Liquid	Contact angle (°)	
	Untreated	APPT treatment
Water	123.4 ± 1.4	83.5 ± 0.7
Diiodomethane	41.5 ± 4.1	10.0 ± 0.5
Glycerol	114.2 ± 1.3)	90.1 ± 2.3

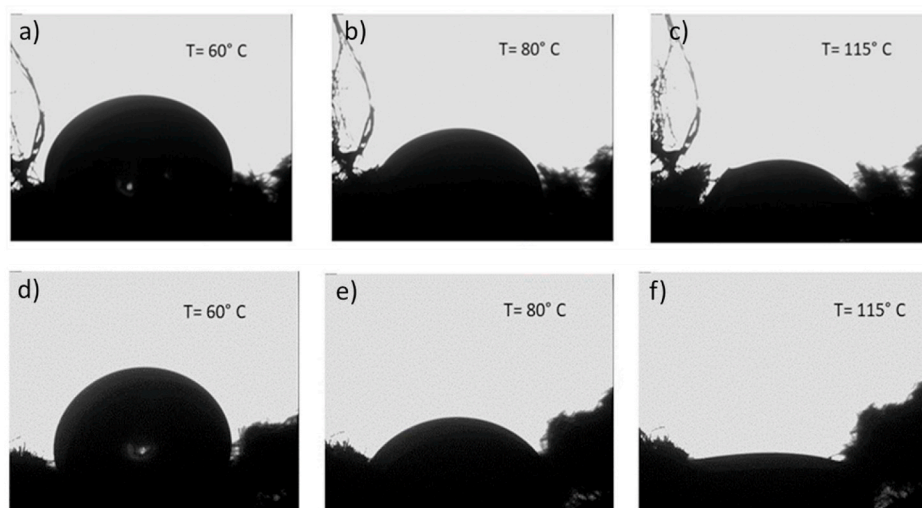


Fig. 6. The contact angle of FF/PBS before (a–c) and after APPT (d–f).

Table 4

Surface energy and its dispersive and polar components of untreated and treated FF.

Samples	SE (mN/m)	Dispersive SE (mN/m)	Polar SE (mN/m)
FF	44.79	41.74	3.05
FF + APPT	30.24	22.71	7.53

presented in Table 5, both treated and untreated fibers' surface mainly consists of carbon and oxygen with negligible amounts of nitrogen. After plasma treatment of flax fibers, the amount of oxygen increased while the amount of carbon decreased. In other words, the ratio of O/C and N/O increased. Therefore, plasma treatment did not affect the surface composition. Both treated and untreated fibers' surface consist of C–C/C–H, R–C–O and R–O–C=O chemical groups. However, the percentages of chemical groups changed after plasma treatment. For example, although the C% decreased, the bond C–C/C–H increased because of water loss during the treatment. In this regard, the corresponding bonds to the water absorption disappeared and carbonyl and C–N polar groups increased, which resulted in higher polar surface energy and better adhesion of fibers to the matrix.

3.3. Mechanical properties

3.3.1. Tensile test

Before performing the tensile test, the weight and volume of specimens were measured to calculate each composite density. Typical tensile stress-strain curves of manufactured composites are plotted in Fig. 7. It can be seen that the tensile properties of PBS-3CF composite were influenced by hybridization with flax fibers. The average values of tensile modulus and strength for each composite are reported in Fig. 8. To better compare modulus and densities between composites, the specific modulus is also calculated and presented with densities in Table 6.

After the replacement of one FF with CF, both tensile strength and tensile modulus increased. However, adding the second FF decreased the tensile properties. The reason can be considered from different aspects. Firstly, according to the fiber-reinforced polymeric matrix theory, the composite modulus can be enhanced since the load applied to the matrix is transferred to fibers during the tensile process. Results indicate that in 2CF/1FF composite, hybridization of natural fibers with carbon fibers has facilitated the load transfer phenomenon. Nevertheless, in the 2CF/2FF composite, because of the lower modulus of flax fiber (27–65 GPa) with respect to the carbon fibers (200–400 GPa), the second FF layer has reduced the modulus and strength of the composite. This is aligned with what has also been reported by literature [15,17,72] that an increase in FF content in hybrid composites leads to a reduction of tensile strength and modulus, mostly due to the lower tensile stiffness of FF with respect to the CF.

Secondly, lower void% of hybrid PBS-2CF/FF composite with respect to the PBS-3CF composite (Table 1) would be another reason for better tensile properties because voids act as stress concentration points and decrease mechanical properties. In the case of PBS-2CF/2FF composite, although the void% is lower than PBS-3CF composite, the tensile strength is lower due to the higher thickness of PBS-2CF/2FF composite. Higher thickness increases the area under load in the tensile test and accordingly decreases the strength in the case of the same applied load.

Another affecting parameter in tensile and modulus of hybrid fiber-reinforced composites is the laminate configuration. Since the carbon

Table 5

XPS compositional analysis for plasma-treated and untreated flax fibers.

	C (%)	O (%)	N (%)	O/C (%)	N/C (%)
Untreated FF	85.6	12.9	1.5	15.0	2.0
Treated FF	80.9	16.1	3.0	20.0	4.0

fibers have a lower failure strain than flax fibers [73], the carbon fibers in hybrid composite reach failure first. In this regard, the alternating arrangement of carbon and flax fibers (PBS-2CF-1FF) is more favorable because broken carbon fibers could be bridged by adjacent flax fibers, delaying the expansion of failed parts [22,73].

In order to validate calculated values, Fig. 8 is also included a study of Liang et al. [74] on PBS-CF (80/20 wt%) composite as a reference. As shown in Fig. 8, with respect to the reference composite, manufactured PBS-3CF composite demonstrates not only significant improvement of strength and modulus with increasing of wt% of CF but also more effective used manufacturing technique.

According to Fig. 9, tensile samples could not be broken during tests completely; there is a partial break of CF and also some delamination of flax and carbon fibers because of the larger failure strain of flax fibers than carbon fibers. Thus, after the tensile test, composites were first freed by liquid nitrogen and then cut with special scissors with micro-tooth to be studied. SEM observations have been performed on the cross-section of composites after tensile loading, as shown in Fig. 10a–f. First of all, the difference between the fibers' morphology is evident in Fig. 10b and d. The CFs have rounded and circular cross-section with respect to the bundled and more flat morphology of flax fibers. It can be seen that the PBS matrix has impregnated the fibers and also penetrated within the fiber layers. In Fig. 10d, the dashed lines are guidelines for FF and matrix interface. Continuity of PBS polymer around the flax fibers indicates the good compatibility between the natural fibers and the matrix, which resulted from APPT treatment on flax fibers.

Moreover, in Fig. 10e and f, different damage types can be detected: fibers pullout and breakage, with some voids left by the pulled-out fibers. The pulled-out fibers are clear of PBS, which indicates the adhesion of matrix and fibers was not enough under the tensile load. This effect was due to the fact that each fiber is made up of numerous filaments. The APPT treatment affects the superficial filaments but not the internal ones; consequently, the union does not become intimate between the matrix and the filaments.

3.3.2. Flexural behavior

Fig. 11A and 10B represent flexural strengths and modulus. The 2CF/1FF is the composite material with the highest values. The substitution of one layer of CF by one or two FF layers made the composite material more resistant per unit area when the samples are subjected to bending. The load per unit area has not increased for the 2CF/2FF composite because of more PBS layers. The flexural strength of 2CF/1FF composite with four PBS layers improved by 18%, while the improvement of 2CF/2FF with five PBS layers was 10%. This less improvement might be due to two reasons. First, an additional thermoplastic matrix layer decreases the composite mechanical properties, as was mentioned before. Second, the flax fibers are unidirectional, which caused them to wave slightly during the hot press process compared to the bidirectional carbon woven. The flax fiber waviness decreases the flexural strength, and when there are two flax fibers, this effect is intensified.

Flexural modulus is similar for the three composites (Fig. 11B) since the values overlap, although there is a slight decrease for the composite with 2CF/2FF. This small decrease is practically not appreciated in the representative flexural stress-strain curve of Fig. 11C.

3.4. Impact properties

The Charpy impact strength of composites (kJ/m^2) is shown in Fig. 12. Results show an increase in the absorbed energy with the increment in natural fiber content. The hybrid composite with two FF layers absorbed more energy per m^2 than the others because natural fibers are tougher and absorb more energy [9,75]. Moreover, pulling out of flax fibers from the matrix (as seen in Fig. 10c–f) without breaking due to their compliance leads to improvement in impact absorption and dissipation energy during the impact testing [9,76]. Consequently, the substitution of one CF layer by two FF layers has improved the impact

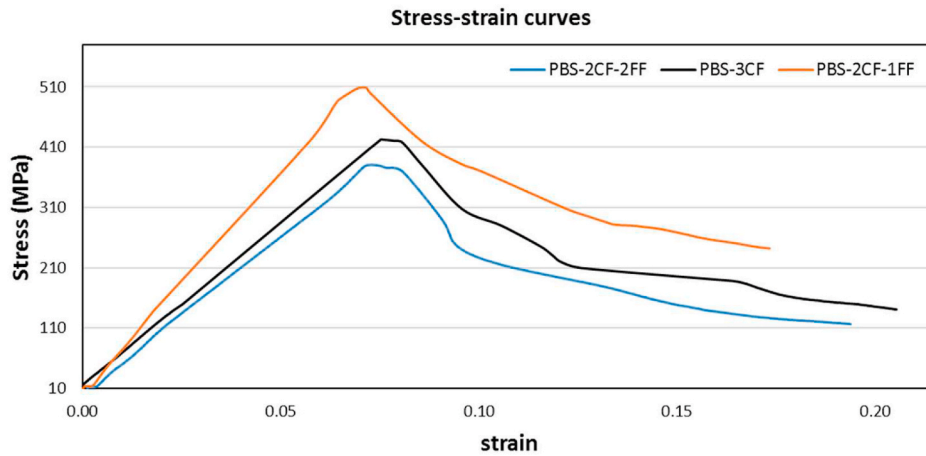


Fig. 7. Stress-strain curves of manufactured composites after tensile test.

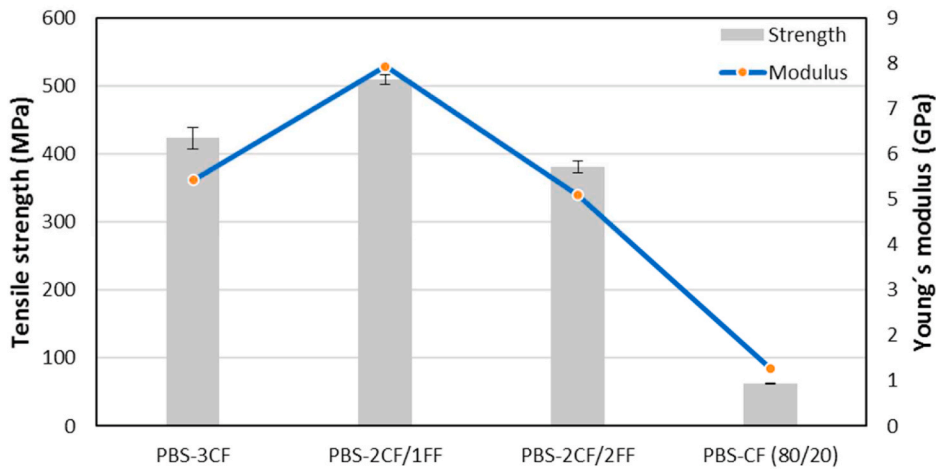


Fig. 8. Young's modulus of PBS-3CF, PBS-2CF/FF and PBS-2CF/2FF composites and PBS-CF (80/20 wt%) reference composite.

Table 6

Density and specific modulus of manufactured PBS-3CF, PBS-2CF/FF and PBS-2CF/2FF composites.

Composite	ρ (g/cm ³)	Specific Modulus (GPa. Cm ³ /g)
PBS-3CF	1.64 ± 0.19	3.30
PBS-2CF/1FF	1.48 ± 0.06	5.36
PBS-2CF/2FF	1.36 ± 0.05	3.73

strength by a 30%. However, for the 2CF/1FF composite, the impact strength is similar to the 3CF composite and just increased 5%. Furthermore, one more thermoplastic matrix layer in the 2CF/2FF composite contributed to increasing the impact resistance [37]. These results have been obtained previously in another experiment with natural fiber-reinforced composites [77]. Another reason for the increase of impact strength might be a combination of failure modes during the impact, such as ply delamination and fiber breaking, supporting dissipating energy during an impact [72].

It seems that in the 2CF/2FF composites structure, both the carbon skins and flax core seemed to be damaged. However, carbon skins have a greater strength against impact load, and the flax core has better flexibility and deformability. As a result, this composite has a higher absorbed energy and impact strength. The composite samples did not break during the impact test.

It is worth mentioning that achieved impact strength for PBS-3CF composite is significantly higher than previous values reported by Li

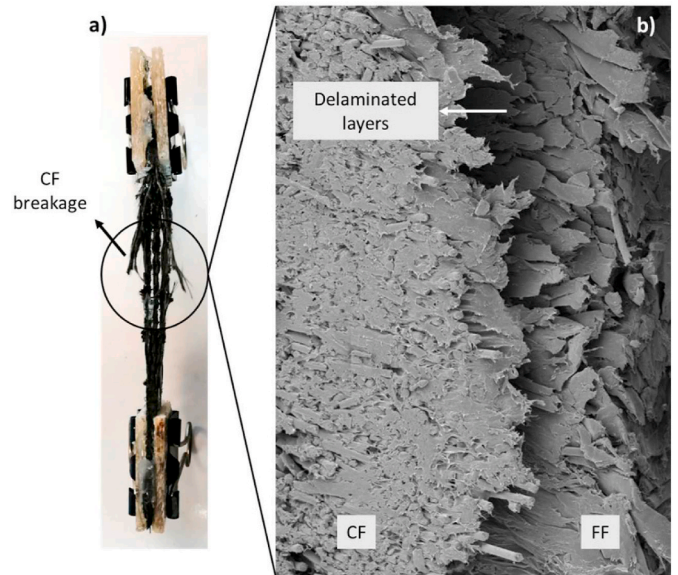


Fig. 9. a) A composite sample after tensile test, b) SEM picture of delaminated layers.

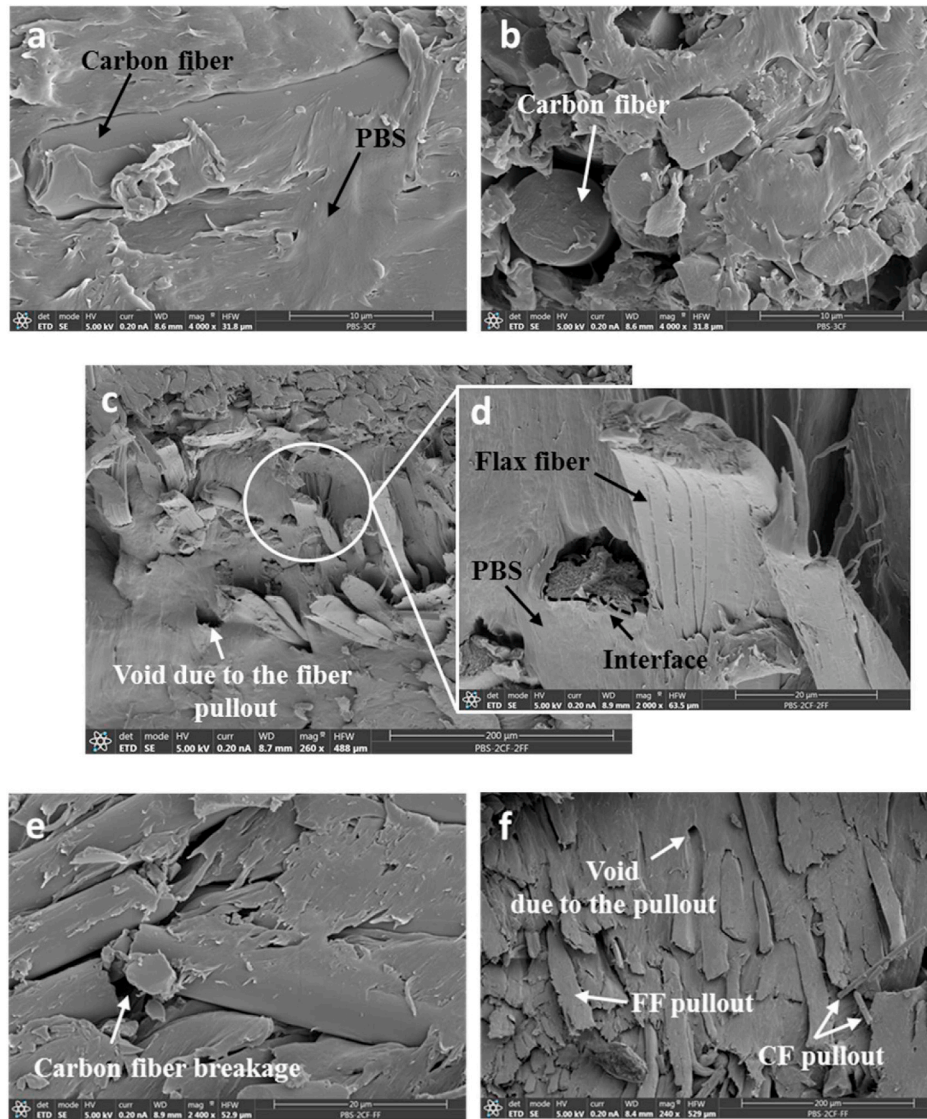


Fig. 10. SEM cross-section micrographs of manufactured composites after tensile test: (a,b) 3CF composite, (c,d) 2CF/2FF composite, (e,f) 2CF/1FF composite.

and Qu [78] (6.9 kJ/m^2 , 393% improvement) and Jicai et al. [74] (11.4 kJ/m^2 , 198% improvement) for the PBS/CF composites. In these references, composites with lower wt% of CF were fabricated with the extrusion technique. It can be concluded that altering manufacturing technique to hot press and also higher wt% of CF with lower ductility have not constrained the PBS matrix and resulted in enhancement of impact strength.

3.5. Differential scanning calorimetry (DSC)

The thermal stability of the three composite materials was evaluated by DSC. As shown in Fig. 13, two different peaks were observed for each composite. The sharp endothermic peaks characteristic of melting and small exothermic peaks assign to the recrystallization process that PBS undergoes during heating since there was T_g (glass transition temperature) at $36 \text{ }^\circ\text{C}$ that corresponds to the amorphous phase [79]. Melting temperature (T_m), recrystallization temperature (T_c), melting enthalpy (ΔH_m), and degree of crystallinity (X_c) that measured based on Eq. (7) are shown in Table 7. The variation in T_m of the composites is not significant. For the 3CF composite, the T_m appeared at $106 \text{ }^\circ\text{C}$. In comparison, the 2CF/1FF and 2CF/2FF composites appeared at $108 \text{ }^\circ\text{C}$, indicating that FF increased entanglement between the molecular chains

of PBS and accordingly increased T_m of composites. The ΔH_m was calculated by the integration of the area under the peak. The enthalpy increased with the FF amount, which means that the natural fibers do not compromise the material thermal properties. The T_c slightly shifted to higher temperatures by replacing CF with FF, indicating that the flax fibers acted as nucleating agents during the matrix recrystallization. As a result, the X_c of PBS/3CF composite increased by 21% and 31% in PBS-2CF/FF and PBS-2CF/2FF composites, respectively. Since the crystalline phase tends to increase stiffness and tensile strength, the PBS-2CF/FF and PBS-2CF/2FF composites with a higher degree of crystallinity exhibit better mechanical properties, as it was shown in part 3.3 and 3.4. Moreover, compared to the pure PBS matrix, the hybrid composites with flax fibers have better thermal properties with respect to the 3CF composite.

4. Conclusion

For the preparation of hybrid composite materials, natural fibers are an excellent alternative to substitute synthetic fibers in terms of availability, price, carbon emissions, and recyclability. In this study, three series of CF reinforced PBS composites were prepared, and two of them were hybridized with FFs. The APPT treatment improved the wetting

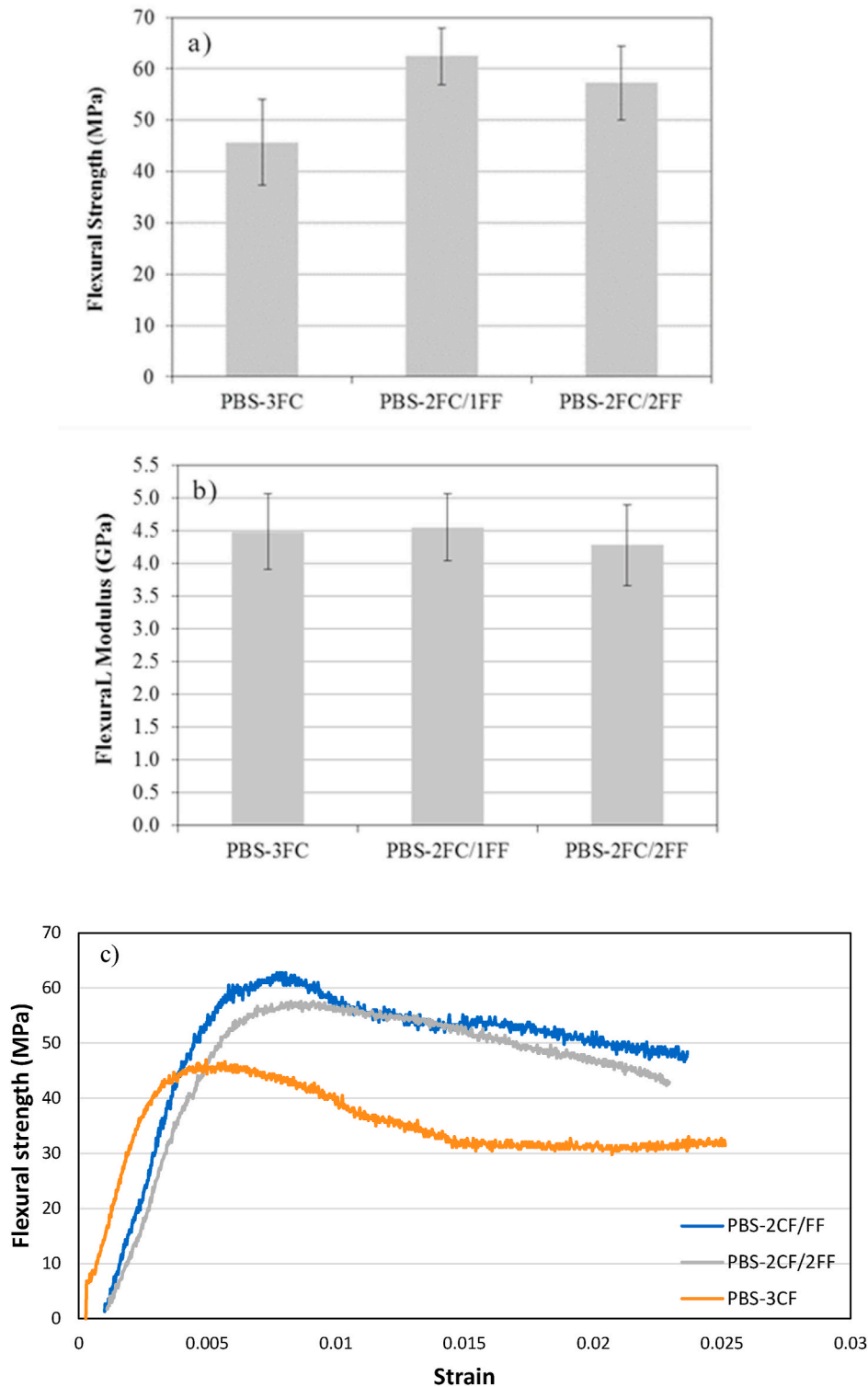


Fig. 11. a) Flexural strength, b) Flexural modulus, and c) Curves of flexural stress-strain for composites.

property of PBS matrix and natural flax fibers and, consequently, their adhesion, which was the major drawback of natural fibers. The characterizations made up by the tensile, three-point bending, and impact tests showed how the flax fibers do not weaken the mechanical properties. On the contrary, the substitution of one CF layer with FF layers resulted in a hybrid composite with superior tensile and flexural properties as well as impact strength. DSC measurements displayed how the FF's presence does not compromise the thermal stability in any composite materials. It is possible to build a lighter hybrid composite

material from natural fibers, i.e., FF, and a biodegradable matrix, i.e., PBS, without compromising the thermal and mechanical properties. These composite materials are an exploitable option for all applications that need lightweight and impact-resistant materials.

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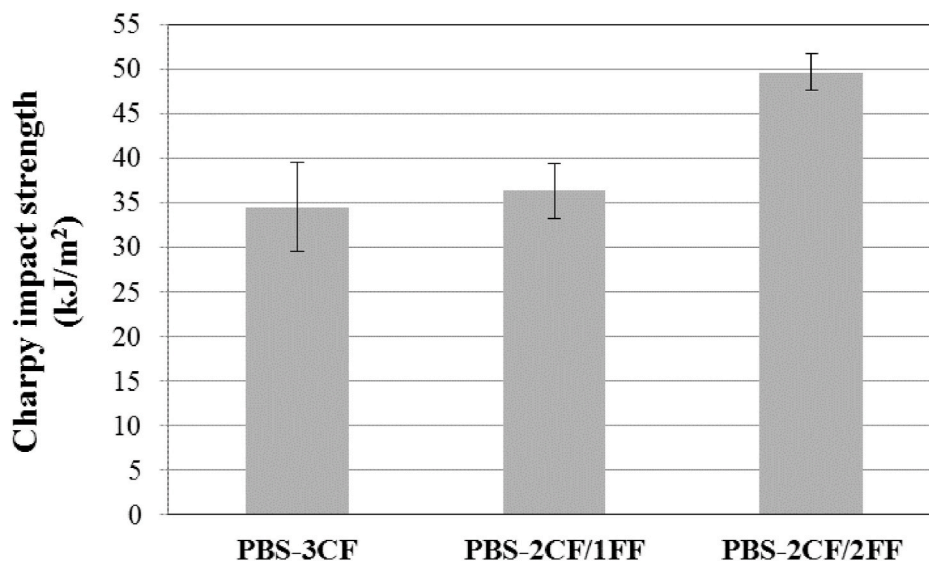


Fig. 12. Charpy impact strength (kJ/m²).

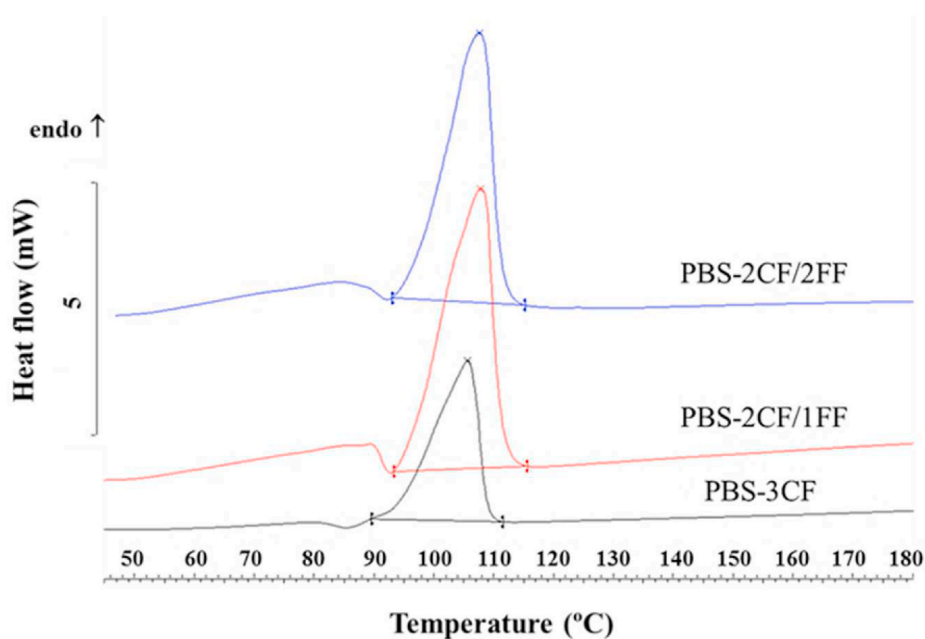


Fig. 13. DSC thermograms for the PBS-3CF, PBS-2CF/FF and PBS-2CF/2FF composites (heating rate:10 °C/min).

Table 7

Thermal properties of studied materials by DSC.

Composite	T _c (°C)	T _m (°C)	ΔH _m (J/g)	X _c (%)
PBS matrix	92	106	38	35
PBS-3CF	86	106	20.25	30.68
PBS-2CF/1FF	93	108	25.82	37.16
PBS-2CF/2FF	93	108	28.78	40.24

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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