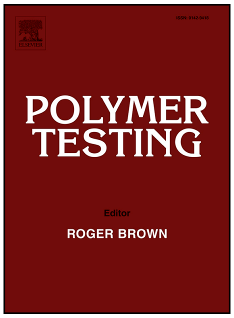

POLYMER TESTING

Editor

ROGER BROWN

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PII: S0142-9418(15)00141-5

DOI: 10.1016/j.polymertesting.2015.06.009

Reference: POTE 4455

To appear in: *Polymer Testing*

Received Date: 12 May 2015

Accepted Date: 20 June 2015

Please cite this article as: E. Frollini, N. Bartolucci, L. Sisti, A. Celli, Biocomposites based on poly(Butylene succinate) and curaua: Mechanical and morphological properties, *Polymer Testing* (2015), doi: 10.1016/j.polymertesting.2015.06.009.

Material Properties

BIOCOMPOSITES BASED ON POLY(BUTYLENE SUCCINATE) AND CURAUA: MECHANICAL AND MORPHOLOGICAL PROPERTIES

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ABSTRACT

Biocomposites based on poly(butylene succinate) (PBS) and curaua fibers have been produced by compression molding, and investigated as a function of fiber length and amount. Mechanical tests, water uptake and morphology studies were carried out in order to assess the composite features according to the characteristics of the reinforcing agents. It turns out that the impact and flexural strengths increase with fiber content. Moreover, the fiber length, varying from 1 to 4 cm for the composite reinforced with 20 wt% of fiber, influences impact strength, which is higher for shorter than for longer fibers. However, flexural strength is not greatly influenced by the length of the fibers. Water uptake studies reveal a higher sensitivity of the material to fiber content rather than fiber size. Biocomposites, which are characterized by enhanced mechanical properties as compared to PBS, can have different applications, for example in rigid packaging or interior car parts.

Keywords: Poly(butylene succinate) (PBS); Lignocellulosic fibers; Biocomposites; Curaua

1. Introduction

The current social awareness of environmental pollution and the need to find polymers that can substitute petro-derived plastics, also due to the oil crisis, have fostered the search for eco-friendly materials, e.g. biopolymers and biocomposites [1-6]. Thus, in the last few decades, polymers reinforced with lignocellulosic fibers have rapidly gained attention due to several advantages [5-7]. Natural plant fibers being renewable materials, they are currently exploited thanks to their biodegradability, high availability, low energy consumption and cheapness. However, fibers also feature other specific and less exploited properties, such as acoustic and thermal insulation, non-abrasiveness for processing tools and low density [3,6-11]. Such features make vegetable fibers very attractive to replace their synthetic counterpart, i.e. glass fibers, in traditional polymeric matrix composites [4,6,7,11]. However, they also have undeniable drawbacks such as low thermal stability, which limits the choice of polymeric matrices [4,7,9], and hydrophilicity, which causes low compatibility with hydrophobic polymers [4,8,9,11,12] and leads to poor resistance against moisture uptake, irreversibly affecting the mechanical, physical and chemical properties of the composite [5,13,14]. Moreover, physical and chemical features of lignocellulosic fibers differ highly within the same species depending on soil, harvesting time, extraction methods and climatic, growing and processing conditions [11,15-17].

Poly(butylenesuccinate) (PBS) belongs to the family of aliphatic polyesters (Fig. 1) and is produced by a two stage polycondensation starting from 1,4-butanediol and succinic acid. Today, the monomers have fossil origin, but their industrial production by biomass fermentation, which will contribute to environmental impact decrement, is in progress [18]. PBS is a semicrystalline polymer with good processability. The melting temperature is around 115 °C and the glass transition temperature occurs at about -30 °C; the tensile strength is similar to those of PE and PP while the stiffness is between LDPE and HDPE [8]. This polymer offers a wide range of applications, going from packaging to flushable hygiene and mulch products. Moreover, PBS is biodegradable due to its aliphatic structure and the presence of the ester bonds which deteriorate in compost, moist soil, fresh water with activated sludge and sea water [19].

Despite all these attractive features, this polyester has not been extensively used mainly because of its high price. Recent works have dealt with PBS reinforced with natural plant fibers [20-31]: the aim being to increase the performance of the biopolymer while reducing the weight of the final material with further economic advantages, as part of the polymer is replaced by less expensive materials. In addition to the properties of PBS already mentioned, the presence of polar groups (carbonyl groups) and nonpolar segments in its structure (Fig. 1) makes it attractive for applications as the matrix in composites reinforced with lignocellulosic fibers, whose main components (lignin, hemicellulose and cellulose) also have polar and apolar groups in their structures. These characteristics favor the interactions at the fiber - matrix interface.

In a previous study, we explored the potential of different lignocellulosic fibers (sugarcane bagasse, coconut, curaua and sisal) to reinforce the PBS matrix [32]. The present paper will further analyze the influence of length and amount of curaua fibers on the final properties of PBS-based composites. Indeed, to the best of our knowledge, no paper has been published so far on this combination of matrix and fibers.

Curaua (*Ananas erectifolius*) belongs to the Bromeliaceae family and is largely cultivated in the Brazilian Amazon region. The fibers extracted from the plant are lignocellulosic material, composed mainly of three fractions: cellulose, hemicelluloses and lignin [33-36]. Their tensile and flexural strength closely resembles that of glass fibers and, unlike the latter, curaua fibers are biodegradable [35]. Therefore, curaua fibers are a very promising material for the preparation of biocomposites: the replacement of glass fibers with curaua fibers in thermoplastic composites has been reported, especially for automotive devices [6, 35].

In this work, biocomposites were produced via thermo-pressed molding, which reaches a maximum processing temperature sensibly lower than the beginning of thermal degradation of curaua fibers. The final properties of the biocomposites have been discussed in terms of thermal behavior, mechanical performance, morphology and water absorption.

2. Experimental

2.1. Materials

Curaua fibers were kindly donated by Pematec Triangel do Brasil Ltda (São Paulo, SP, Brazil) while polybutylene succinate (Natureplast PBE003) was supplied by Natureplast.

2.2. Fiber and polymer characterization

In order to improve the adhesion between the lignocellulosic material and the polymer, the fibers were subjected to an extraction process using a mixture of ethanol and cyclohexane (1:1 v/v, 10 min, reflux) to remove the substances responsible for the poor wettability, e.g. waxes, terpenes, fatty acids [37,38]. Afterwards, they were washed with water and dried in an air-circulating oven at 45 °C overnight and later at 105 °C for 4 hours. The chemical characterization and crystallinity index, $I_{c,}$ was carried out as described by Paiva and Frollini [10,32,39].

The thermal stability of curaua fibers, PBS and composites was determined as described elsewhere [31].

2.3. Morphological characterization

The morphological characterization of the composites was performed by SEM, Zeiss DSM 940, 440, electron acceleration 20 kV. All the samples were previously coated with an ultrathin coating of gold in a sputtering coating system.

2.4. Composite preparation

A mechanical mill was used to crush the PBS pellets: the powder was further dried at 80 °C in an air-circulating oven overnight. Compression molding was employed to manufacture randomly oriented fiber reinforced composites. The composite samples were prepared first by varying the fiber amount (10, 20 and 30 wt.%) and keeping their length constant (3 cm), then they were prepared with different fiber lengths (1, 2, 3 and 4 cm) while their percentage was fixed at 20 wt.%. The mold, filled with the samples, was heated up from room temperature to 50 °C and kept there 15 minutes, afterwards the temperature was increased to 75 °C and kept there 15 minutes more, then it was increased to 100 °C, kept there 30 minutes and, finally, it was increased to 125 °C and kept there 30 more minutes. The heating rate was set at 2.5 °C min^{-1} while a pressure of 33 MPa was applied during the overall process.

2.5. Composite mechanical characterization

The Izod impact resistance was performed according to ASTM D256-06 using a CEAST Resil 25 equipment. The tests were carried out at room temperature using notched specimens (incident energy of 2.75 J). The flexural properties were determined according to the three-point bending method described in ASTM D790-02. The equipment used was an Instron Universal testing Machine, model 5569, equipped with a 50 kN load cell. The rate of crosshead motion was equal to 1.36 mm min^{-1}.

2.6. Composite water absorption

The test was carried out as described elsewhere [32], in triplicate following a procedure adapted from ASTM D570-98. The samples were weighed at different time intervals and the percentage increase in weight corresponds to the water absorption (*A%*), as shown by the following equation, whereas m_t is the measured weight at different times and m_o the initial weight:

$$A\% = \frac{m_t - m_0}{m_0} \times 100$$

3. Results and discussion

3.1. Fiber and polymer characterization

The characterization of the fibers, reported and discussed in a previous paper [32], revealed that cellulose is the main component of the curaua fibers (62.9 wt%), followed by hemicellulose (29.4 wt%) and lignin (5.5 wt%). The initial crystallinity I_c, equal to 57 %, is reasonably high thanks to the high content of cellulose, which is a semicrystalline polymer, whereas hemicellulose, a branched amorphous structure, and lignin, a reticulated polymer, do not contribute to the I_c value. As to the thermal characterization of fibers, the thermogravimetric analysis indicates that the fibers are thermally stable up to 250 °C: indeed, at temperatures lower than 250 °C the weight loss is negligible and essentially related to water desorption. At T > 250 °C hemicellulose shows its decomposition, followed by cellulose and lignin, maximum decomposition rates of which are located at 360 °C and 400 °C, respectively. PBS undergoes one main decomposition stage, starting at approximately 280-300 °C. Moreover, from DSC analysis, PBS is a semicrystalline polymer, characterized by the glass transition temperature centered at -27 °C, the melting temperature at about 115 °C and crystallization from the melt at a temperature of 68°C.

The morphology study on cross section and surface of the fibers, as indicated in the previous work [32], showed a compact structure, composed of several connected ultimate microfibers which strengthen the fibers themselves. Moreover, it must be noted the presence of a rough surface with defects that can foster the formation of mechanical bonding resulting in strong adhesion between the phases.

3.2. Composite characterization

3.2.1. Effect of fiber loading

The results of the mechanical characterization of the composites prepared with different fiber loadings (10, 20 and 30 wt%) while their length is kept constant (3 cm), are reported in Fig. 2-4. The Izod impact resistance of composites is higher than that of PBS homopolymers (approximately 52 Jm^{-1}) and notably increases with the percentage of fibers, with the highest value for 30 wt% curaua fibers (345 Jm^{-1}). It is worth noting that this result clearly indicates the potential of lignocellulosic fibers as a reinforcing agent for PBS, since the impact resistance of thermoplastics, especially polyolefins, may decrease when they are reinforced by lignocellulosic fibers due, among other factors, to poor wettability during processing and not enough adhesion at interface in the end product, [34]. A larger surface area is probably available for the stress transfer leading to superior mechanical strength [40]. It can also be observed that the composites loaded with 30 wt%, which presents the highest strength, are characterized by a greater standard deviation, probably due to the formation of a more heterogeneous material.

The flexural strength has the same trend of impact resistance with the increase of fiber content, but in this case the improvement up to 30 wt% of loading was not as significant as in the impact resistance. In the case of flexural modulus, the increment of approximately 64 % as compared to the neat polymer is significant. In the flexural strength test, due to its inherent characteristics, the result is strongly influenced by the outer surface of the specimen [34], which is mostly composed of PBS in these composites. In addition, all the specimens did not break at 5% strain, thus suggesting that the ductility of PBS was retained.

SEM images of the samples are reported in Fig.5. It is worth noting that the fiber-matrix interfaces show good adhesion, despite the high fiber content reached in the composites. This could be due to the end chain hydroxyl groups and carbonyl groups of PBS which can interact with the polar groups of lignocellulosic fibers. Moreover, the strong interactions are favored by the curaua rough surface. These characteristics fully justify the observed enhancement of mechanical properties. However, in some regions of the sample prepared with 30 wt% of curaua (Fig.3c), it is clear that the PBS matrix, though to a small extent, was not enough to flow and wet all the fibers. This indicates that a further fiber loading would lead to an incomplete wetting of the fiber by PBS and, therefore, to a decrease in mechanical properties.

As to the water absorption tests (Fig. 6), it can be stated that, because the reinforcing agents are hydrophilic, they enhance affinity of the material for moisture and the water uptake increases with the fiber content [5,13]. Indeed the total water absorbed rises from 0.19 wt% for the pristine polymer to 1.2 wt% for the composite loaded with 10 wt% of curaua fibers, 1.7% when the fiber content is 20 wt% and 3.1% if the fiber load is 30 wt%. However, it should be emphasized that this test was carried out under extreme conditions, i.e., the specimens were immersed in water without any side protection. Such circumstances are not representative of a real application of these materials, i.e. the increase in the hydrophilic character of the composite, compared to the neat polymer, does not represent a significant restriction to the application of these materials.

3.2.2. Influence of fiber length

The investigation on the impact of the fiber length on the properties of the materials prepared in this work was carried out considering the composite reinforced by 20 wt% fiber. This composite did not feature the best mechanical properties, since 3-cm fiber lengths were used, but it was chosen primarily to check whether the impact resistance, an important property of composites, would be improved when the fiber length is increased (4 cm) or decreased (1 to 2 cm).

In Fig. 7, the Izod impact resistance is reported for the samples containing 20 wt% of curaua fibers, characterized by different lengths (1-4 cm). In all the composites, the mechanical performance increases with respect to PBS. Shorter fibers (1-2 cm) results in composites with the best mechanical properties. This behaviour is probably due to a better distribution of the fibers in the matrix. Indeed, agglomeration mostly occurs if longer fibers are used, causing non uniform dispersion with local clumping of fibers and matrix-rich areas, that can lead to a poor stress transfer efficiency favoring crack initiation and affecting the overall mechanical performance [6].

Regarding flexural tests, the results reported in Fig. 8 and 9 show that, when the fiber content is 20 wt% and the fiber length varies from 1 cm to 4 cm, the flexural strength gradually increases. However, the energy variation with fibers length, taking also into account the standard deviation, seems negligible. Moreover, no specimen broke under the current load conditions. Again, these results may stem from the fact that the outer surface of the material, mainly consisting of PBS in these composites, exerts a strong influence in the flexural test. As to the flexural modulus values, they are indicating a reinforcing effect of the fiber that is not affected by their length.

Fig. 10 reports SEM images obtained for composites with different fiber lengths (4, 2 and 1 cm). In all cases good interaction between the matrix and the fibers can be observed but, in the case of the composite with 4 cm fibers, as evidenced in the microstructure of Fig. 11a obtained after the impact, matrix-rich areas are clearly visible along with local agglomeration of fibers. In composites loaded with shorter fibers (Fig. 11b), some matrix-rich areas are present but they are sensibly smaller. The microstructure appears more homogeneous with superior distribution of the reinforcement in the bulk of the material. Thus, the efficiency of the stress-transfer from the matrix to the fibers increases, and probably leads to the superior Izod impact strength of composites manufactured with fibers of 1 and 2 cm. Moreover, the good distribution of fibers makes a greater surface area available for mechanical interlocking.

In Fig. 12, the results on water uptake are reported. It can be observed that shorter fibers present higher water absorption. Although the increase is small, it can be stated that shorter fibers generate a greater numbers of tips which can be considered as defects where water molecules can accumulate. Indeed, water absorption in composite materials is also affected by fiber distribution into the matrix: all the parameters influencing fiber distribution (fiber length, shape and content) also influence water uptake by the composite [41]. Despite their lower mechanical properties, longer fibers hinder water absorption showing better resistance.

4. Conclusions

Biocomposites were prepared via thermo-pressed molding. The polymeric matrix was PBS, which is one of the emerging bio-polymers, obtainable from monomers derivable from sugar fermentation and biodegradable. Curaua fibers have an elevated cellulose content which confers high crystallinity and good mechanical properties; they are thermally stable up to 250 °C. The melting temperature of PBS (T_m = 115 °C) allows the use of a maximum processing temperature during molding sensibly lower than the beginning of thermal degradation of natural plant fibers.

The results of Izod impact and flexural tests carried out on the composites prove that the mechanical performances increased with the fiber content and show an optimum result for a loading of 30 wt% of curaua fibers (3 cm). On the other hand, when a constant fraction of fibers was used (20 wt%), a size reduction resulted in a more homogeneous microstructure, as revealed by SEM images. The efficient stress transfer was increased leading to superior mechanical characteristics. In the considered length range, the optimum fiber dimension was located between 1 cm and 2 cm. Water uptake showed the great sensitivity of the material to the lignocellulosic content rather than fiber dimension. The greater water absorption capability due to lignocellulosic fibers can be beneficial or disadvantageous depending on the application of the final material: indeed, water is known to enhance the biodegradability of PBS. However, as already emphasized, water absorption tests were performed under far more aggressive conditions than those related to the potential applications of these materials.

The biocomposites with better mechanical properties than those of the neat polymer prepared in this work could have varied applications, for example in rigid packaging and interior parts of cars.

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Figure Captions

Fig.1. Chemical structure of PBS.

Fig.2. Izod impact resistance of PBS reinforced with different curaua contents (10-30 wt%, 3 cm).

Fig.3. Flexural strength of PBS reinforced with different curaua contents (10-30 wt%, 3 cm) measured as the force to deform the specimen up to 5%.

Fig.4. Flexural modulus of PBS reinforced with different curaua contents (10-30 wt%, 3 cm).

Fig.5. SEM images of the interphase between the polymer and the fibers (3 cm) of the fractured composites. (a) 10 wt%, (b) 20 wt%, (c) 30 wt%.

Fig.6. Composites water absorption.

Fig.7. Izod impact strength of PBS reinforced with curaua of different fiber lengths (1-4 cm, (20 wt%).

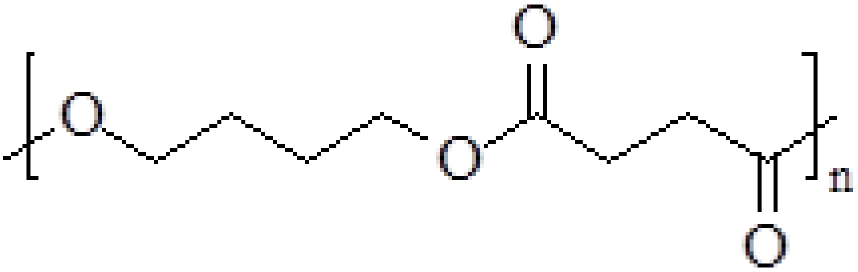
Fig.8. Flexural strength of PBS and composites (20 wt%, 1-4 cm) measured as the force to deform the specimen up to 5%.

Fig.9. Flexural modulus of PBS and composites (20 wt%, 1-4 cm).

Fig.10. SEM images of the interphase between the polymer and the fibers (20 wt%) of the fractured composites. (a) 4 cm, (b) 2 cm, (c) 1 cm.

Fig.11. SEM images of the fractured surfaces of the composites (20 wt%) (a) 4 cm, (b) 2 cm.

Fig.12. Composites water absorption.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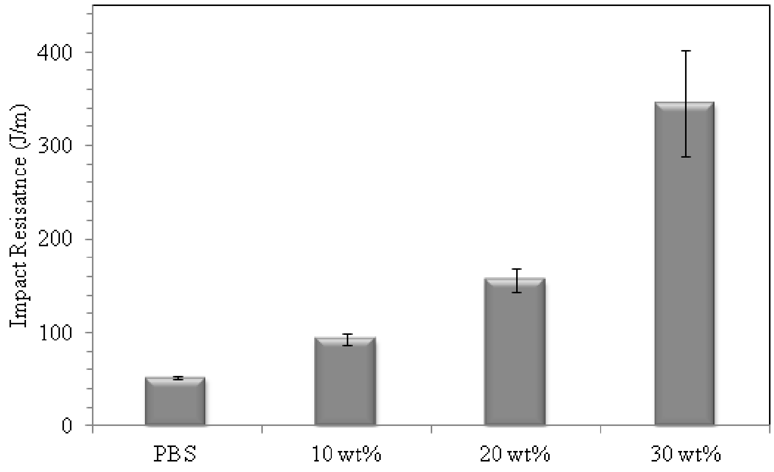
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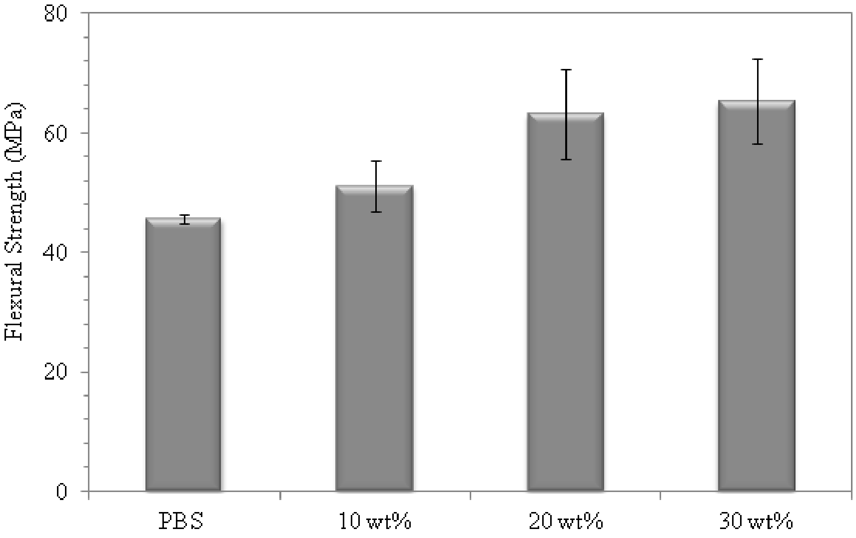
20 wt%
30 wt%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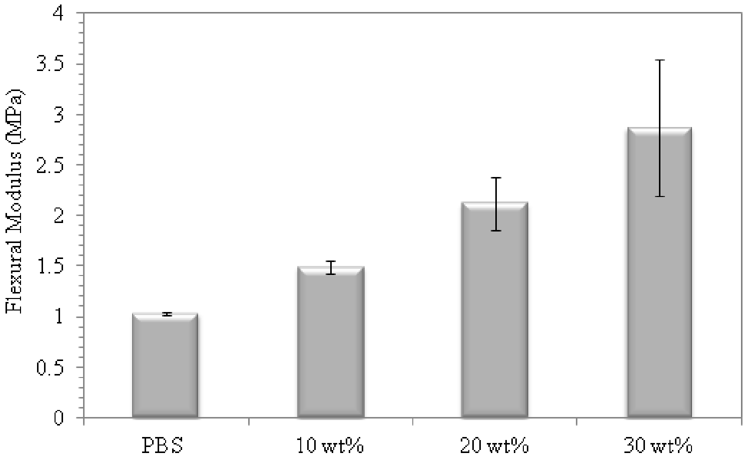
30 wt%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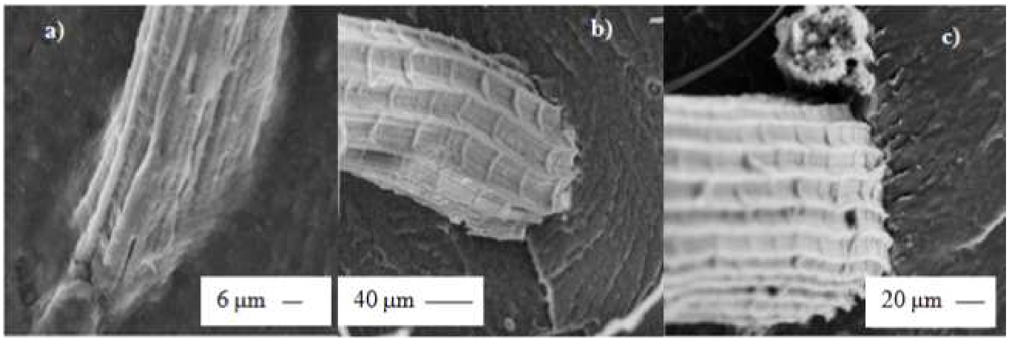
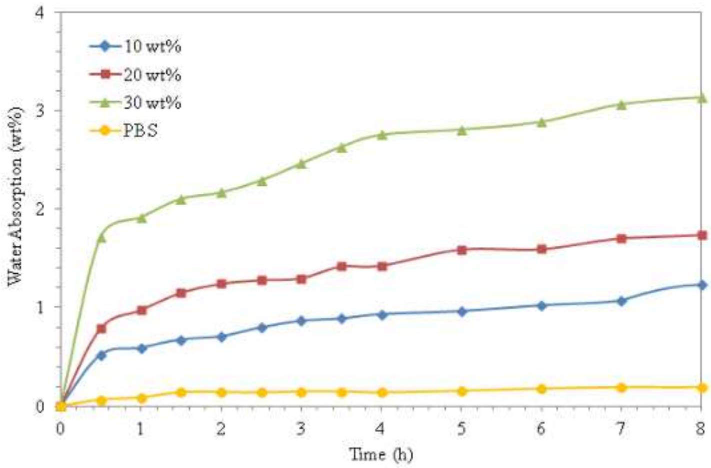
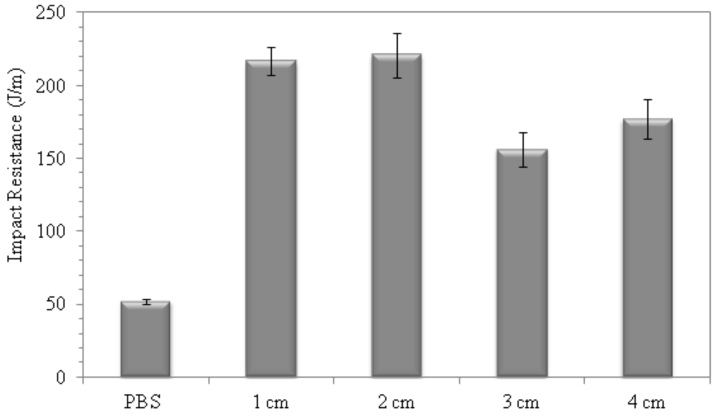
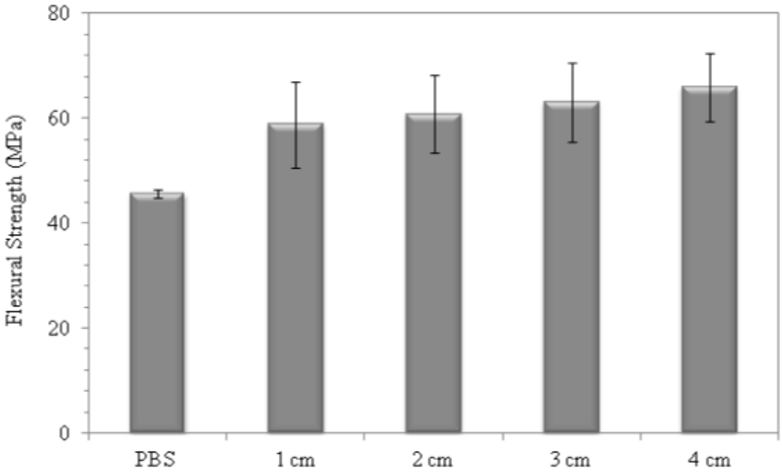
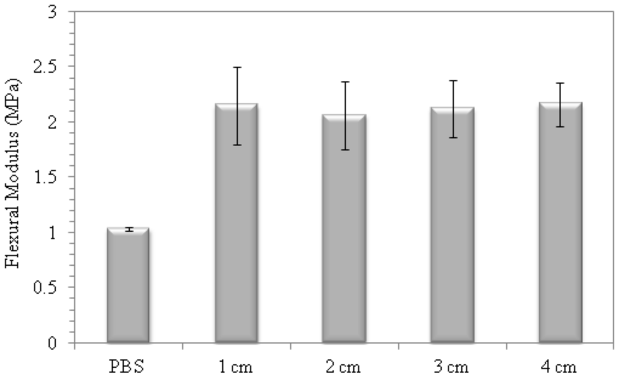
3
2.5
2
1.5
1
0.5
0
PBS
10 wt%
20 wt%
30 wt%

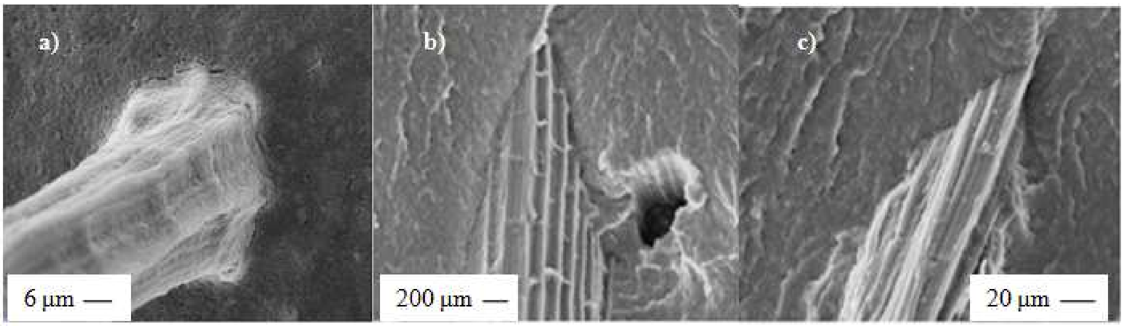

a)
6 µm
40 µm
b)
c)
20 µm

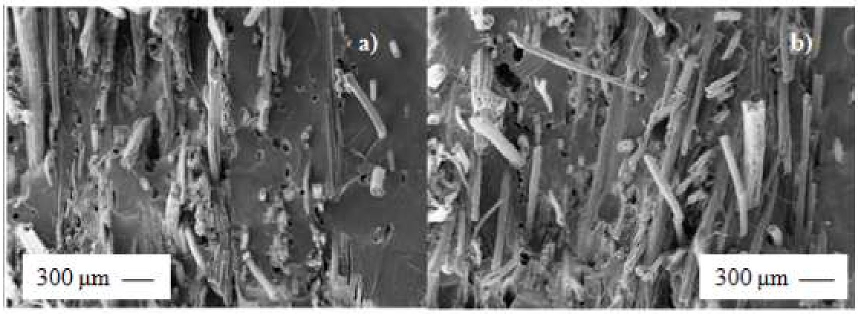

4
3
2
1
0
Water Absorption (wt%)
0
1
2
3
4
5
6
7
8
Time (h)
10 wt%
20 wt%
30 wt%
PBS

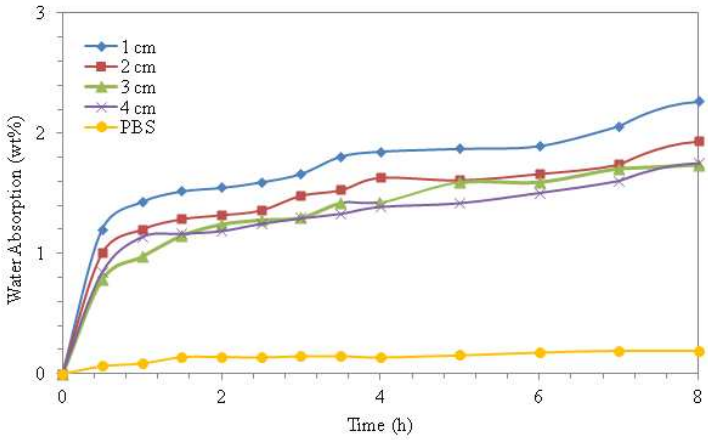

Impact Resistance (J/m)
250
200
150
100
50
0
PBS
1 cm
2 cm
3 cm
4 cm


Flexural Strength (MPa)
0
20
40
60
80
PBS
1 cm
2 cm
3 cm
4 cm


Flexural Modulus (MPa)
3
2.5
2
1.5
1
0.5
0
PBS
1 cm
2 cm
3 cm
4 cm


a)
6 µm
b)
200 µm
c)
20 µm


a)
300 µm
b)
300 µm


3
2
1
0
Water Absorption (wt%)
0
2
4
6
8
Time (h)
1 cm
2 cm
3 cm
4 cm
PBS