

Mechanical and Biodegradable Properties of Jute/Flax Reinforced PLA Composites

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Abstract: Green composites possessing biodegradable or recyclable characteristics have gained interest in recent years due to their ecofriendly, sustainable and lightweight characteristics over the conventional plastic-based materials. In this study, flax and jute natural fibers have been used individually and as hybrid reinforcement into Poly Lactic Acid (PLA) matrix. The composites developed are suitable to be used in biodegradable products in packaging and automobile industries. Hot press compression molding was used to fabricate samples of PLA/flax, PLA/jute and PLA/flax/jute (hybrid composites). The concentration of natural fibers in individual fiber-based composites was varied (between 0-50 %) by weight to investigate its effect on tensile and impact properties. Maximum tensile properties were obtained for 40 wt% single-fiber reinforced into PLA. This reinforcement content (40 wt%) was used as reference for hybrid composites. Hybrid composites were fabricated with different combinations of jute and flax fibers by keeping the total concentration of reinforcement equal to 40 % by weight. Tensile and Charpy impact tests were performed to evaluate the mechanical properties of the composites. Scanning Electron Microscopy of the tensile fractured surfaces was performed to observe the failure mechanism and adhesion at fiber-matrix interfaces in the composites. Further characterizations included Fourier Transform Infra-Red spectroscopy and Biodegradability tests, which were performed according to ASTM standards. Fourier Transform Infrared analysis revealed interaction between the natural fibers (jute and flax) and PLA matrix in hybrid composites. The enhanced interaction in hybrid composites resulted in their improved impact resistance. Based on the results obtained in this study, the improved mechanical and biodegradable properties of these composites make it suitable for use in applications like food-packaging and indoor plastic products in automobiles, to reduce synthetic plastic pollution.

Keywords: Poly lactic acid, Natural fibers, Hybrid composites, Mechanical properties, Biodegradability

Introduction

The concerns regarding petrochemicals have raised due to their anti-environmental characteristics and continuous depletion [1,2]. The researchers now are seeking interest in alternatives that are environmentally sustainable, biodegradable, ecofriendly, non-abrasive, non-toxic and independent of petrochemicals [1-5]. Green composites consisting of natural fibers (NFs) and biodegradable polymers are the potential replacement of the conventional petrochemical based materials [6,7]. Biodegradable polymers can be naturally available, processed from renewable sources, microbial synthesis or petrochemical synthesis [6]. PLA is used in this research which is the most extensively researched biodegradable polymer, derived from renewable sources [2,6]. PLA is manufactured with wide range of applications; including packaging industry, as matrices for drug delivery, it performs key roles in medical applications like bone fracture internal fixation, dissolvable sutures, in textile industry and in agricultural sector as agricultural plant growth promoter [8,9]. The brittle nature of PLA reduces the variation in applications of PLA but addition of reinforcements is one of the ways to improve its properties [5,10]. NFs reinforced PLA have found application in packaging industry even for mass

transportation and also in manufacturing of non-structural parts of automotive, marine and aerospace industry [5,11]. Moreover, several researchers have also been interested in substituting petroleum-based products with PLA and its composites [12]. The reinforcements in a green composite is in the form of NFs which are derived from plants, animals or minerals [6]. NFs extracted from plants like jute and flax are part of this research. Jute is the most easily available NF with good mechanical and thermal properties [5]. Flax fiber provides advantages of low density, high strength, high toughness and high stiffness [5]. In past, more research is available on NFs and non-biodegradable polymer composites, but currently interest has been shifted toward totally biodegradable green composites which involves NFs as reinforcement and biopolymers as matrix [13-17]. In addition to good mechanical and thermal properties, these materials are environmental friendly and non-toxic in nature [1,18].

The strength of a green composite depends on the reinforced natural fiber. The strength and stiffness of a fiber is provided by the amount of arrangement of cellulosic microfibrils present in each fiber. Natural fiber reinforcements in polymer may enhance certain desired properties of composites including stiffness, strength and biodegradability [12]. Ku *et al.* [19] found that increasing the fiber content increases the tensile strength of NF reinforced composites up to an optimum value, beyond this point further increase in fiber content reduces the tensile strength of the composite.

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Tao *et al.* [20] studied mechanical and thermal properties of PLA/jute and PLA/ramie composites. They concluded that 30 % NFs in PLA gives maximum mechanical properties; TGA test showed that degradation temperature of PLA was improved by addition of NFs. Compatibility between PLA and kenaf fiber was investigated by Kamarudin *et al.* [21]. They showed improvement in fiber matrix interface till 40 % of kenaf fiber was added in PLA, further addition of reinforcement caused poor filler matrix compatibility, resulting earlier fracture of the composite. The mechanical performance of a composite highly depends on the nature of fiber, orientation of fiber and also the fiber/matrix interaction [12]. Mechanical properties of hybrid green composites with and without treatment of NFs was investigated by Ranjan *et al.* [3]. They concluded that mechanical properties of PLA were increased by adding NFs in it, but surface treated NFs further enhanced the mechanical properties. Biodegradability of PLA has been a topic of research in various studies [22-25]. Siakeng *et al.* [12] studied that temperature, time, impurities, residual catalyst concentration and molecular weight of PLA are the factors which influence the biodegradability of PLA.

In this investigation; firstly, green composites of PLA/flax and PLA/jute were fabricated separately by hot press compression molding process. Different percentages of natural fibers by weight (between 0-50 %) were added in PLA and characterized. Maximum mechanical properties were obtained at 40 wt. % fiber in PLA for both PLA/flax and PLA/jute composites. In the next step, hybrid composites of flax and jute reinforced into PLA were made in different combinations of flax and jute fibers by keeping the total weight of reinforcement equal to 40 % of the composite. The properties of hybrid composites were investigated. Tensile test, impact test, Scanning Electron Microscopy (SEM), Fourier Transform Infrared Spectroscopy (FTIR) and biodegradability tests were performed. This study indicates improvement of mechanical properties of the composites after reinforcement of fibers compare to pure PLA. Flax fiber-based composites have been found to have higher tensile strength as compared to jute fiber-based composites and hybrid composites. Hybrid composites have displayed higher impact energy absorption capacity as compared to individual flax and jute fibers-based composites. FTIR study indicated enhanced interaction between the natural fibers and PLA matrix in the hybrid composites. SEM study of tensile fractured surfaces indicated better interfacial adhesion between PLA and flax fibers as compare to PLA and jute fibers. Moreover, biodegradability of the composites was proven by Soil Burial test. Results are explained in details in the results and discussion section of this paper.

Experimental

Materials

The material used in this research includes PLA (as

Table 1. Properties of PLA, jute and flax [3,26]

| Properties | PLA | Jute | Flax |
|-----------------------------|-----------|---------|----------|
| Density (g/m ³) | 1.24 | 1.45 | 1.5 |
| Tensile strength (MPa) | 18.77 | 400-800 | 800-1500 |
| Youngs modulus (GPa) | 3.38-3.62 | 10-30 | 60-80 |

matrix) and jute and flax fabrics (as reinforcement). PLA is extensively researched biopolymer and has relatively good availability. PLA shows outstanding physical and mechanical properties; it has been the frontrunner among many biopolymers [12]. For this research, PLA was supplied by Huaian Ruanke Trade Co, Ltd., China, (Melting Temperature 155-180 °C). Bi-directional woven flax fabric was purchased from Easy Composites (UK), while jute fabric was purchased from Future Composites (Pakistan), in woven bi-directional form. The density of the fibers and PLA was obtained from the corresponding suppliers. Tensile properties of PLA were evaluated in laboratory during this research while reference values of the tensile properties of the reinforcement have been cited from the published literature [3,26]. Table 1 illustrates various properties of constituents used in this research.

Composites Fabrication

PLA/jute, PLA/flax and PLA/jute/flax (hybrid composites) were fabricated using hot press compression molding technique. Flax and jute fabrics were cut to 250 mm×250 mm square pieces. The concentration of fibers in PLA was varied between 0-50 % by weight. Table 2 explains different weight fractions of jute and flax fibers in PLA matrix with nomenclature. For composites fabrication, the preheated material was placed in an open heated mold cavity. A plug member was used to close the mold and pressure was

Table 2. Weight percentage of reinforcement in matrix

| Sr No | PLA (%) | Flax (%) | Jute (%) | Nomenclature |
|-------|---------|----------|----------|--------------|
| 1 | 100 | 0 | 0 | PLA |
| 2 | 90 | 10 | 0 | 10F |
| 3 | 80 | 20 | 0 | 20F |
| 4 | 70 | 30 | 0 | 30F |
| 5 | 60 | 40 | 0 | 40F |
| 6 | 50 | 50 | 0 | 50F |
| 7 | 90 | 0 | 10 | 10J |
| 8 | 80 | 0 | 20 | 20J |
| 9 | 70 | 0 | 30 | 30J |
| 10 | 60 | 0 | 40 | 40J |
| 11 | 50 | 0 | 50 | 50J |
| 12 | 60 | 20 | 20 | 20F20J |
| 13 | 60 | 30 | 10 | 30F10J |
| 14 | 60 | 10 | 30 | 10F30J |

applied so that the material comes in contact with all the mold areas. 16 MPa pressure and 185 °C temperature was kept maintained for 2-3 minutes [27], then the molding material was unloaded and allowed to cure. 250 mm×250 mm sheets of composites were prepared and specimens were cut according to ASTM standards for tensile, impact, and biodegradability tests.

Characterizations

Following characterizations were conducted to examine the properties of PLA/Flax, PLA/Jute and Hybrid composites.

Tensile Test

Specimen of Pure PLA and composites were tested for tensile properties according to ASTM D3039. Tensile tests were performed on universal testing machine WDW-100E with a load cell of 100 kN. The cross-head speed was set at 2 mm/min.

Impact Test

Charpy Impact test was performed to evaluate the energy absorbed by the specimen before fracture on JP-300B (Time Group Inc., China) with 14 kg hammer, with sample sizes according to ASTM D256.

Scanning Electron Microscopy (SEM)

Fiber-matrix interfaces and failure behavior of tensile fractured specimens were observed using SEM images. MIRA 3 TESCAN equipment was used to conduct this study. The images of samples were obtained by focusing fractured surfaces with the high energy beam of electrons.

FT-IR Spectroscopy

FT-IR spectroscopy was performed to study possible interactions between the constituents of the composites. The experiment was performed on FTIR equipment (Perkin Elmer Spectrum 100) in dry air and at ambient temperature with spectra range from 4000 to 400 cm^{-1} .

Soil Burial Test

Biodegradability of the composites was evaluated by soil burial test. The specimens were weighed and then buried underneath the soil for a time period and then taken out to check its loss in weight. Specimens (40 mm×8 mm×3 mm) of all composites were buried in a sand-soil (50-50 %) mixture. Temperature was kept at 30 ± 2 °C; to maintain moisture level constant 30-40 % of water was added to the sand-soil mixture every three days [28]. Weight of the samples was measured after each 10 days and then reburied. The process was continued for one month to calculate the rate of biodegradation.

Results and Discussion

Fourier Transform Infrared Spectroscopy (FTIR)

As the hybrid composite contains two different types of natural fibers which may have possible interaction among themselves and with the PLA matrix. FTIR was carried out to investigate the functional groups present in the matrix and

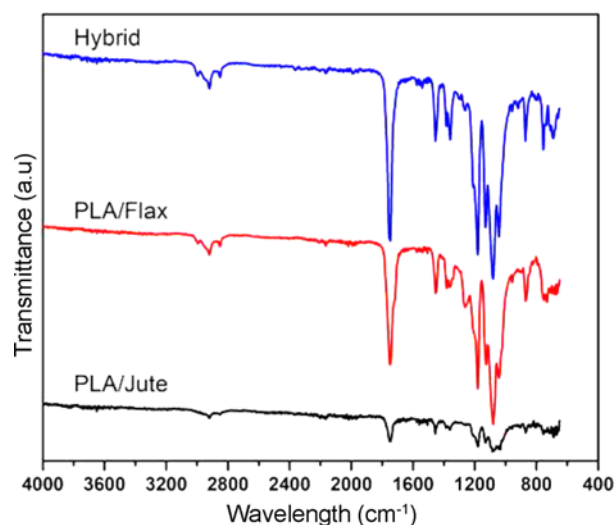


Figure 1. FTIR Spectra of PLA/jute, PLA/flax and hybrid composites.

reinforcements, and possible formation of functional group by hybridization of flax and jute fibers in PLA. In order to study the appearance and shifting of the transmittance peaks, FTIR spectrum of hybrid composite has been compared with the spectra of PLA/jute and PLA/flax composites. Figure 1 shows the FTIR spectra of FTIR spectra of PLA/Jute, PLA/Flax and Hybrid composite. Table 3 shows various bonds and the corresponding wavelength of PLA/Flax/Jute hybrid composite.

Appearance of new peaks and shift of the existing peaks in the FTIR spectrum of hybrid composite is compared to the spectra of individual PLA/jute and PLA/flax composites. An additional peak at 1755 cm^{-1} for the hybrid composites can be observed (Figure 1) which belongs to C=O stretching [29]. Two peak shifts at 1078 cm^{-1} and 869 cm^{-1} were also observed, indicating strong C-O stretching and strong C-H bending respectively [30].

Formation of these functional groups in the hybrid

Table 3. Several bonds and the corresponding wavelength of PLA/Flax/Jute hybrid composite [21,22]

| Sr. No. | Bond | Wavelength (cm^{-1}) |
|---------|--|---------------------------------|
| 1. | C-H stretching in cellulose and hemicellulose | 2870-2920 |
| 2. | C=O stretching due to acetyl group present in hemicellulose | 1747 |
| 3. | Bending deformation in CH_2 and CH_3 compounds | 1456 |
| 4. | OH-group | 1382 |
| 5. | O-C stretching in carboxylic acid and its derivatives | 1263 |
| 6. | O-C=O stretching of ester bonds | 1087-1180 |
| 7. | Out of peak O-H bending of alcohol and phenol | 680-760 |

composite denote better interaction between the two kinds of natural fibers (jute & flax) and the PLA matrix providing better mechanical properties. No additional interaction was observed in the hybrid composite instead the existing interaction is further enhanced by combination of different natural fibers, which is in correspondence with the literature [31].

Tensile Properties

The tensile strength of pure PLA was compared with PLA/flax, PLA/jute and PLA/flax/jute hybrid composites as shown in Figure 2. An obvious increase in the tensile strength of the composites was observed after reinforcement of Jute, Flax and hybrid Jute/Flax in PLA matrix. Increase in tensile strength of PLA/Flax and PLA/Jute composite was recorded up to 40 wt.% fiber reinforcement in PLA. The strength of composites was however observed to decrease after further reinforcement of both flax and jute fibers beyond 40 %. When fiber content exceeds a certain amount, formation of fiber-fiber interaction may occur, and the applied load cannot be transferred from matrix to the fiber which results in drop of tensile strength [32]. Keeping this finding in view the combined weight percentage of jute and flax fibers was kept as 40 % by varying the individual amount of each fiber to find optimum combination of both jute and flax in the hybrid composites. The composition of

the hybrid composite along with the nomenclature has already been explained in Table 2.

PLA/flax composites were found to display the maximum tensile strength, while the hybrid composites have higher tensile strength than PLA/jute composites. One possible reason is that flax fiber itself has higher tensile strength than that of jute fiber due to its higher cellulosic content [33]. In this study tensile strength of pure PLA (18.77 MPa) has been observed to increase to 72 MPa (283.6 %), 45 MPa (139.7 %) and 61.89 MPa (229.7 %) after flax, jute and hybrid reinforcement in PLA respectively. The hybrid composite has higher tensile strength than PLA/jute composite because of the presence of flax as well as the enhanced interaction between the hybrid reinforcement and matrix which has been discussed in FTIR spectroscopic results. For details, average tensile properties of individual and hybrid fiber composites with maximum tensile strength, modulus and elongation at breakup are shown in Table 4.

Scanning Electron Microscopy (SEM)

The SEM fractographs of tensile fractured surfaces of PLA/flax, PLA/jute and hybrid composites are shown in Figure 3. Well dispersed fibers in PLA matrix can be observed in the SEM micrographs of all the composites. To emphasize on the failure behavior, fiber breakage has been observed in the SEM images of PLA/flax composite as a result of tensile fracture (Figure 3(a)), while PLA/jute composite suffered from fiber breakage as well as fiber pull-out at some locations (Figure 3(b)). No fiber pull-out has been observed in the hybrid composite which contains both flax and jute fibers (Figure 3(c)). The fiber breakage phenomenon indicates strong interfacial bonding between the reinforcement and matrix in the PLA/flax composites; while, the presence of fibers pull-out shows some poor interfaces in PLA/jute composite. Interfacial bonding is one strong reason why PLA/flax composites have higher tensile strength than that of PLA/jute composites. Absence of fibers pull-out in the hybrid composite is a sign of enhanced interfaces as a result of interaction between flax, jute and the PLA matrix which is also reported in the FTIR results.

Impact Properties

The impact energy of pure PLA was compared with PLA/flax, PLA/jute and PLA/flax/jute hybrid composites as shown in Figure 4. Impact energy absorption capability of the composites was observed to increase after addition of jute, flax and hybrid reinforcements (jute/flax) in the PLA matrix. 40 wt% reinforcement in PLA matrix resulted in maximum impact energy absorption for both PLA/flax and PLA/jute composites. The fiber increases the amount of energy required to cause fracture. In fiber reinforced polymer composites, toughness is dependent upon fiber, polymer matrix and their interfacial bond strength. Hybrid composite (30F10J) was found to absorb higher impact energy than

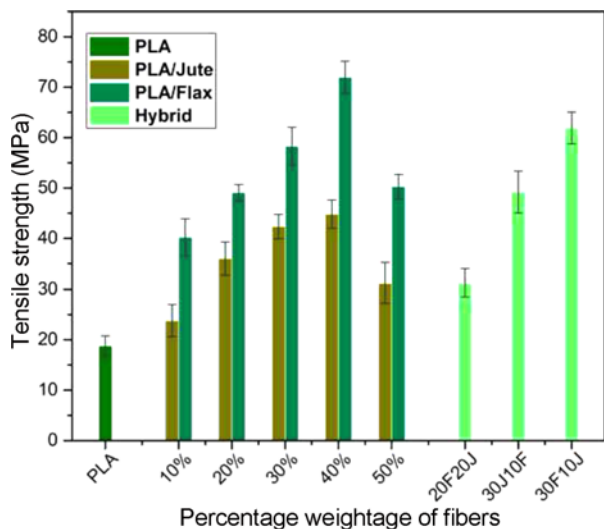


Figure 2. Tensile strength of pure PLA and PLA/Fibers composites.

Table 4. Average tensile properties of selected composites

| S. No. | Composites | Tensile strength (MPa) | | Tensile modulus (GPa) | % Elongation at breakup |
|--------|------------|------------------------|--------------------|-----------------------|-------------------------|
| | | Average value | Standard deviation | | |
| 1 | 40F | 72 | 3.19 | 23.1 | 2.6 |
| 2 | 40J | 45 | 2.79 | 17.83 | 1.28 |
| 3 | 30F10J | 61.89 | 3.14 | 17.75 | 1.90 |

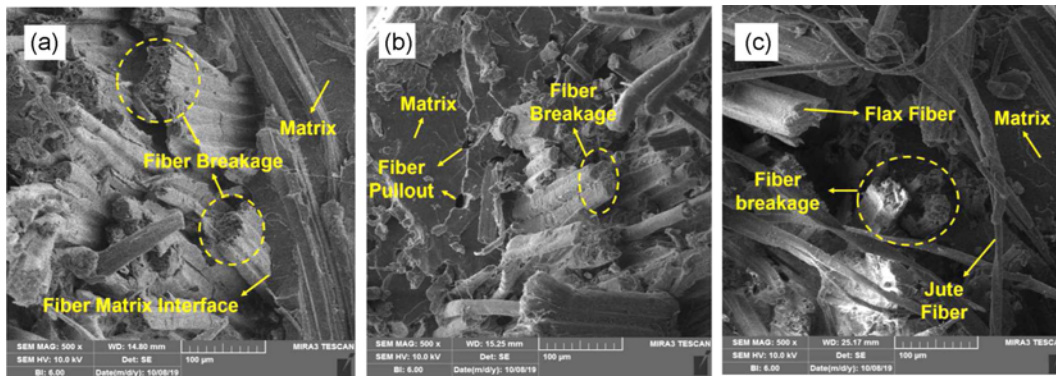


Figure 3. SEM images of tensile fractured surfaced of (a) PLA/flax(40F), (b) PLA/jute(40J), and (c) hybridcomposite (30F10J).

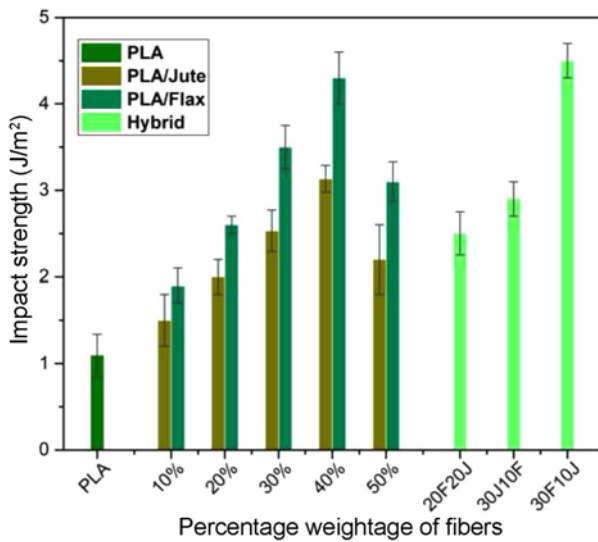


Figure 4. Charpy Impact Energy of Pure PLA and PLA/Fiber composite.

those of PLA/flax and PLA/jute composites. The impact energy absorbed by pure PLA 1.1 J/mm² has been observed to increase to a maximum of 4.3 J/mm² (290.9%), 3.13 J/mm² (184.5%) and 4.5 J/mm² (309.1%) after reinforcement of flax, jute and hybrid (flax/jute) in PLA, respectively.

Encouraging results were obtained from impact test showing that the impact energy absorption capacity of hybrid composites is greater than PLA/flax and PLA/jute composites. This is supported by the fact that flax fiber

increases the amount of energy required to pull-out fibers. Similar results were also observed by Siakeng *et al.* [34], where hybridization of coir and pineapple leaf fibers increased impact strength as compared to single fiber reinforced composites. Impact strength of a material is controlled by toughness; and for composite materials it depends on the toughness of individual constituents and their interfacial shear strength. The results indicate that hybridization improves the interfacial adhesion and resists the crack propagation due to impact loading. The coefficient of friction between two different fibers is greater compared to similar fibers, so two layers of different fibers (hybrid) need more energy to delaminate under impact loading as compared to layers of same fibers. The hybrid fiber layers consume the impact energy more effectively over the interfaces as compared to those of individual fibers.

Biodegradability

Biodegradability of the composites was investigated by soil burial test. The test was performed on pure PLA and PLA/flax and PLA/jute composites having maximum mechanical properties as identified above (i.e. 40F and 40J composites). The purpose of this study was to identify the biodegradability rate of the composites. Specimens of pure PLA, 40F and 40J were buried in soil/sand mixture for 10, 20 and 30 days. Weight of the specimens was measured before and after the test. Results of the soil burial test are published in Table 5.

Before burial the weight of the samples was measured as

Table 5. Mass loss after soil burial (all values are in grams)

| | PLA (g) | | 40J (g) | | 40F (g) | |
|---------------|---------------|--------------------|---------------|--------------------|---------------|--------------------|
| | Average value | Standard deviation | Average value | Standard deviation | Average value | Standard deviation |
| Initial mass | 1.000 | - | 1.500 | - | 1.350 | - |
| After 10 days | 0.971 | 0.0074 | 1.490 | 0.0023 | 1.346 | 0.0012 |
| After 20 days | 0.947 | 0.0058 | 1.479 | 0.0045 | 1.339 | 0.0032 |
| After 30 days | 0.925 | 0.0082 | 1.462 | 0.0048 | 1.331 | 0.0026 |

1 g of PLA, 1.5 g of 40J and 1.35 g of 40F. The weight loss of PLA, 40J and 40F has been observed to decrease to 0.925 g (7.5 %), 1.462 g (2.6 %) and 1.331 g (1.43 %) after 30 days. Further loss in weight may be recorded after burial of the specimens for longer periods. Biodegradability depends on the nature of constituents, their interfacial bonding and the environmental conditions. Biodegradation also relies upon the effective motion of these microorganisms over substrate. Diffusion of moisture from soil notably helps the growth and effective motion of microorganisms [35]. PLA and natural fibers are hydrophilic in nature [36]. The fiber matrix interface is the weak zone of composite providing excessive surface area for microbial attack. Natural fiber being hydrophilic act as a path way for microorganisms to biodegrade composite, in addition to the biological contact occurring at composite-environment interface [37]. The hydrophilic group in PLA and natural fibers promote the absorption of moisture in their territory and help microorganisms to act and hence reducing the weight of the samples. Thus, the natural fibers reinforced PLA composites are biodegradable.

Conclusion

Biodegradable hybrid green composites of flax and jute fiber reinforcements in PLA matrix were developed using hot press compression molding technique. An increase in the mechanical properties of the composites was observed after reinforcement of jute, flax and hybrid jute/flax fibers in PLA matrix. Maximum mechanical properties were recorded after 40 wt% reinforcement of natural fibers in the PLA matrix. Tensile strength of PLA/flax composites was found to be higher than PLA/jute and hybrid composites. SEM study established that the tensile properties of flax fibers-based composites are higher because of better fiber-matrix interfaces as compared to those of jute fibers based composites. Hybrid composites also displayed better interfacial properties. The same was verified in the FTIR spectroscopy where interaction between matrix and reinforcements has been established through new peaks generation and peaks shifting. The enhanced impact strength of hybrid green composites makes them suitable in high impact loading applications as compared to the individual fiber based composites. The hybrid composites have capability to absorb more impact energy due to higher coefficient of friction between dissimilar fibers and improved interfaces. The weight loss of composites as a result of soil burial shows that composites are biodegradable. The degradability rate of PLA/flax and PLA/jute composites is different because of the individual composition and water absorption characteristics of the flax and jute fibers. Overall, the hybrid composites outperformed the individual fiber reinforced composites in terms of impact properties. Moreover, the strength was comparable to conventional plastic-based materials and

recently researched natural fiber reinforced polymers. The composites demonstrated in this research have excellent mechanical properties with potential to replace the conventional polymers and composites in indoor applications, non-structural parts of automobiles, marine and aerospace industries.

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