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Plant-Derived Fiber-Reinforced Polymer Composites: The Impact of Compatibilizer on the Fundamental Characteristics and Degradation Properties

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Abstract

By using compression molding, unidirectional polypropylene (PP) composites reinforced with banana fiber were created both with and without the use of stearic acid (SA) as a coupling agent. Four filler loading levels (10%, 20%, 30%, and 40% by weight) were employed during the composite manufacturing process, using raw banana fiber. The resulting composites underwent mechanical testing (tensile, bending, and impact characteristics). The ideal combination of mechanical properties was found in 30% fiber-reinforced composites, depending on the fiber loading. To improve its compatibility with the polymer matrix, optimized banana fiber underwent a chemical treatment using SA. When compared to the raw composites, SA-treated banana fiber-reinforced composites produced superior mechanical properties. Scanning electron micrographs of the tensile fractured samples revealed better adhesion between banana and PP matrix upon treatment with SA, which provided additional information regarding the fiber-matrix adhesion. Investigations were also conducted into the composites' water uptake and weathering simulation tests. Unidirectional PP composites reinforced with banana fibers were developed using compression molding, with and without SA as a coupling agent. Four filler loadings (10%, 20%, 30%, and 40% by weight) were used, and mechanical tests revealed that composites with 30% fiber loading had optimal properties. Chemical treatment of banana fibers with SA improved fiber-polymer matrix compatibility, resulting in enhanced mechanical performance compared to untreated fibers. SEM analysis confirmed improved fiber-matrix adhesion with SA treatment. Additionally, the composites were evaluated for water absorption and weathering resistance, providing insights into their durability.

Keywords: Banana fiber, polypropylene, composite, stearic acid, mechanical properties

INTRODUCTION

The use of natural fiber to reinforce thermoplastics in composite materials has been the subject of extensive study and development worldwide. These materials are on the market right now, mostly to the automobile sector [1–3]. Natural fibers are a renewable resource for materials that are widely available, especially in tropical areas, have high mechanical properties, and are typically inexpensive. Natural fibers have several advantages over synthetic fibers, including lower cost, low density, renewability, and biodegradability. Recently, researchers have been interested in the use of natural fibers as reinforcement in polymeric matrices. When considering the environment, using natural fibers would be more healthful than using synthetic ones. Natural fibers are currently utilized in the construction, automotive, and packaging

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industries. Examples of these fibers include jute, cotton, banana, bamboo, coir, flax, hemp, and sisal. All of these fibers have been demonstrated to be efficient reinforcement in thermoplastic matrices [4, 5].

Banana fiber, also known as Manila hemp, is one of the most important natural fibers in the world and is derived from the bast fiber of the *Musa textilis* plant. It is a member of the Musacea plant genus, which also includes bananas. Bananas, renowned for their resistance to elements like stone impact, exposure to the elements, and moisture, are the first natural fiber that meets the strict requirements for materials used on the exterior of motor vehicles [6]. Tropical locations have an abundance of banana plants and other agricultural plants. An additional financial investment is not required for the industrial application of banana fiber, which is a byproduct of banana cultivation. Table 1 displays a few of the typical attributes of banana fiber [7]. Because of Daimler Chrysler's inventive application of banana fiber in underfloor protection for passenger cars, banana fiber reinforced composites are becoming more and more common these days [8]. There are several articles on banana fibers that detail the usage of banana fibers as a reinforcing agent in thermoplastics like polyethylene (PE) and polypropylene (PP) [9–11]. Thermoplastic engineering polymers are a class of materials with several important and useful properties, such as a high Vicat softening point, good flex life, sterilizability, good surface hardness, and good resistance to scratches and abrasions. For applications requiring normal temperatures, PP's excellent and desirable physical, thermal, and mechanical properties were taken into consideration. Its low specific density, lack of environmental pollution upon burning, and significant market share in wood plastic composites further contributed to PP's selection as the composite's matrix material [12–14].

Both natural and banana fiber display strong hydrophilicity due to the attraction or interaction between the hydroxyl groups of the fiber components and water molecules. Interactions between fiber and water begin in the non-crystalline region and progress toward the crystalline region. Water is absorbed by hygroscopic materials such as hemicellulose and crystalline and non-crystalline cellulose by a process known as hydration that uses available hydroxyl groups. When a cellulose molecule absorbs a water molecule, an exothermic event takes place that produces cellulose hydrate, which serves as a catalyst for more absorption [15, 16]. Due in part to its high moisture sensitivity, lignocellulosic fibers can cause dimensional instability in composite materials, which limits the use of fiber as reinforcement. Due to their hydrophilic nature, natural fibers often have low interfacial properties that limit their potential as reinforcing agents; to improve the fiber interface, chemical changes and chemical coupling agents are considered. The majority of substances with two unique functions function as chemical coupling agents. Reactions between the hydroxyl groups of the cellulose and the functional groups of the matrix are involved in the first and second tasks, respectively [17]. The fibers' ability to absorb moisture can be reduced by chemical treatments such as acetylation, mercerization, methylation, cyanoethylation, benzoylation, and permanganate treatment [18–20].

It has been accepted that stearic acid (SA) is a coupling agent in addition to the coupling agents mentioned above. As a coupling agent, though, its full potential has not yet been thoroughly explored. Kim and colleagues [21] developed PE composites using SA-coated zeolite as the filler [21]. The flexibility of the polymer matrices was enhanced by the SA coating on the zeolite surface. $CaCO_3$-LLDPE (LLDPE = linear low-density polyethylene) composites treated with SA were produced by Bellayer et al. [22] in order to gain a better understanding of the intumescence mechanism. By permitting $CaCO_3$ to diffuse widely across the LLDPE matrix, SA aided in the formation of a coherent network when the thermal stress during burning was high. After a thorough review of the literature, it is discovered that SA has not yet been investigated for use in cellulosic fiber composites as a coupling agent. Therefore, the goal of this work was to look into the use of SA in the creation of banana-PP composites as a coupling agent. In the current work, a detailed investigation has been conducted into the change in mechanical properties, including tensile, bending, and impact properties, of banana fiber-reinforced polypropylene composites formed by compression molding. In this effort, composite materials will be made from banana fiber that has been treated with SA and PP. Furthermore, the impact

of fiber loading on the mechanical properties and morphology of banana fiber reinforced PP is reported. The composites' ability to absorb water and imitate weathering was also investigated.

EXPERIMENTAL

Materials

PP granules weighing 0.90 g/cm^3 and containing 2.16 kg of melt at 230°C per minute were purchased from Cosmoplene Polyolefin Company Ltd. (401, Ayer Merbau Road, Singapore). To make banana fibers with fiber diameters between 0.20 and 0.25 mm, which were used as filler, banana trees grown in Bangladesh were utilized. With long fibers that firmly overlap to form a pseudostem, the banana plant is a tall herbaceous plant that grows to a height of 2 to 16 meters. A total of 30 large leaves, each spanning more than 2 meters in length and 30 to 60 cm in width, are often produced by banana plants. More chemicals, including toluene, acetone, and SA, were sold by Aldrich Chemical Co. Inc. (USA). Solvents were used just as sent.

Composites Manufacturing

The banana fibers were carefully separated by hand, and then the adhering pith, if any, was removed. The fibers were then uniformly cut to a length of 150 mm. The well-separated fibers were submerged in acetone for 30 minutes at room temperature in order to eliminate unnecessary material. When the cleaned samples had dried in the air, the desired amount was weighed. Toluene and acetone were used to dissolve SA, and the ratio of SA to acetone was 1: 10: 10 (w/w/w). An aluminum tray was lightly coated with banana fibers, which were added to the solution in a 1:3 (w/w) ratio. The solvent combination was allowed to evaporate at room temperature after the tray was covered with aluminum foil and let to stand for two hours. Additionally, SA-coated banana fibers were dried in an oven set at a constant 60°C for 48 hours. To make one polypropylene sheet, roughly 6 g of polypropylene granules were placed between two steel plates and pressed in a hot press (Carver, Inc, USA, Model 3856). The press was operated at 180ºC. Steel plates were compressed to 7 MPa for 2 minutes.

After that, the plates were cooled for 3 minutes at room temperature and under 7 MPa of pressure in a different press. The resulting PP sheet was cut to the necessary dimensions (150 × 120 × 0.25 mm^3) for composite fabrication. After that, the fibers were positioned at random between PP sheets in a mold measuring 150 ×120 × 0.25 mm^3. Composites with different fiber loadings (e.g., 10–40 wt%) were made by sandwiching four layers of fibers between five layers of pre-weighted PP sheets. To make composites, this sandwich was squeezed for five minutes at 190°C and 10 MPa. Then, using a separate press (Carver Laboratory Press, USA, Model 2518) operating in a cooling environment, the composite sample composed of steel plates was cooled.

Morphological Examination

The composites' tensile fracture surface morphology was investigated using a 10 kV scanning electron microscope (SEM, model JEOL JSM-5310) both with and without SA treatment. The samples were vacuum-coated with a thin layer (25 nm) of gold using a BAL-TEC SCD 050 sputter coater layer to avoid electrical charge accumulation prior to SEM.

The Water Absorption of the Composites

Rectangular specimens with dimensions of 25.4 mm × 76.2 mm were chosen for the water uptake test in compliance with ASTM D-579-99. The samples were dried by heating them to 50°C in an oven for about 24 hours. Afterward, they were cooled in a desiccator and weighed. The samples were then immersed in a beaker with 100 mL of deionized water at room temperature (25°C) for varied durations, up to 30 days. Samples were taken out of the beaker after a certain period of time, wiped five times with a dry cloth, and then their weights were recorded once again. The samples were returned to the water after each measurement. The samples' increased weight as a result of consuming water was measured as a weight difference and displayed as a percentage increase over baseline weight.

Examine the Simulated Weathering

For a total of 300 hours, the untreated and treated composite samples were exposed to sunshine for four hours (65°C ± 2°C) using the UV lamp (313 nm) and dews and condensation for 2 hours (45°C ± 2°C) using a tester (Q-UV-26200, Q-panel Co.). After that, the samples were weighed, dried for 30 minutes in the oven, and their mechanical properties evaluated. In each of the previously described tests, at least five distinct samples are assessed, and the average of the experimental data yields the trustworthy outcomes.

RESULTS AND DISCUSSION

Influence of the Banana Content on the Composites' Mechanical Properties

The mechanical properties of the composites were found to be significantly impacted by the fiber content (wt%). Figure 1 displays the tensile strength (TS) and bending strength (BS) values of untreated banana fiber composites at various fiber loadings. It was found that TS and BS rose as fiber loading increased up to 30%; however, TS and BS dropped for composites with 40% fiber loading (Figure 1). It turns out that the TS and BS of the untreated composites with a 30% banana content were 39.2 and 45.6 MPa, respectively.

Figure 2 illustrates the results of a study on the effects of fiber loading on the tensile modulus (TM) and bending modulus (BM) of banana-polypropylene composite. For every 10% increase in fiber loading up to 40%, the TM and BM of PP increased steadily. The increase was brought about by the natural fibers' first reinforcement effect [23], which permitted stress transfer from the continuous polymer matrix to the scattered fiber phase. The composites were determined to have heights TM and BM of 0.90 and 0.94 GPa, respectively. In untreated banana fiber reinforced PP composites, the variation in Charpy impact strength (IS) with fiber loading is displayed in Figure 3. Up to 30% fiber loading caused the IS to rise; yet, composites with 40% fiber loading produced lower IS values than those with 30% fiber loading.

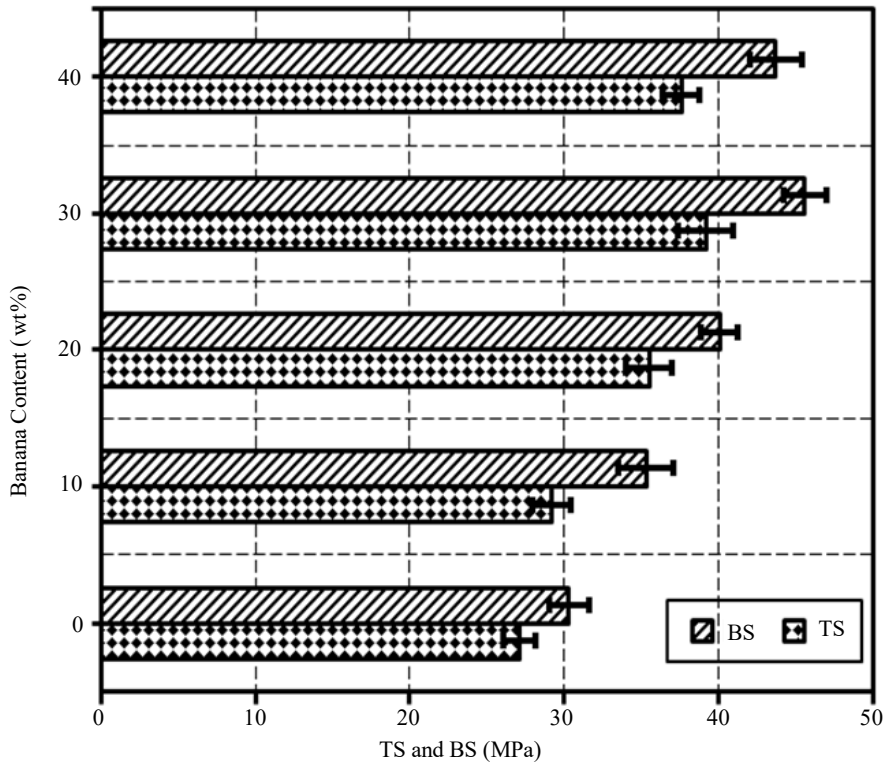

Figure 1. Effect of fiber loading on banana fiber/polypropylene (PP) composites' tensile and bending strengths.

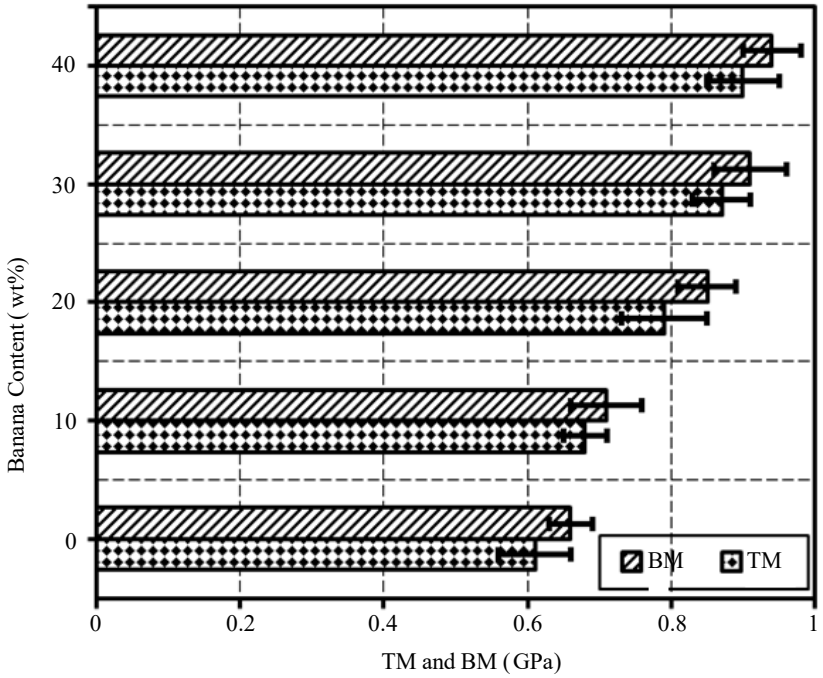

Figure 2. Effect of fiber loading on banana fiber/polypropylene (PP) composites' tensile and bending modulus.

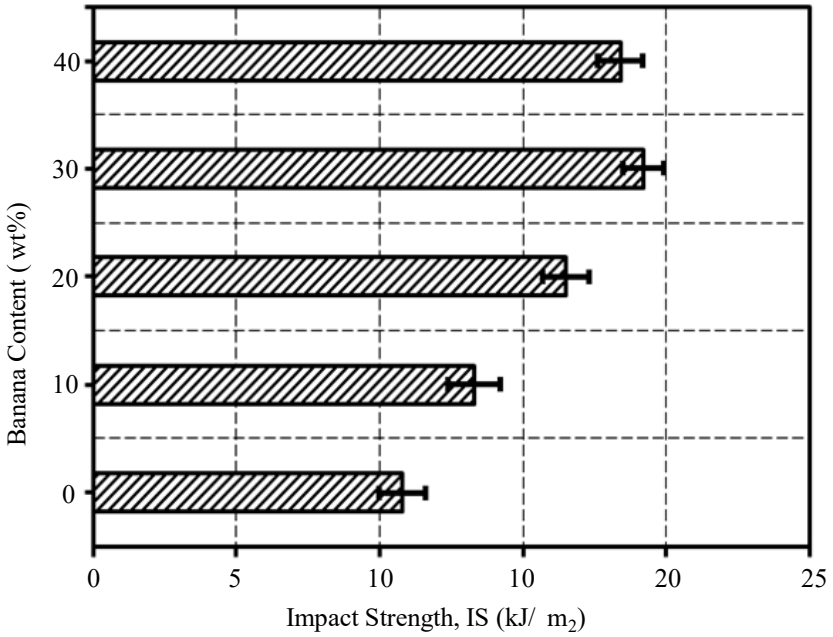

Figure 3. Effect of fiber loading on banana fiber/polypropylene (PP) composites' impact strength.

The height value of IS was found to be 19.2 kJ/m^2 at 30% banana content in the composite. Therefore, it was determined that the mixture with 30% banana content was optimum. The composites' TS, BS, TM, BM, and IS, at 30 wt% of banana fiber loading, shown improvements of 47%, 51%, 43%, 38%, and 78%, respectively, in comparison to the virgin PP matrix. The observed improvement in mechanical characteristics indicates that banana fibers possessing a high aspect ratio and uniform cross section can effectively facilitate the transformation of stress from the matrix. When compared to 30 wt% of banana fiber loading, the degree of mechanical performance gain at low banana fiber loading is less because

fiber orientation is a big factor in the composites' performance. A greater free area for fiber movement as a result of poor fiber alignment at lower fiber loading lowers the effective stress transfer from fiber to matrix [24]. Moreover, fiber loading at 10 wt% may not be sufficient to adequately strengthen the PP matrix and transfer stress since a minimum critical weight ratio fiber loading is needed to reinforce the matrix. The key weight percentage of banana fiber loading in this study is 30 wt%. At this weight, the fibers are sufficient to constrain the matrix, resulting in a uniform distribution of stress and providing the matrix with effective reinforcement. However, it was shown that fiber loading decreased mechanical characteristics above 30 wt% by 4%, 4.2%, and 4.2% in TS, BS, and IS, respectively. Poor adhesion between the fibers and matrix, which encouraged microcrack formation at the interface, and non-uniform stress transfer as a result of fiber aggregation in the matrix could be the cause of the strength loss at increased fiber loading [24]. Increased fiber loading also causes voids to occur in the composites as a result of fiber-fiber agglomeration brought on by H-bonding between the fibers, which causes the PP matrix's mechanical characteristics to deteriorate. Conversely, the TM and BM of PP showed a continuous rise with fiber loading from 10 to 40 wt%, indicating a reinforcing effect. Fiber breakage and pullout, delamination and disbanding, and crack initiation and propagation in the resin matrix are all factors in the failure process that is reflected in the impact response of fiber reinforced composites. The fibers' interaction with the crack formation and function as a stress-transferring medium can account for the rise in notched impact strength. A decline in impact strength was noted at high fiber loadings of 40 wt% because the composites contained a large number of fiber ends, which may cause cracks to form and ultimately lead to composite failure [25]. The breakdown of composites can be explained similarly to the likelihood of fiber aggregation that occurs in areas of stress concentrations where a fracture propagates with less energy.

Impact of Stearic Acid Treatment on the Composites' Mechanical Characteristics

Figures 4 and 5 visually represent the findings of an evaluation of the coupling agent's (SA) impact on the static mechanical characteristics (TS, BS, TM, BM, and IS) of 30 wt% banana fiber reinforced PP composites (BFRP). When comparing the mechanical parameters of all samples with the unidirectional composites without SA at 30 wt% fiber loading, there was an additional improvement with the addition of SA. When compared to 30 wt% reinforced BFRP without SA, the maximum TS and BS of 30 wt% reinforced BFRP at 10 wt% of SA loading rose by 10% and 7%, respectively (Figure 4). When compared to those of the 30 wt% BFRP composites, it was discovered that the highest IS of unidirectional banana fiber/PP composites rose by 36% as a result of the SA coating treatment on the fiber surface (Figure 4). However, as compared to those of the BFRP composites at 30 wt% fiber loading, the maximum TM and BM of the banana fiber/PP composites improved by about 49% and 55%, respectively, as a result of the SA coating treatment on the fiber surface, as shown in Figure 5. The presence of hydroxyl and other polar groups in the fiber surface causes weak interfacial adhesion between the hydrophilic fiber and the hydrophobic PP matrix, which in turn causes debonding and lower mechanical performance of the untreated BFRP composites. But when SA was added, it interacted with the hydrophilic banana through its carboxylic group, giving the surface of the abaca some degree of hydrophobicity and making the banana compatible with the hydrophobic PP. Acid-base interactions allow SA to interact with the fiber surface in a suitable way. Better matrix to fiber stress transmission results from enhanced adhesion and contact between the fibers and matrix, which can be achieved either by covalent bonding, acid-base interaction (like H-bonding), or a combination of the two. In addition to chain entanglement between SA and PP chains, SA provides possible covalent interaction between the hydroxyl and carboxylic groups of the banana fiber, resulting in an effective stress transmission at the interface. Additionally, according to Bisanda and Ansell [26], surface treatment of natural fiber enhances the adhesive characteristics of banana fiber by eliminating hemicelluloses. This creates a rough surface topography that enhances the mechanical properties and adhesion of the fiber-matrix interface. However, a decline in the BFRP's mechanical performance was noted when the weight percentage of SA increased from 10 to 13 wt%. This is primarily due to the SA chains' self-entanglement with one another rather than with the polymer matrix or the fibers, which causes slippage at the interface and lowers mechanical performances [25].

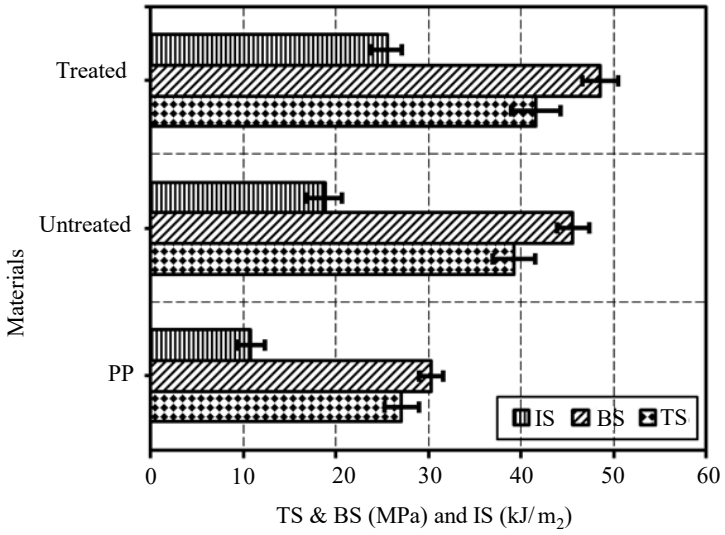

Figure 4. Treated- and untreated-level tensile, bending, and impact resistances of banana/polypropylene (PP) composites.

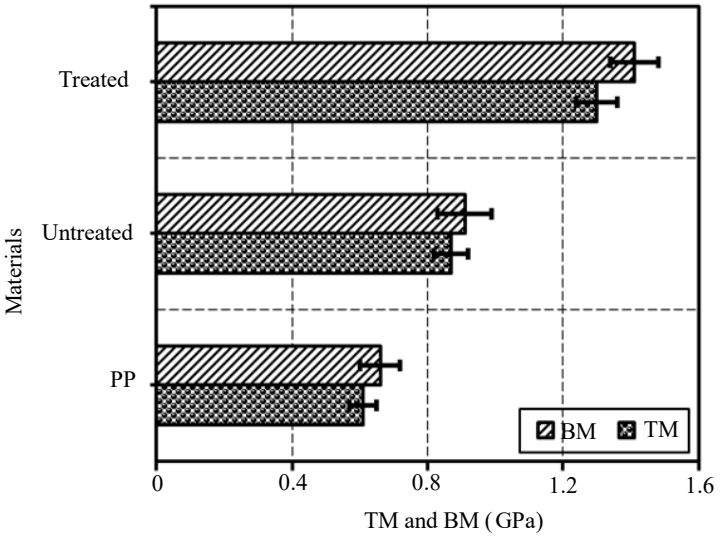

Figure 5. Treated- and untreated-level tensile and bending modulus of banana/polypropylene (PP) composites.

Fractured Surface Morphology

To comprehend the fracture mechanism, a scanning electron micrograph (SEM) of the composites samples' tensile surface was collected. Information regarding a SA's impact on the composites' interphase can be obtained by scanning electron microscopy examination of the fracture surfaces. The tensile fractographs for the treated and untreated composites at 30 weight percent fiber loading are shown in Figure 6(a) and (b). Poor dispersion was observed in the untreated fiber composites, as illustrated in Figure 6a, where the fibers were observed to clump together into bunches and numerous holes were left behind following fiber pullout from the matrix due to the application of tensile stress. However, enhanced adhesion and fiber dispersion were seen in treated fiber composites (Figure 6b), which led to a decrease in the frequency of fiber pullouts and voids because the fibers did not completely emerge from the matrix. In addition, the distribution of the broken surfaces was more uniform than it was for the untreated fiber composites. Using SA coating chemicals to modify its hydrophilic surface, lignocellulosic banana fiber was employed to create unidirectional banana fiber/PP composites. The hydroxyl group (-OH) of the banana and the carboxylic group of SA interacted to improve the adhesion between the fiber and matrix. Better mechanical properties were demonstrated by these composites.

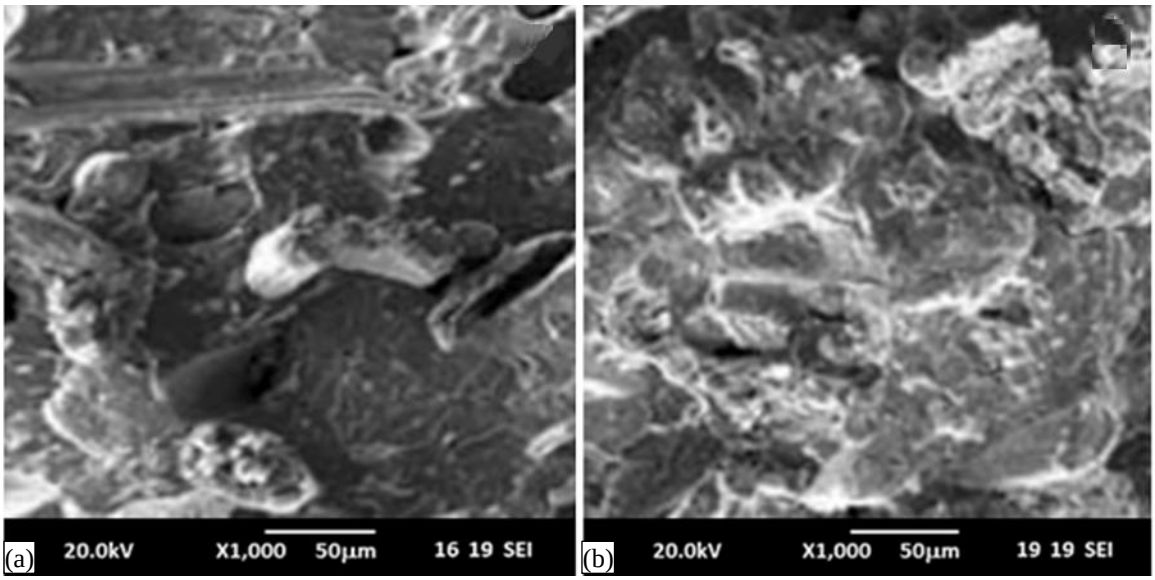

Figure 6. Tensile fracture surfaces (with 30% by weight of fiber loaded) as seen in a scanning electron micrograph: Untreated banana fiber composite (a) and stearic acid (SA)-treated banana fiber composite (b).

The Composites' Water Absorption

It is well known that the absorption of water by lignocellulosic-based composites results in unfavorable dimensional changes in the finished product [27]. Water absorption can also quickly result in debonding, delamination, and a loss of structural integrity [28], which deteriorates the mechanical capabilities of the material. The composite tensile strength diminishes with longer immersion times in water [29].

The volume and kind of lignocellulosic filler a composite made of lignocellulosic materials used greatly influences how much water it absorbs in water immersion tests. It follows that a higher absorption of water is anticipated for composites that comprise lignocellulosic material, such banana fiber. Figure 7 displays the produced composites' water absorption characteristics when subjected to fiber loading. The only composite samples taken are the untreated and SA-treated samples in order to provide better comparison.

With an increase in fiber loading up to 40 wt%, the untreated composite's water absorption (%) rose [30]. When the fiber loading was 40 wt%, the untreated composite absorbed 1.71% of the water, but the sample treated with SA was only able to absorb 0.68%. Relative to the untreated composites, it was found that the SA pre-treated composites contained less water. This could be because of the influence of the polarity of the functional groups: the hydroxyl group in the cellulose molecule of virgin banana fiber is polar and absorbs moisture from the atmosphere; on the other hand, the carboxylic group in stearic acid is less polar than the hydroxyl group. Consequently, the pre-treated composites absorbed less water, which could enhance the dimensional stability of the composites.

The Effect of Simulated Weathering

An investigation on the environment's degrading properties was done using the weathering effect. The impact of weathering on various physical parameters was investigated using two composite samples, namely the untreated and the SA-treated samples. In this regard, a rigorous weathering test was conducted on both treated and untreated samples for a duration of 300 hours, replicating alternate cycles of sunshine and condensation. The weathering tester was equipped with a humidity control system, water spray, and a high-intensity mercury or xenon arc. Periodically, the samples' tensile properties-TS and TM, in particular-were assessed. Table 1 displays the TS and TM value losses.

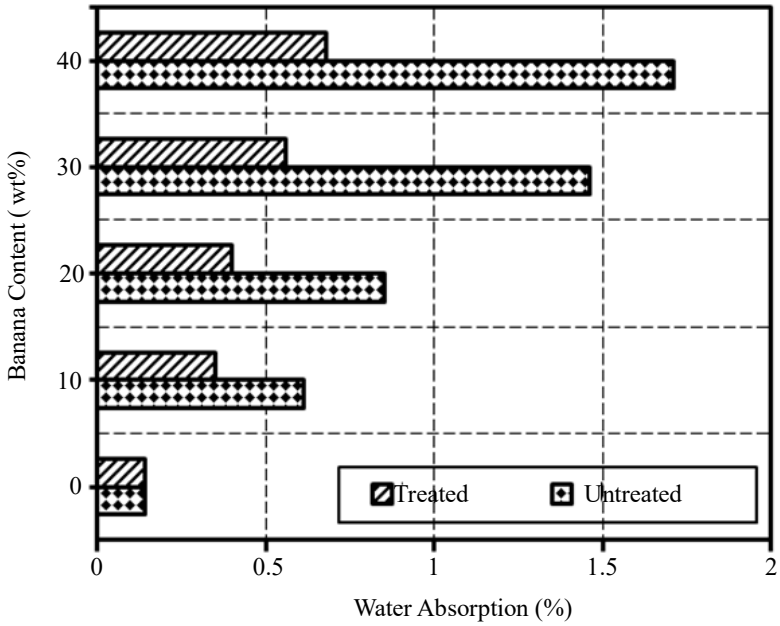

Figure 7. Water absorption variations between untreated and stearic acid (SA)-treated banana fiber/polypropylene (PP) composites at varying fiber loadings.

Table 1. Deterioration of tensile strength (TS) and tensile modulus ™ in both treated and untreated composites during simulation weathering test.

Materials	Degradation time (h)	Degradation in medium	
		LTS (%)	***LTM (%)***
UBPC	50	8.4 ± 0.6	9.3 ± 0.4
	100	11.3 ± 0.5	10.1 ± 0.6
	150	13.3 ± 0.4	12.4 ± 0.5
	200	15.7 ± 0.5	14.3 ± 0.5
	300	16.8 ± 0.7	14.7 ± 0.6
TBPC	50	4.1 ± 0.04	5.3 ± 0.5
	100	6.3 ± 0.06	6.6 ± 0.6
	150	7.8 ± 0.05	8.3 ± 0.5
	200	9.6 ± 0.07	8.7 ± 0.4
	300	11.4 ± 0.8	9.4 ± 0.5

PP, polypropylene; UBPC, Untreated banana/PP composite; TBPC: treated banana/PP composite, LTS, loss of TS (%); LTM, loss of TM (%).

CONCLUSIONS

The mechanical properties of PP-based unidirectional composites reinforced with banana fiber were investigated in this work at different fiber weight percentages. The study asserts that SA covers the banana fiber to make it hydrophobic so that it can be utilized with the hydrophobic PP matrix. The experimental results of the study lead to the following conclusions:

1. The composite materials exhibited an increase in tensile strength, bending strength, and impact strength up to a 30 wt% increase in fiber loading. On the other hand, the tensile and bending moduli of the materials showed a consistent rise as the fiber loading approached 40 wt%.
2. The mechanical characteristics of the treated banana fiber reinforced composites showed that their values for tensile strength, bending strength, impact strength, tensile and bending moduli were greater than those of the virgin polypropylene and even higher than those of the raw composites.

3. Scanning electron micrographs of the fracture surface of the composites showed that the improved tensile properties displayed by the composites with SA-treated banana fiber could be attributed to changes in the fiber's structural properties as well as changes in the fiber/matrix interfacial properties in the case of strength.
4. The treated composite exhibited a significantly lower trend in its water uptake behavior as compared to the untreated composite. Simulated weathering experiments revealed that the treated sample's tensile strength and tensile modulus may be lower than the untreated sample's, based on the amount of time needed for their degradation.

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