Effect of Fiber Ratio and Chemical Treatment on Properties of Banana and Betel Nut Fiber Reinforced Hybrid Polypropylene Composites

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Abstract. The term hybrid composite refers to the composite containing more than one type of fiber material as reinforcing fillers. It has become attractive structural material due to the ability of providing better combination of properties as compared to single fiber composite. In present research, banana and betel nut fiber reinforced polypropylene composites were prepared by hot pressing technique. Raw banana and betel nut fiber were chemically treated with sodium hydroxide to increase adhesion of those fibers with polypropylene. Both raw and alkali treated fibers at 15 wt% were utilized during composite preparation. Banana and betel nut fiber ratios were varied at 1:1, 3:1 and 1:3. Scanning electron microscopic and thermogravimetric analysis, mechanical (tensile, flexural, impact and hardness) and water absorption tests of prepared composites were subsequently conducted. According to microscopic analysis, composites containing treated banana and betel nut fiber at 3:1 ratio had the best adhesion between the fibers and matrix. Polypropylene composites reinforced with banana and betel nut fiber 3:1 ratio had highest tensile and flexural properties, while composites reinforced with banana and betel nut fiber 1:3 ratio showed highest impact properties. According to water absorption test, higher banana fiber containing composite had higher water absorption as compared to higher betel nut fiber containing composite. Polypropylene composites reinforced with equal amount of banana and betel nut fiber had highest thermal stability among all prepared composites. Sodium hydroxide treatment of fibers increased flexural and hardness properties of composites as compared to raw fiber composites. On the other hand, opposite trend was observed in case of tensile, impact and water absorption properties.

1. Introduction
For many years composites have shown many advantages for its light weight, high corrosion resistance and many other qualities. Among the composites fiber reinforced polymer composites have grown popular now a days because of the good compatibility between the fiber and the matrix and light weight of fiber as well as the polymer. Polymer composites have been used in many important fields like automobile, aircraft, bridge, construction etc. and many other daily life uses. Natural fiber reinforced composites give some extra advantages like natural fibers are really abundant in nature, they are
biodegradable so they do not cause any environmental harm, they are extremely light in weight, they have higher strengths than the matrix polymer. The experiment was done by using two kinds of natural fibers as reinforcement and polypropylene as matrix. Banana fiber and betel nut fiber were used as the reinforcing agent which are abundant in nature and have higher strength. Growing attention is being paid to banana fiber as a promising reinforcing material because it is relatively inexpensive, abundantly available and the energy requirement in their extraction is low, which has a considerable engineering advantage [1]. Betel nut husk (BNH) fiber is among a newly explored type of agro-waste fiber. The BNH fiber is extracted from betel nut fruit after a 5-day water retting process. The observed tensile properties of BNH fiber are comparable to kenaf fiber, but with significantly higher elongation at break values [2]. Polypropylene is a thermoplastic polymer and can be made by polymerizing molecules. Scientists prefer thermoplastic matrices than thermostets due to low production cycle, lower cost of processing and high reparability of thermoplastics [3]. In present research, betel nut and banana fiber were added to the polypropylene matrix as reinforcing materials to improve its properties. Mechanical and water absorption tests and microscopic and thermogravimetric analysis of the composites were performed to examine the differences among the polypropylene composites containing different ratio of banana and betel nut fiber. Effect of sodium hydroxide treatment of banana and betel nut fiber on properties of prepared composites was also evaluated.

2. Experimental Procedure

2.1 Materials
Polypropylene was used as matrix and banana fiber and betel nut fiber were used as reinforcement in present research. Polypropylene was white in color and granular in size and its melting point was 160°C. It was collected from market. Banana fibers were collected from bark and betel nut fibers were collected from fruit.

2.2 Alkali treatment
Both banana fiber and betel nut fiber were treated with 5% NaOH to remove impurities and other greasy contents. Fibers were immersed in two separate beakers and kept at room temperature for about two and half hours. Then fibers were washed with distilled water until all NaOH were removed. Fibers then placed in an oven at 100°C for about two hours.

2.3 Composite preparation
Composites were prepared using hot press technique. Initially fiber loading was fixed at 15 wt%, while banana fiber and betel nut fiber ratio was varied at 1:1, 3:1 and 1:3 during composite preparation. Treated hybrid fiber reinforced composite was prepared using banana fiber and betel nut fiber ratio of 3:1. To produce composite fibers were cut into 3 mm size. Fibers and PP were weighted and dried in oven at 80°C for 20 minutes. Fibers were mixed properly. Then releasing agent (silicone spray) was sprayed over the clean mold surfaces. Then PP, fibers, PP layers were made and covered by other silicone sprayed mold and placed in hot press machine. This machine was hydraulic type having capacity of maximum load of 50 KN and maximum temperature of 300°C. The fiber matrix mixture was allowed to press at 30 KN pressure and temperature was initially raised to 160°C and hold there for 20-25 minutes. After that temperature was raised to (190-195)°C depending on the thickness required. Since the compression temperature was higher than PP melting point (160°C) but lower than the fibers melting point (>220°C), so only matrix melted. Then water was passed through the dies and cooled to room temperature. The pressure was released and product was withdrawn from the die.

2.4 Mechanical tests
Tensile, flexural, impact and hardness tests were conducted. For each test, three sample were prepared, tested and average value was taken. Tensile test were carried out according to ASTM D 638-01 using an instron machine (system Id3369J8567, maximum capacity 50 KN). The three point flexural test was
carried out according to ASTM D 790-00 using the same instron machine. The Charpy impact test of the composite was conducted using an impact tester MT 3016 and Specimen was prepared according to ASTM D 6110-97. Hardness was measured using shore durometer in its Shore D scale.

2.5 Water absorption test
Weight of the specimens were taken then immersed in a beaker of water for 24 hours at room temperature. Wet specimens weight were taken after removing excess water. Percentage of water absorption was then calculated.

2.6 Scanning electron microscopy
Surface morphology of the prepared composites was observed by using a scanning electron microscope. The surface of the fiber was made conductive by giving platinum coating using a sputtering machine. The fiber was then observed in vacuum condition into the SEM machine.

2.7 Thermo gravimetric analysis
TGA was carried out in a thermo gravimetric analyzer of model TGA Q50 W/FMC. Its temperature range is ambient +5ºC to 800ºC and heating rate is 0.1 to 100ºC/min. Temperature range of 25ºC to 500ºC and a constant heating rate of 10ºC/min was used in present research.

3. Results and Discussion

3.1 Tensile properties
Figure 1 presents tensile properties of prepared composites against fiber ratio and chemical treatment. It can be observed that for 3:1 banana:betel nut fiber reinforced polypropylene composite, tensile strength was found higher (Figure 1(a)). This is because banana has higher cellulose content as compared betel nut fiber and higher cellulose content fiber has higher tensile strength [4]. Thus higher amount of banana fiber promoted higher tensile strength to the composite. Young’s modulus of banana fiber is also higher than that of the betel nut fiber [2, 4]. So, higher concentration of banana fiber incorporation demands higher stress for the same deformation and results into the increase of Young’s modulus with increased banana fiber ratio [5, 6] (Figure 1(b)).

![Figure 1. Variation of (a) tensile strength and (b) Young's modulus against banana and betel nut fiber ratio and chemical treatment.](image)

Figure 1 also shows that due to the alkali treatment tensile strength and Young’s modulus decreased. This may be because of the change in cellulose structure [7]. The alkali treatment transforms the cellulose I structure partially to cellulose II structure which is amorphous and this results in poor tensile properties of the fiber. Besides damages in the fiber structure such as increase in deep pores and thinning of the fiber cell walls might occur due to the alkali treatment,
which in turn reduced the tensile properties [7, 8]. Again, the alkali treatment partially removes hemicelluloses and lignin which act as the binder substance to cement the microfibrils together. So, the removal of these substances might have created less rigid and less dense fiber and decreased the resistance of the microfibrils of the natural fiber relative to stretching [9-11]. The range of tensile strength of prepared composites was 19.5-21.5 MPa, while the same of hybrid composites containing either banana or betel nut fiber found elsewhere was 18.0-23.0 MPa [12, 13]. The range of Young's modulus of prepared composites was 900-2300 MPa, while the same of hybrid composites containing either banana or betel nut fiber found elsewhere was 680-1600 MPa [12, 13].

3.2 Flexural properties
Variation of flexural properties against fiber ratio and chemical treatment is shown in Figure 2. In Figure 2 (a) it can be observed that for 3:1 banana-betel nut fiber reinforced polypropylene composite, flexural strength increased by 14.56%, which is higher than 1:3 banana-betel nut fiber reinforced polypropylene composite for which flexural strength increased by 14.07%. It is due to higher amounts of cellulose in banana than betel nut [14]. Though alkali treatment increased interfacial bonding, excess treatment increased brittleness, thus flexural strength decreased. In Figure 2 (b) it can be observed that for 3:1 banana-betel nut fiber reinforced polypropylene composite, flexural modulus increased by 20.05%, which is higher than 1:3 banana-betel nut fiber reinforced polypropylene composite for which flexural modulus increased by 16.86%. After treatment modulus increased due to increase in brittleness. The range of flexural strength of prepared composites was 37-42 MPa, while the same of hybrid composites containing either banana or betel nut fiber found elsewhere was 31-42 MPa [12, 13]. The range of flexural modulus of prepared composites was 1300-1700 MPa, while the same of hybrid composites containing either banana or betel nut fiber found elsewhere was 1200-1800 MPa [12, 13].

![Figure 2](image_url)

**Figure 2.** Variation of (a) flexural strength and (b) flexural modulus against banana and betel nut fiber ratio and chemical treatment.

3.3 Impact strength and hardness
Due to incorporation fiber in matrix, void reduces and more force is needed to pull out the fiber. So, Charpy impact strength increases [13, 17]. Betel nut fiber has higher pull out strength than banana fiber [14]. For this reason, by incorporating banana fiber to 75% (fiber ratio 3:1) impact strength increased by 13.33%. Whereas by incorporating betel nut fiber to 75% (fiber ratio 1:3), impact strength increased by 23.33% as shown in Figure 3 (a). After alkali treatment mechanical interlocking develops between fiber and matrix, which minimizes fiber pull out. This decreased
impact strength [14]. When fiber was incorporated into PP, void decreases, flexibility decreases and composite becomes more rigid. Thus, hardness increases. It can be observed in Figure 3 (b) that with increasing banana fiber to 75% (fiber ratio 3:1), hardness decreased by 0.59%, which is negligible. When betel nut fiber was increased to 75% (fiber ratio 1:3), no change in hardness was observed. After treatment, matrix-fiber adhesion increased, so hardness increased slightly (0.42%). The range of hardness of prepared composites was 72.8-73.2 Shore D, while the same of hybrid composites containing either banana or betel nut fiber found elsewhere was 64-75.2 Shore D [12, 13].

![Figure 3. Variation of (a) impact strength and (b) hardness against banana and betel nut fiber ratio and chemical treatment.](image)

3.4 Water absorption characteristics

When fiber reinforced PP is exposed to a process of water absorption, the fiber swells and then micro-cracks occur due to the increased stress at the fiber-matrix interface region. So, water can diffuse more readily through the interface of fiber and matrix [5]. Variation of water absorption of prepared composites is shown in Figure 4. For 3:1 banana-betel nut fiber reinforced polypropylene composite water absorption was found higher because banana has higher amount of cellulose content which increases the hydrophilic nature of fiber [5]. After the alkali treatment, water absorption should have been reduced because of the reduction of cellulose and hemi-cellulose content which are hydrophilic and the improvement of fiber-matrix interface adhesion [8]. However, in present research, treatment might not be done properly and over treating occurred for which micro-gaps was increased due to the removal of interfibrillar matrix material, such as lignin and pectin. For increased micro gaps water could more easily penetrate through the fiber-matrix interface. That is why higher water absorption might occurred after the treatment [6, 18, 19].

![Figure 4. Variation of water absorption against banana and betel nut fiber ratio and chemical treatment.](image)
3.5 SEM analysis
The surface morphology of composites with banana and betel nut fiber at 3:1, 1:1 and 1:3 ratios and treated banana and betel nut fiber at 3:1 ratio was observed under scanning electron microscope and is shown in Figure 5. From the SEM images, better bonding is observed for composites containing banana and betel nut fiber at 3:1 ratio as compared to other two composites. This was due to lower fiber pull out and better adhesion between fibers and matrix in case of higher banana fiber reinforced composite (Figure 5 (c)). Alkaline treatment increases fiber surface roughness resulting in better mechanical interlocking. It also increases the amount of cellulose exposed on the fiber surface, thus increasing the number of possible reaction sites. Also the main purpose of alkali treatment is to disrupt hydrogen bonding in the network structure and remove some hemicellulose, lignin, wax and oils, thereby increasing surface roughness and reducing its hydrophilic nature [19]. SEM micrographs in Figure 5 (d) show better bonding of fiber with matrix in case of treated fiber composite.

![SEM micrographs](a) (b) (c) (d)

Figure 5. SEM micrographs of raw banana and betel nut fiber ((a) 1:3, (b) 1:1, (c) 3:1) and (d) treated banana and betel nut fiber (3:1) reinforced PP composites.

3.6 TGA results
From the TGA curves (Figure 6), it can be stated that an initial weight loss of 2.15% was observed in untreated 1:1 banana-betel nut fiber reinforced composite at around 231.4°C. For 3:1 and 1:3 ratio of banana-betel nut fiber reinforced composite, the initial weight loss was found to be 2.6% and 2.7% respectively at temperature of 229.5°C. So, among the three ratios, the 1:1 ratio of banana- betel nut fiber reinforced composite has the highest thermal stability. The initial weight loss is also higher for the 3:1 and 1:3 ratios; that may happen because of the presence of slightly increased amount of water. Among the 3:1 and 1:3 ratio, the 1:3 ratio of banana-betel nut fiber reinforced composite has higher initial weight loss because betel nut has higher amount of hemicelluloses which makes it more hydrophilic. NaOH catalyses pyrolysis process, Thus thermal stability decreased in case of NaOH treated fiber reinforced PP. So, the thermal degradation started to occur at lower temperature in comparison with the untreated fiber reinforced PP. The amount of cellulose decomposition or the derivative weight (%) change of cellulose got decreased in the NaOH treated fiber reinforced PP than the untreated 3:1 and the 1:3 banana-betel nut fiber reinforced PP. This is because the treatment decreased the amount of cellulose and changed the structure of the remained cellulose, which was more reactive than the previous structure. In chemical
treatment, the hydrogen of hydroxyl group of cellulose was substituted by acetyl group, which is more reactive [5]. Exposure of more reactive cellulose components on the fiber surface may have promoted rough surface and more effective surface area for better bonding between fiber surface and polymer matrix via mechanical interlocking mechanism [9]. Thus decomposition of the cellulose got decreased.

![Figure 6. TGA curves of raw banana and betel nut fiber (a) 1:1, (b) 3:1, (c) 1:3 and (d) treated banana and betel nut fiber (3:1) reinforced PP composites.](image)

4. Conclusion
In present research, raw and chemically treated banana and betel nut fiber reinforced polypropylene composites were prepared by hot pressing technique. Both raw and alkali treated fibers at 15 wt% were utilized during composite preparation. Banana and betel nut fiber ratios were varied at 1:1, 3:1 and 1:3. Higher percentage of banana fiber containing composite showed better tensile strength, flexural strength, flexural modulus and Young’s modulus than the untreated ones. Higher percentage of banana fiber containing composite showed higher water absorption than higher betel nut fiber containing composite. 50% banana fiber reinforced composite showed the highest thermal stability.

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