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EFFECTS OF CHEMICAL MODIFICATIONS OF PINEAPPLE LEAF FIBRE ON THE PROPERTIES OF POLYPROPYLENE COMPOSITES

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ABSTRACT

The effects of chemical modifications of pineapple leaf fibre on the mechanical and end-use properties of pineapple leaf fibre filled polypropylene composites were studied within fibre content, 0 – 10 wt. % and particle size, 75 µm. The pineapple leaf fibre was alkali treated using sodium hydroxide (NaOH) and bleached using hydrogen peroxide (H_2O_2). Maleic anhydride-graft-polypropylene (MA - g - PP) was used as a compatibilizer. Analysis of the pineapple leaf fibre showed that it contains an appreciable quantity of cellulose (65.30 %) and has a density of 1.89 g/cm^3, moisture content 6.10 %, oil absorption 1.96 g/100 g, ash content 1.94 %, and pH 6.90. The pineapple leaf fibre composites were prepared in an injection moulding machine. Result showed that unfilled polypropylene exhibited higher tensile strength (39.17 MPa) than the filled polypropylene composites. The addition of MA - g - PP and the use of treated pineapple leaf fibres improved the tensile strength of the composites. The untreated pineapple leaf fibre prepared composites exhibited the least tensile strength and had higher elongation at break than the filled composites. The elongation at break of filled polypropylene composites decreased with increase in fibre content while the addition of MA – g - PP and treated pineapple leaf fibre improved the elongation at break of the composites. Generally, the hardness (BHN), specific gravity, and water absorption indices of the composites increased with increase in fibre content investigated. This study has shown that the use of MA - g - PP, alkali refined, and bleached pineapple leaf fibres in compounding polypropylene increased the hardness and specific gravity of the resulting composites and decreased their water absorption properties. These property improvements should justify the use of pineapple leaf fibre in processing polypropylene composites especially where the improved properties are of utmost consideration.

Keywords: Polypropylene, Composite, Pineapple leaf fibre, Tensile strength, Water absorption, Compatibilizer.

INTRODUCTION

Natural fibres are presently gaining importance in polymer composite production as a result of the increasing demand for environmentally friendly fillers, and the need to reduce the cost of traditional fillers (i.e. carbon, glass, ceramics, etc.). Natural fibre reinforced composites are used in the automobile industry for seat coverings, trunk panels, headliners, dashboards, door panels, and many other industries for domestic and engineering applications.

These fibres, which are available in different countries of the world can be extracted from leaflets (coconut, palm), leaves (sisal, banana), fruit (coir), stems (jute, linen, hemp) and trunk [1, 2]. Natural fibres are characterized by cellulose micro – fibrils present in the amorphous matrix of lignin and hemicellulose that run along the length of the fibres where each fibril resembles a single wood fibre [3]. Natural fibres have the following advantages over the traditional reinforcing fillers: low cost, availability, low density, flexibility during processing, non – abrasive to processing equipment, acceptance specific strength properties, biodegradability, ease of separation, enhanced energy recovery, reduced thermal and respiratory irritation, and high toughness [4][5]. The performance of plant fibres in composite production, however, depends on many factors such as the chemical composition and physical properties of the fibres [6] [7]. The use of natural fibres in reinforcing polymers has been reviewed by many authors [3, 8 – 11].

There are certain disadvantages associated with the use of natural fibres in compounding thermoplastic and thermosetting composites. These have been recognised and include weak compatibility between the fibre and polymer interface, especially at high temperatures, the inherent high water absorption of fibres which result to dimensional instability in composites, lower durability and processing temperature permissible due to the tendering of the fibres to degrade [12, 13]. The problem of lower processing temperature limits the use of natural fibres in compounding thermoplastic commodities such as poly (vinyl chloride), polyethylene, polypropylene and polystyrene. The weak interfacial adhesion between the highly hydrophilic natural fibres and the non - polar hydrophobic polymer matrix leads to considerable decrease in the properties of the composites, thereby, limiting their industrial utilization and production. The poor interfacial adhesion between natural fibres and polymer matrices can be improved by using coupling agents such as maleic anhydride – graft – polyethylene, and surface modifications of fibre surfaces. Surface modification of natural fibres can be achieved by using chemicals such as alkali treatment [14], benzoylation [15], silane [16], acetylation [17], acrylation [18], and isocyanate treatment [19]. However, these fibre surface modification methods vary in performance, and is dependent on the chemical structure of the reagent used [5].

Polypropylene, an olefin polymer is widely utilized in industry when filled for electrical and automotive engineering applications because of its excellent stiffness property which enables it to substitute conventional materials in demanding engineering applications [20]. The interest to compound polypropylene with filler is growing in importance because of improved compounding technology, and new compatibilizing agents that allow the use of high filler content [21]. Thus, the following bio - fillers have been used to compound polypropylene: flax [22], green coconut fibre [23], seaweed fibre [24], kenaf and raw bamboo fibre [25], and cauraua fibre [26].

The present study reports the utilization of chemically treated pineapple leaf fibre in compounding polypropylene. Maleic anhydride – graft - polypropylene was used as a compatibilizer. The pineapple leaf fibre was sieved to 75 mm particle size, and used within filler content, 0 to 10 wt. %. Pineapple leaf presently, has no identifiable industrial utilization in Nigeria, and litters farm yards as wastes. The present investigation is part of our ongoing efforts at putting the usefulness of pineapple leaf fibre as filler in compounding polymers. Previous reports from our laboratory have reported the use of pineapple leaf fibre in compounding natural rubber, and high density polyethylene with encouraging results [27 - 30]. The successful utilization of pineapple leaf fibre in the composite industry will be a source of economic benefit to rural farmers. Fortunately, pineapple leaf fibre is naturally available, of low cost, eco-friendly, and easy to process.

EXPERIMENTAL

Polypropylene

The polypropylene used in this study was obtained from Ceeplast Industries, Aba, Nigeria. It has a density of 0.92 g/cm^3 and melt flow index of 16 g/10 min.

Pineapple Leaf Fibre

The pineapple leaves from where the fibre was prepared were collected from a pineapple orchard near Umuagwo, Owerri, Imo State, Nigeria.

Maleic anhydride – graft - Polypropylene

Maleic anhydride-g-polypropylene (MA - g - PP) was used as a compatibilizer in this study. It was obtained from Rovet Scientific Limited, Benin City, Nigeria and is a product of Sigma Aldrich, USA. The maleic anhydride content of the MA - g - PP is approximately 8 – 10 wt. %, and has a density of 0.934 g/cm^3.

Preparation of Materials

Pineapple Leaf Fibre

Pineapple leaves collected from Umuagwo, Imo State, Nigeria was cut into smaller sizes, and sun dried for fourteen days. The dried leaves were later oven dried for 24 h at 80^{o}C prior to grinding. A manual grinder was used to grind the chopped dry pineapple leaves into powdery form. The pineapple leaf fibre obtained was sieved to 75 µm particle size and stored in a tight lid container for subsequent use.

Fibre Treatment

The pineapple leaf fibre was alkali treated and bleached according to the method of Rosa *et al*. [31].

Bleaching

500 g of pineapple leaf fibre was immersed in 1000 cm^3 solution containing 150 cm^3 (30%, w/w) H_2O_2 and 16 g NaOH at 85^{o}C and stirred for 1 h. The fibre was subsequently washed with distilled water several times and dried in an oven at 50^{o}C to a constant weight.

Alkali Treatment

1000 g of pineapple leaf fibre was immersed in 1000 cm^3 of 10% (w/w) NaOH solution for 3 h at 70^{o}C with occasional shaking followed by washing with distilled water several times to remove any absorbed alkali. The water used in washing the fibre was tested with a red litmus paper. The alkali was removed completely from the fibre when the litmus dipped in the water was neutral. The alkali treated fibre was placed in a hot air oven maintained at 70 ^{o}C, and later cooled and kept in a desiccator for subsequent use.

Analysis on Pineapple Leaf Fibre

The pineapple leaf fibre was analyzed for the following using standard methods: ash content (ASTM D 4607 - 94), moisture content (ASTM D – 1509 - 13), oil absorption (ASTM D 1510, 1983), pH (ASTM D 1512 - 05). The chemical composition of pineapple leaf fibre was determined as described previously [28].

Preparation of Polypropylene Composites

The pineapple leaf fibre filled polypropylene composites were prepared using the formulation scheme shown in Table 1. The composites were prepared by thoroughly mixing high density polypropylene with the appropriate pineapple leaf fibre quantities as shown in Tables 1 and 2. Maleic anhydride-graft-polypropylene was used as a compatibilizer and was used at 3% based on the weight of pineapple leaf fibre.

Table 1. Composition of polypropylene composites.

S/N	PP content (g)	Fibre Content (g)		
		Untreated PLF	Bleached PLF	Alkali treated PLF
1	200	0.0	0.0	0.0
2	197.5	2.5	2.5	2.5
3	195.0	5.0	5.0	5.0
4	192.5	7.5	7.5	7.5
5	190.0	10.0	10.0	10.0

Table 2. Composition of treated pineapple leaf fibre / polypropylene composites.

S/N	Untreated PLF (g)	MA – g – PP (g)
1	197.5	0.15
2	195.0	0.50
3	192.5	0.75
4	190.0	1.00

Tests on Polypropylene Composites

The following tests were performed on polypropylene composites using standard methods: tensile tests (ASTM D 638), water absorption (24 h)(ISO 180), specific gravity(ASTM D 792), and hardness (ASTM D 2240).

RESULTS AND DISCUSSION

Physical and Chemical Analysis of Pineapple Leaf Fibre

The results of physical and chemical analysis of pineapple leaf fibre (PLF) are shown in Tables 3 and 4 respectively. The pH of aqueous slurry of pineapple leaf fibre was determined to be 6.90; an indication that the fibre is slightly acidic. The acidity of a filler has a bearing on its reinforcing effect; the more acidic a filler is, the lower the reinforcement on composites [16]. The moisture content of 6.10% recorded for pineapple leaf fibre is due to its organic origin. High moisture content in a filler often leads to weak filler-matrix adhesion, hence, poor mechanical strength properties of the composites.

Table 3. Physical properties of pineapple leaf fibre.

Property	Content
Ash (%)	9.40
Moisture (%)	6.10
pH of aqueous slurry	6.90
Density (g/cm^3)	1.96
Oil absorption (g/100 g)	1.89

The chemical composition of pineapple leaf fibre presented in Table 4 shows the presence of appreciable quantity of cellulose (65.3%) and low lignin content (13.2%). The higher mechanical property of pineapple leaf fibre is related to its high cellulose content. The results of PLF based polymer composites show excellent stiffness and strength properties as compared to other cellulose based composite material [17].

Table 4. Chemical composition of pineapple leaf fibre.

Property	Content(%)
Ash	2.8
Cellulose	65.3
Holocellulose	71.6
Lignin	3.2
Extractives	2.6

Mechanical Properties of Polypropylene Composites

Tensile Strength

Fig. 1 shows the effect of pineapple leaf fibre content on the tensile strength of polypropylene composites. The tensile strength of unfilled polypropylene is 3.71 MPa. From Fig. 1, it is evident that the tensile strength of polypropylene composites decreased with increase in filler content and are lower than the tensile strength of unfilled polypropylene. The decrease of tensile strength with increase in filler content may be due to heterogeneous dispersion of pineapple leaf fibre in polypropylene matrix, and incompatibility of polypropylene and pineapple leaf fibre (PLF). The heterogeneous dispersion of pineapple leaf fibre in the polypropylene matrix would cause a very significant stress condition which would lead to the formation of micro voids and quick development into crack during deformation [18].

The decrease in tensile strength with increase in filler content observed in this study is in agreement with the findings of Vu *et al*. [32] and Mctlenry and Stachunki [33] who reported that the tensile strength of polypropylene composites decreased with fibre contents. Eze *et al*. [34] who worked on pineapple leaf fibre filled high density polyethylene reported increases in tensile strength of the composites with filler content and attributed the findings to better filler dispersion in the polymer, and filler-matrix interactions.

The effects of treated pineapple leaf fibre, and compatibilizer on the tensile strength of polypropylene composites are also shown in Fig. 1. From the figure, it was observed that the treatment of the fibres and the addition of MA - g - PP during polypropylene compounding appreciably increased the tensile strength of polypropylene composites within the filler content investigated but the values were lower than the tensile strength of unfilled polypropylene.

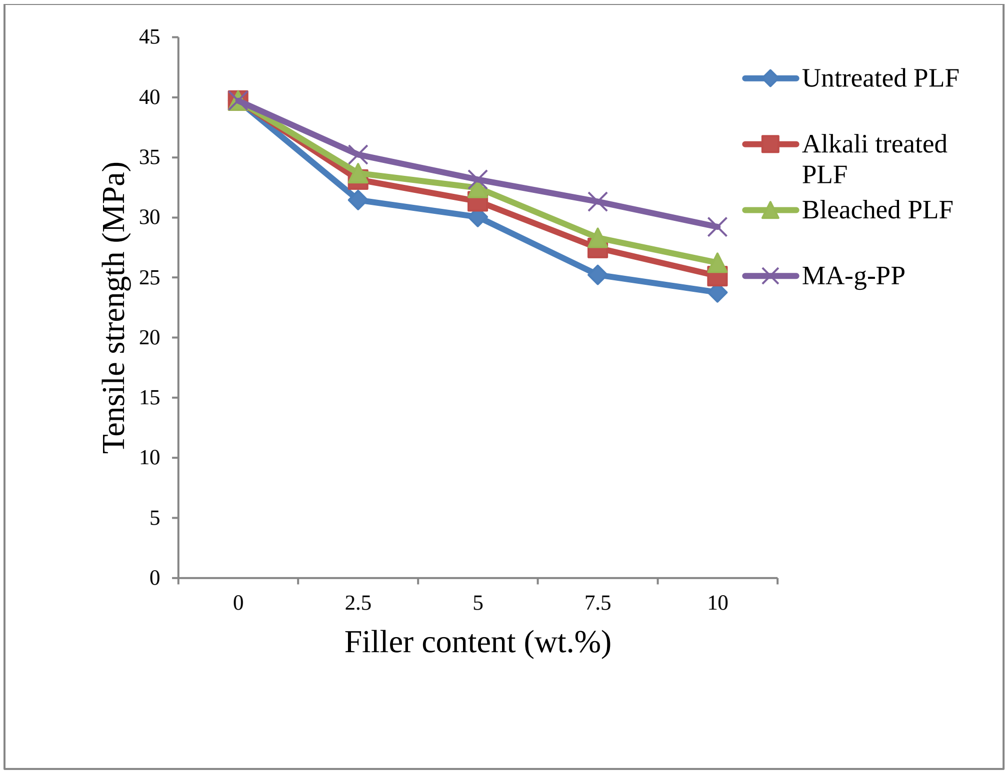

Fig. **1** Effect of pineapple leaf fibre content on tensile strength of polypropylene composites.

It has been reported that alkali treatment helps in the removal of lignin and hemicelluloses which affect the tensile characteristics of fibres [20]. Sodium hydroxide and hydrogen peroxide treatments removed the hydrogen bonding in the network structures of the fibres thereby increasing the fibre surface softness [24]. When the treated fibres are stretched, there are rearrangements among the fibrils resulting in better load sharing, and hence, higher stress development in the fibres. The addition of 10 wt.% MA – g - PP (based on fibre content) the increased the tensile strength of the composites for all the fibre contents investigated; an indication that MA – g – PP can be used as compatibilizer in processing pineapple leaf fibre/polypropylene composite. These findings are attributed to better interfacial adhesion between the fibre and polymer matrix arising from better chemical modification of the interface between the fibre and polypropylene [24]. MA - g - PP has a hydrophilic group that is compatible with the fibre and a hydrophobic group that is compatible with polypropylene for improved adhesion in composites. This property enables it to react with the surface of polypropylene to form bonds. The determined chemical composition of pineapple leaf fibre showed that it contains cellulose and hemicelluloses, both polar groups which are capable of bonding with maleic anhydride-graft-polypropylene for improved adhesion in composites. The observed order on the improvement of tensile strength of polypropylene composite is: MA-g-PP > bleached PLF > alkali treated PLF > untreated PLF.

Elongation at Break

Fig. 2 shows the effect of fibre content on the elongation at break of pineapple leaf fibre/polypropylene composites. From the figure, it is seen that the elongation at break for all the composites decreased with increase in fibre content. Elongation at break of untreated pineapple leaf fibre/polypropylene composite was found to decrease by 44.24% at 2.5 wt.% fibre content and 54.96% at 10.0 wt.% fibre content. Generally, fillers can be considered as structural elements embedded in polymer matrices, and at the concentrations considered (0 – 10 wt.%), the fibre content investigated was not sufficient to restrain the polypropylene molecules thereby resulting in a matrix that was not ductile. Similarly, poor filler/ matrix interaction or incompatibility can be responsible for the poor ultimate performance. Some workers have reported the decrease in elongation at break of composites with increase in filler contents [28, 35].

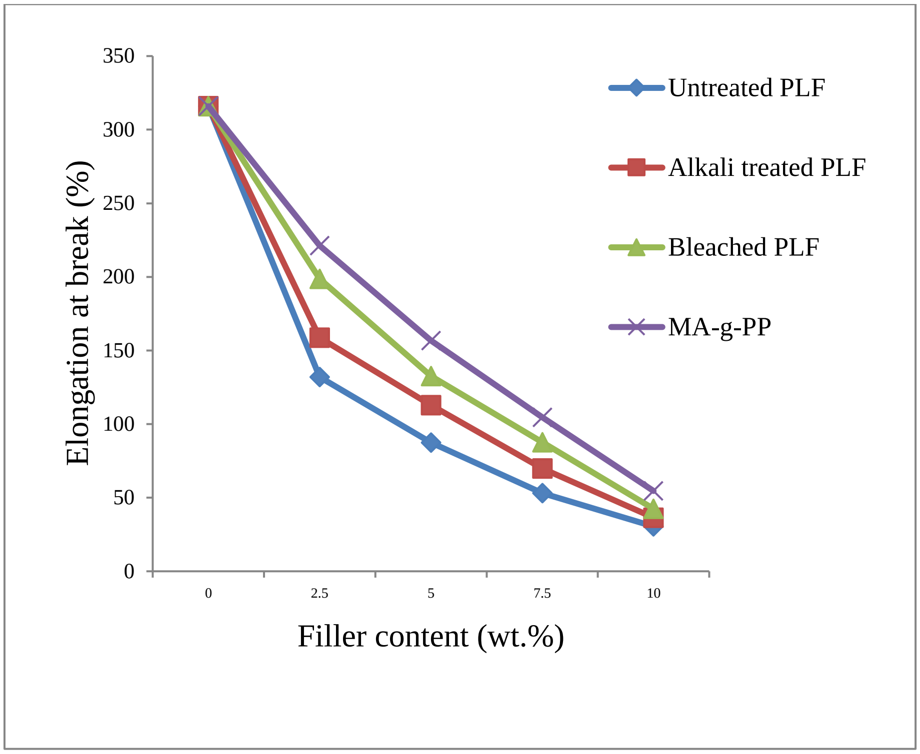

Fig. **2** Effect of pineapple leaf fibre content on elongation at break of polypropylene composites.

The elongation at break of pineapple leaf fibre/polypropylene composites were improved significantly especially within fibre contents, 0 – 5 wt.%. The use of treated pineapple leaf fibre and addition of MA - g - PP into the polypropylene matrix increased the elongation at break of the composites. This result is attributed to improved adhesion between the pineapple leaf fibre and the polypropylene matrix which resulted to increases in the elongation at break of the composites. Generally, surface treatment and addition of compatibilizer make fillers to become stiffer than untreated fillers [26]. Therefore, the combined effects of surface treatment and compatibilizer make the composite to be stiffer (higher modulus).

Hardness

Fig. 3 shows the variation of the hardness (BHN) of polypropylene composites with fibre content. The hardness number of unfilled polypropylene is 30.1. There is a general increase in the hardness of the composites with fibre content both for the treated and untreated fibre, and in the presence of maleic anhydride - graft – polypropylene. The increases were more pronounced at high fibre content (10.0 wt.%). Wang *et al*. [36] reported that the addition of fillers into polymer matrices makes the resulting composites to be harder because fillers are naturally stiffer than polymers.

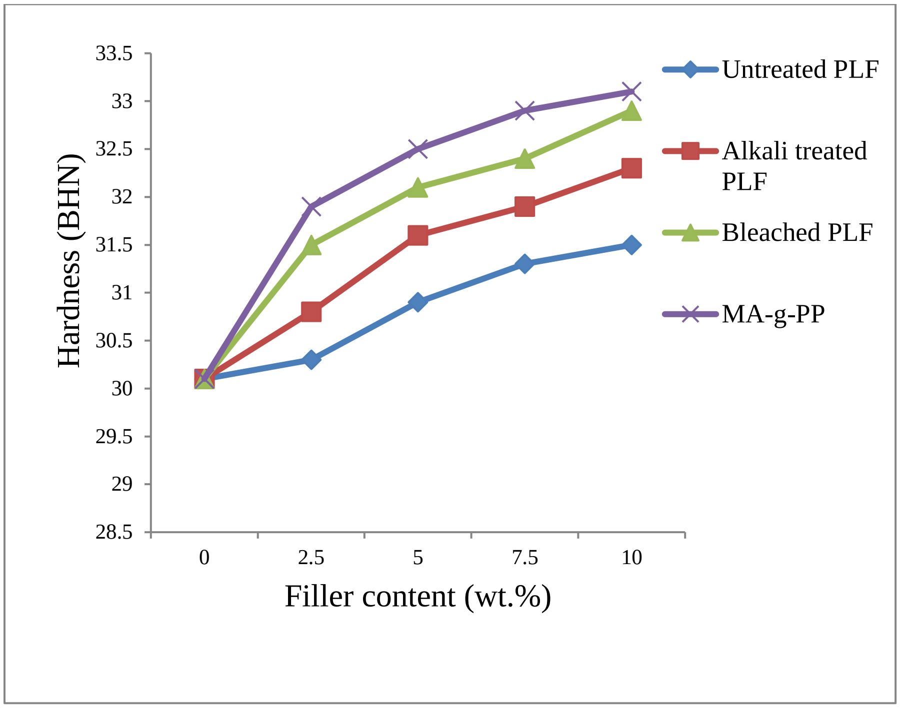

Fig. **3** Effect of pineapple leaf fibre content on hardness of polypropylene composites.

Thus, as the composites become stiffer with increasing fibre content, the hardness increased. Such increase in the hardness of polypropylene composites with increasing fibre content has been reported [37]. The addition of MA - g - PP into polypropylene matrix increased the hardness of the composites more than what was obtained for the treated and untreated fibres. The general order on the improvement of the hardness of polypropylene composites is: MA - g - PP > bleached PLF > alkali treated PLF > untreated PLF. The addition of MA - g - PP and the use of treated fibres must have appreciably increased the interfacial bonding between the polypropylene/matrix interface and the pineapple leaf fibre resulting to increased composite hardness.

Specific Gravity

Data on the specific gravity of polypropylene composites are illustrated graphically in Fig. 4. The specific gravity of untreated polypropylene composite is 1.38. Fig. 4 shows a general increase of the specific gravity of the composites with fibre content. All the filled polypropylene composites exhibited higher specific gravity than the unfilled at 2.5 – 10.0 wt. %fibre content investigated in the study The untreated pineapple leaf fibre composites exhibited higher specific gravity than the alkali treated, and bleached pineapple leaf fibres.

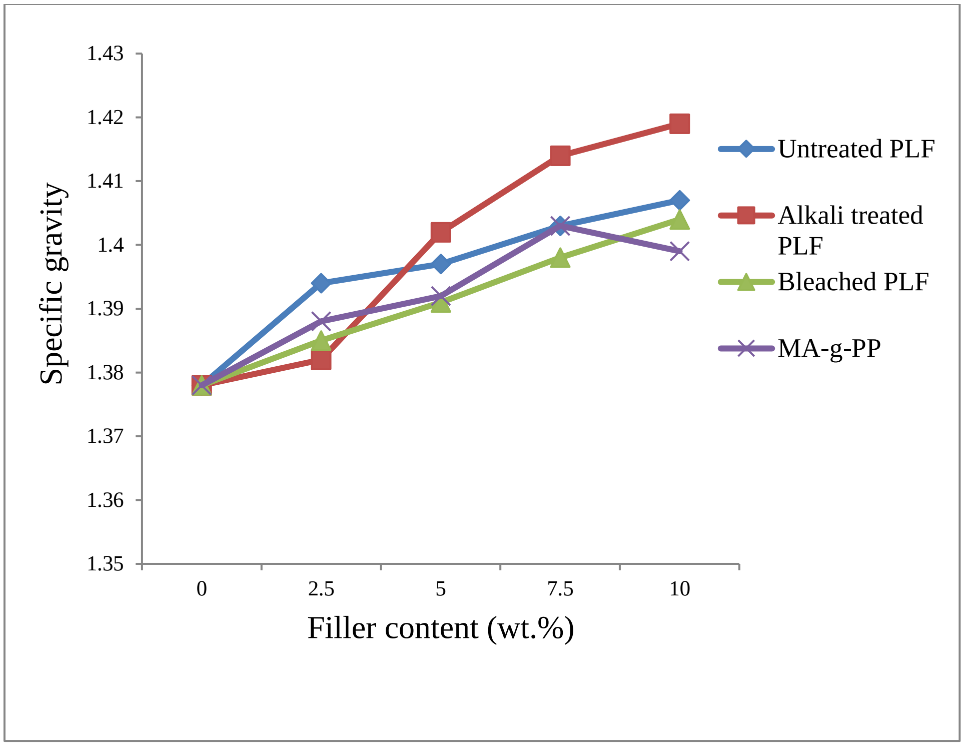

Fig. 4 Effect of pineapple leaf fibre content on the specific gravity of polypropylene composites.

The MA - g - PP processed polypropylene composites exhibited increases in specific gravity between fibre contents, 2.5 – 7.5 wt.%, and thereafter, the specific gravity decreased at 10 wt.% fibre content. At 2.5 wt.% fibre content, the alkali treated pineapple leaf fibre composite had the least specific gravity while at 7.5 and 10 wt.% fibre content, it exhibited higher specific gravity than the other processed polypropylene composites. The high specific gravity of the polypropylene composites obtained with alkali treated pineapple leaf fibre may be attributed to improved interaction, and consequent adhesion between the polymer matrix and the fibre. The specific gravity of polypropylene composites obtained in this study within the fibre contents investigated (0 – 10.0 wt.%) is higher than that of a 40 % (wt/wt) glass – polypropylene composite which is 1.23 [13].

Water Sorption

The water sorption (24 h cold water) indices of pineapple leaf fibre filled polypropylene composites are illustrated graphically in Fig. 5. All the polypropylene composites generally showed increases in water absorption with increase in fibre content. The hydrophilic nature of pineapple leaf fibre is believed to be responsible for the water absorption in the composites. Polypropylene has small water absorption index (3.46). The use of MA - g - PP decreased the amount of water absorbed by composites. This decrease in water absorption is attributed to some hydrophilic-OH group present in the fibres reacting with acid anhydride of maleic anhydride - graft - polypropylene to form ester linkages with the resultant decrease in water absorption. Alkali treated and bleached pineapple leaf fibre composites equally showed decreases in water absorption indices when compared to the untreated fibre at any fibre content considered in this study. Haque *et al*. [38] in their studies on coir fibre filled polypropylene found that the amount of water absorbed by the composites increased with coir content and was dependent on the type of chemical treatment of coir fibre. Karina *et al*. [39] who studied kenaf, banana, water hyacinth, wood, and oil palm empty fruit bunch fibre filled polypropylene reported that water absorption by recycled and virgin polypropylene composites was due to the presence of hydrophilic fibres in the composites. The order in the amount of water absorbed by the composites in this study is: untreated PLF > bleached PLF > alkali treated PLF > MA - g - PP compatibilizer.

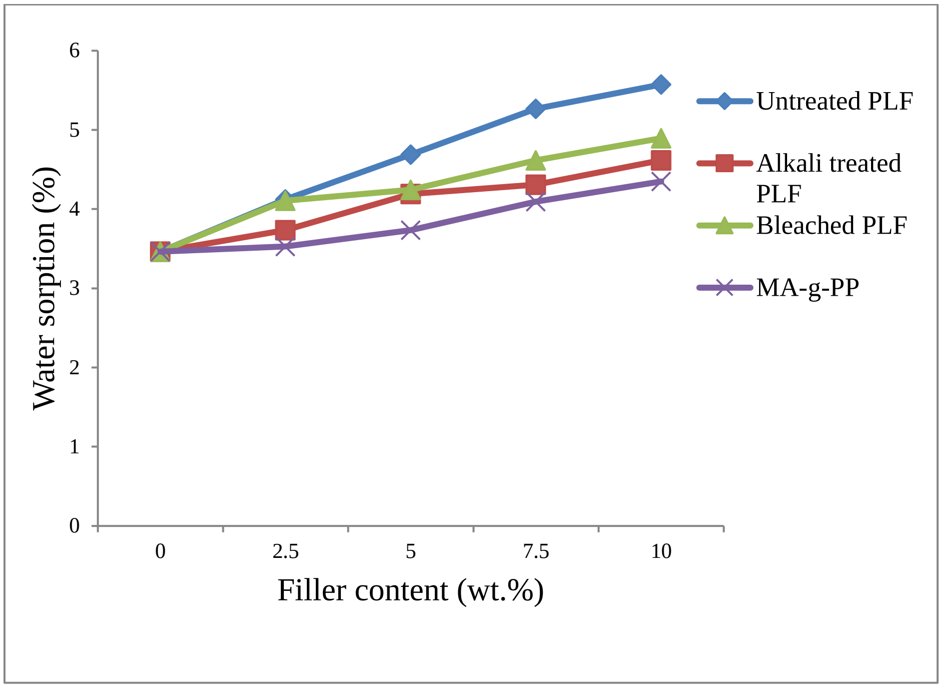

Fig. 5 Effect of pineapple leaf content on water absorption of polypropylene composites.

CONCLUSION

This study has shown that chemical modification of pineapple leaf fibre using NaOH and H_2O_2 affected the mechanical and end-use properties of polypropylene composites.

1. The tensile strength and elongation at break of the untreated pineapple leaf fibre filled polypropylene composites decreased with increase in fibre content. The hardness, specific gravity and water absorption indices of the untreated pineapple leaf fibre filled polypropylene composites increased with increased fibre content.
2. The use of MA - g - PP compatibilizer, alkali treated, and bleached pineapple leaf fibre improved the tensile strength, elongation at break, hardness, and specific gravity of polypropylene composites and decreased their water absorption property.
3. MA - g - PP, alkali treated, and bleached pineapple leaf fibre filled polypropylene composites exhibited higher hardness and specific gravity than the untreated pineapple leaf fibre filled polypropylene composites.
4. The MA - g - PP filled polypropylene composites absorbed the least amount of water followed by the alkali treated, bleached, and untreated pineapple leaf fibres in that order.

The present study has demonstrated the utility of pineapple leaf fibre in compounding polypropylene composites. The properties of the composites were improved upon on the incorporation of MA – g – PP, alkali treated, and bleached pineapple leaf fibres into polypropylene. Fortunately, pineapple leaf which is the source of pineapple leaf fibre is locally available in quantity. The fibre is not toxic, easy to process, does not abrade the processing equipment, and is biodegradabe. Furthermore, the successful utilization of pineapple leaf fibre in compounding polypropylene will be a source of economic benefit to the rural farmers where this crop grows.

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