

A Review on the Mechanical Properties of Natural Fibre Reinforced Polypropylene Composites

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Abstract: This paper is a review on the mechanical properties of natural fibre reinforced polymer composites. Natural fibres have recently become attractive to researchers, engineers and scientists as an alternative reinforcement for fibre reinforced polymer (FRP) composites. Due to their low cost, fairly good mechanical properties, high specific strength, non-abrasive, eco-friendly and bio-degradability characteristics, they are exploited as a replacement for the conventional fibre, such as glass, aramid and carbon. The tensile properties of natural fibre reinforced polymers (both thermoplastics and thermo sets) are mainly influenced by the interfacial adhesion between the matrix and the fibres. Several chemical modifications are employed to improve the interfacial matrix-fibre bonding resulting in the enhancement of tensile properties of the composites. In general, the tensile strengths of the natural fibre reinforced polymer composites increase with fibre content, up to a maximum or optimum value, the value will then drop. However, the Young's modulus of the natural fibre reinforced polymer composites increase with increasing fibre loading. Khoathane et al. [1] found that the tensile strength and Young's modulus of composites reinforced with bleached hemp fibers increased incredibly with increasing fiber loading.

Keywords: Polymer-matrix composites (PMCs), Mechanical properties, Mechanical testing Compression moulding

INTRODUCTION

A fibre reinforced polymer (FRP) is a composite material consisting of a polymer matrix imbedded with high-strength fibres, such as glass, aramid and carbon [2]. Generally, polymer can be classified into two classes, thermoplastics and thermosettings. Thermoplastic materials currently dominate, as matrices for bio- fibres; the most commonly used thermoplastics for this purpose are polypropylene (PP), polyethylene, and poly vinyl chloride (PVC); while phenolic, epoxy and polyester resins are the most commonly used thermosetting matrices [3]. In the recent decades, natural fibres as an alternative reinforcement in polymer composites have attracted the attention of many researchers and scientists due to their advantages over conventional glass and carbon fibres [4]. These natural fibers include flax, hemp, jute, sisal, kenaf, coir, kapok, banana, henequen and many others [5]. The various advantages of natural fibres over man-made glass and carbon fibres are low cost, low density, comparable specific tensile properties, nonabrasive to the equipments, non- irritation to the skin, reduced energy consumption, less health risk, renewability, recyclability and biodegradability [3]. These composites materials are suitably applicable for aerospace, leisure, construction, sport, packaging and automotive industries, especially for the last mentioned application [3, 6]. However, the certain drawback of natural fibres/polymers composites is the incompatibility between the hydrophilic natural fibres and the hydrophobic thermoplastic matrices. This leads to undesirable properties of the composites. It is therefore necessary to modify the fibre surface by employing chemical modifications to improve the adhesion between fibre and matrix [3] here are many factors that can influence the performance of natural fiber reinforced composites. Apart from the hydrophilic nature of fibre, the properties of the natural fibre reinforced composites can also be influenced by fibre content / amount of filler. In general, high fibre content is required to achieve high performance of the composites. Therefore, the effect of fibre content on the properties of natural fibre reinforced composites is particularly significance. It is often observed that the increase in fibre loading leads to an increase in tensile properties [7]. Another important factor that significantly influences the properties and interfacial characteristics of the composites is the processing parameters used. Therefore, suitable processing techniques and parameters must be carefully selected in order to yield the optimum composite products. This article aims to review the reported works on the effects of fiber loading, chemical treatments, manufacturing techniques and process parameters on tensile properties of natural fiber reinforced composites.

- **Tensile Properties**

Generally, the tensile properties of composites are markedly improved by adding fibers to a polymer matrix since fibers have much higher strength and stiffness values than those of the matrices. [3, 8].

In general, higher fiber content is desired for the purpose of achieving high performance of short fiber reinforced polymer composites (SFRP) [7]. It is often observed that the presence of fiber or other reinforcement in the polymeric matrix raises the composite strength and modulus [5]. Therefore, the effect of fiber content on the tensile properties of fiber reinforced composites is of particular interest and significance for many researchers [7].

Nonwoven mats from hemp and polypropylene fibers in various proportions are mixed and hot pressed to make composite materials. The effect of hemp fibre content and anisotropy are examined on the basis of tensile properties of the resultant composite materials. The tensile strength, with fibres in the perpendicular direction, tended to decrease with increasing hemp fibre content (a maximum decrease of 34 % at 70 % of hemp) as depicted in Figure 1. Whereas, the tensile strength, with fibres in the parallel direction, showed a different trend and a maximum value was found with increasing fibre loading. It was found that the tensile strength of composites with fibres in the perpendicular direction was 20 – 40 % lower than those of composites with fibres in parallel direction. Since the fibres lay perpendicular to the direction of load, they cannot act as load bearing elements in the composite matrix structure but become potential defects which could cause failure. As expected, better tensile properties are found in the specimens cut from the composite sheets parallel to the direction of carding as depicted. [9].

From the above citations and discussions, it can be found that the values of the tensile strength of natural fibre reinforced composites increased with increasing fibre loading up to a maximum or optimum value before falling back. However, it is generally true that the values of the Young's modulus increased progressively with increasing fibre loading. On the other hand, some researchers found totally the opposite trend to the increase of composite strength with increasing fibre content. This can be attributed to many factors such as incompatibility between matrix and fibers, improper Manufacturing processes, fiber degradation and others.

The hydrophilic nature of natural fibers is incompatible with hydrophobic polymer matrix and has a tendency to form aggregates. These hydrophilic fibers exhibit poor resistant to moisture, which lead to high water absorption, subsequently resulting in poor tensile properties of the natural fiber reinforced composites. Moreover, fiber surfaces have waxes and other non-cellulosic substances such as hemi-cellulose, lignin and pectin, which create poor adhesion between matrix and fibers. Therefore, in order to improve and develop natural fiber reinforced polymer composites with better tensile properties, it is necessary to increase fibers' hydrophobicity by introducing the natural fibers to surface chemical modification (surface treatment). The fiber modification is attempted to improve fibers hydrophobic, interfacial bonding between matrix and fiber, roughness and wettability, and also decrease moisture absorption, leading to the enhancement of tensile properties of the composites [13-17]. The different surface chemical modifications, such as chemical treatments, coupling agents and graft copolymerization, of natural fibers aimed at improving the tensile properties of the composites were performed by a number of researchers. Alkali treatment, also called mercerization, is one of the most popular chemical treatments of natural fibres. Sodium hydroxide (NaOH) is used in this method to remove the hydrogen bonding in the network structure of the fibres cellulose, thereby increasing fibres surface roughness [13]. This treatment also removes certain amount of lignin, wax and oils covering the external surface of the fibres cell wall, depolymerises the native cellulose structure and exposes the short length crystallites [14]. Acrylic acid treatment was also reported to be effective in modifying the natural fibres surface. A study on flax fibres-reinforced polyethylene biocomposites by Li et al. found that the efficiency of such a treatment was higher than alkali and silane treatment [14]. The chemical coupling method is also one of the important chemical methods, which improve the interfacial adhesion. In this method the fiber surface is treated with a compound that forms a bridge of chemical bonds between fiber and matrix. The chemical composition of coupling agents allows them to react with the fiber surface forming a bridge of chemical bonds between the fiber and matrix. Most researchers found these treatments were effective and showed better interfacial bonding [13]. Among different coupling agents, maleic anhydride is the most commonly used. In general, the literature reports improvements in tensile strength and elongation at break when maleic anhydride grafted matrices are used as compatibilizers (coupling agent) [15].

Hu and Lim [18] investigated that alkali treatment significantly improved the tensile properties of hemp fiber reinforced polylactic acid (PLA) compare to those untreated. Figures 14 and 15 showed that the composites with 40% volume fraction of

alkali treated fibre have the best tensile properties. The tensile strength and tensile modulus of the composites with 40% treated fiber are 54.6 MPa and 85 GPa respectively, which are much higher than neat PLA, especially for the tensile modulus which is more than twice of that of neat PLA (35 GPa).

Fuqua and Ulven reported that fibre loading of treated (alkali and bleached) and untreated flax fiber without compatibilizer (maleic anhydride grafted polypropylene or MAPP) in PP composites caused inferior tensile strength (even compared with pure PP) [19]. However, treated fiber loading with compatibilizer resulted in favourable tensile strength as depicted in the continuously increased trend of composite modulus can be found in all cases (untreated, bleached and treated) and reached a maximum value at 65/5/30 (% wt PP/MAPP/ fiber loading) [19]. This can be argued that the introduction of alkali treatment with 5% MAPP in the natural fiber reinforced plastic composites helped to improve both tensile strength and Young's modulus of the composites compare to those without MAPP.

Liu et al. evaluated the effects of different fiber surface modifications, 2%NaOH, 2+5%NaOH (Note that 2+5% NaOH treatment is a continuation treatment from 2%NaOH process and then soaked with 5% NaOH) and coupling agent, on jute / polybutylene succinate (PBS) biocomposites [20]. The experiment results showed that surface modifications could remove surface impurities, increased surface roughness and reduced diameter of jute fiber, subsequently, significantly increased the tensile strength and modulus of the composites but decreased breaking elongation as depicted in Figures 18 through 20. It was observed that the biocomposites of jute fibers treated by 2%NaOH, 2+5%NaOH or coupling agent, obviously had their tensile properties increased when compared to those untreated and yielded an optimum value at fiber content of 20 wt%. The results also showed that the strength and stiffness of composites were dependent on the types of treatment. In Figures 21 and 22, the 100/0/0 referred to w/t % of PP (100%), MAPP (0%) and fibre loading (0%); while 65/5/30 referred to w/t % of PP (65%), MAPP (5%) and fibre loading (30%).

Li et al. [14] studied flax fiber reinforced polyethylene biocomposites. In the study, flax fibers, containing 58 w/t % of flax shives were used to reinforce polyethylene (high density polyethylene and linear low density polyethylene). The composites contained 10 w/t % of fibre and processed by extrusion and injection molding. Five surface modification methods, alkali, silane, potassium permanganate, acrylic acid, and sodium chlorite treatments, were employed to improve the interfacial bonding between fibers and matrix. Figures 21 (LLDPE) and 22 (HDPE) showed that the biocomposite tensile strengths were increased after surface modifications. Among these surface modification techniques, acrylic acid was found to be a relatively good method in enhancing tensile properties of both flax / HDPE and LLDPE biocomposites [14].

Fuqua and Ulven investigated the different MAPP loading (0, 5 and 10 w/t %) effects on tensile properties of corn chaff fiber reinforced polypropylene composites [19]. They also investigated the effect of various treatments, silane z-6011, silane z-6020 and 5 w/t % MAPP, on corn chaff fiber & distilled dried grains (DDGS) reinforced polypropylene composites [19]. It was found that 5 w/t % MAPP yielded the optimum value for the composites in term of tensile strength and modulus as shown in Figures 23 and 24 respectively [19]. The strength reduction observed with high MAPP loading was caused by the interaction between the compatibilizer (MAPP) and the fibre/matrix system. The anhydride units of MAPP maintain loop confirmations within the composite systems, since they all can act with equal probability with the cellulose in the corn fibers. Coupled with MAPP's low average molecular weight, the interaction between the PP matrix and MAPP becomes dominated principally by Van der Waals' forces; since chain entanglement of PP and MAPP is virtually impossible. MAPP that is not utilizes for fibre/matrix adhesion and is therefore mechanically harmful to the composites, which leads credence to the significant performance variation between 5 and 10 w/t % loadings. However, through the use of 5 w/t % MAPP, it was found that the tensile properties of the composites increase, especially tensile strength compared to neat resin and those untreated.

Sain et al. investigated the effect of a low-molecular weight MAPP on tensile properties of polypropylene reinforced with the varieties of natural fibers such as old newsprint, kraft pulp and hemp [20]. Figures 25 and 26 showed that the optimum level of the coupling agent (MAPP) by weight of the old newsprint-filled PP composites was 4 percent for tensile strength and 1.5 percent for tensile modulus respectively [20].

Another important factor that significantly influences the properties and interfacial characteristics of the composites is processing techniques and parameters used. Common methods for manufacturing natural fibre reinforced thermoplastic composites are extrusion-injection moulding and compression moulding. Tungjitpornkull and Sombatsompop researched on the difference in the tensile properties of E-glass fiber (GF) reinforced wood/PVC (WPVC) composites, manufactured by twin screw extrusion and compression moulding processes respectively [22]. The experimental results suggested that the

GF/WPVC composites produced from compression moulding gave better tensile modulus than those from their counterparts as depicted in Figure 27. The shear stress in compression moulding was lower than that in twin screw extrusion, as a result there was less thermal degradation of PVC molecules and less breakage of glass fiber, resulting in longer fibre length in the composites manufactured by compression moulding. The composite manufactured by compression moulding would have higher specific density, which resulted in less void and air and was then stronger than its counterpart [22]. The study by Siatong et al. aimed to determine the optimum values for fiber content by mass (0%, 12.5% and 25%), extrusion barrel zone temperatures (75-110-120-130-140 °C and 75-120-130-140-150 °C) and extrusion screw speed (110 and 150 rpm) for the production of flax fiber reinforced polyethylene (HDPE and LLDPE) composites [23]. Response surface methodology was applied as optimization technique over three response variables: density deviation (%), tensile strength (MPa) and water absorption (% mass increase) of the composites. According to statistical analysis, the optimum values that yield the highest tensile strength (17.09 MPa for LLDPE composite and 21.70 MPa for HDPE composite) were: fiber content of 6.25%, barrel zone temperatures of 75-116-126-136-146 °C and screw speed of 118 rpm for LLDPE composites, and fiber content of 5%, barrel zone temperatures of 75-118-128-138-148 °C and screw speed of 128 rpm for HDPE composites. The optimum values of temperatures (T) were closer to the higher levels (75-120-130-140-150 °C) because lower temperatures result in inconsistent melt of resin that can lead to non-uniform dispersion of the fibers in the composites and eventually lower the tensile strength. The optimum values of screw speed were closer to the lower level (110 rpm). This was because the higher screw speed led to shorter residence time, non-uniform dispersion of fibers, high porosity, and consequently, lowers tensile strength. However, the unexpected result was the very low optimum level of the fiber content. Theoretically, an increase of flax fibers should improve the mechanical properties of the composites, yet, the results of tensile strength negated this [23].

Li et al. determined the appropriate value of injection temperature and pressure for flax fiber reinforced high-density polyethylene biocomposites. The results showed that higher fiber content in composites led to higher mechanical strength [24]. Injection temperature of lower than 192 °C was recommended for better composite quality because at higher temperature, fibre degradation (fibre degradation temperature \approx 200 °C) might have occurred, therefore, lead to inferior tensile properties. However, the injection temperature should not be lower than 160 °C in order to ensure adequate melting of matrix. In comparison with injection temperature, the influence of injection pressure was not obvious. However, higher injection pressure is preferred to obtain better composite tensile properties [24].

The optimum pressure was determined for the natural fibre mat (hemp and kenaf) reinforced acrylic resin manufactured by high-tech vacuum compression process. Figure 28 showed that the maximum pressure for the composites was at 60 bars. Above this value, there was a decrease in tensile properties of the composites due to the damage of the fiber structure. The advantages of using vacuum technology are to allow a reduction of the press time to a minimum without decreasing the performance of the cured materials. In addition, the work conditions were significantly improved when the vacuum chamber process was used. [25].

Khondker et al. studied the processing conditions of unidirectional jute yarn reinforced polypropylene composites fabricated by film stacking methods [26]. From optical micrographs obtained, they suggested that there must be an optimum processing temperature for which this composite might perform better in tensile properties. According to the optical microscopy results, they showed that the composites moulded at a temperature of 160 °C for 15 minutes and under 2.0 MPa molding pressure, would have the PP matrix films fused and the PP melted completely and penetrated into the fiber bundles. This temperature was considered favourably ideal for the processing of composites that used lignocellulosic fibers as reinforcement, as most lignocellulosic fibers cannot withstand processing temperatures higher than 175 °C for longer duration, and hence limiting their ability to be used with some thermoplastic resins [26].

The effect of the melting-mixing technique parameters on the tensile properties of sisal fiber reinforced polypropylene composites were optimised by varying the 29 through 32, mixing time of 10 min, rotor speed of 50 rpm and a mixing temperature of 170 °C were found to be the optimum mixing conditions. For mixing times (Figures 29 and 30), below the optimum value, the tensile strength and Young's modulus were low because of ineffective mixing and poor dispersion of the fiber in PP matrix. As the mixing time was increased, melting of PP resin became extensive and resulted in better fiber distribution into the matrix. When mixing time was more than 10 minutes, fiber breakage and degradation would happen, leading to a decrease in tensile properties. For mixing temperatures (Figure 31), the performance of short fiber composites was controlled directly by fiber aspect ratio, quality of dispersion and interface between fiber and polymer. Below the optimum value, viscosity as well as shear stress generated in the mixture was very high, resulting in the break down of fibers to shorter lengths during mixing, leading to a lower tensile strength of the composites. On the other hand, if mixing

temperature was above the optimum, the thermal degradation of fibers would occur, leading to the decrease of tensile properties. For mixing speeds (Figure 32), low tensile strength was observed at speeds lower than the optimum value due to poor dispersion of fibers in molten PP matrix. Above the optimum rotor speed, there was a reduction in strength because of fiber breakage at high rotor speed [27].

4. Discussions and conclusions

The scientific world is facing a serious problem of developing new and advanced technologies and methods to treat solid wastes, particularly non-naturally-reversible polymers. The processes to decompose those wastes are actually not cost-effective and will subsequently produce harmful chemicals. Owing to the above ground, reinforcing polymers with natural fibres is the way to go. In this paper, most of the natural fibers mentioned were plant-based but it should be noted that animal fibres like cocoon silkworm silk, chicken feather and spider silk have also been used and the trend should go on. Those fibres, both animal- and plant-based have provided useful solutions for new materials development, in the field of material science and engineering. Natural fibers are indeed renewable resources that can be grown and made within a short period of time, in which the supply can be unlimited as compared with traditional glass and carbon fibers for making advanced composites. However, for some recyclable polymers, their overall energy consumption during collecting, recycling, refining and remoulding processes have to be considered to ensure the damage of the natural cycle would be kept as minimal. On top of it, Natural fibers are low cost, recyclable, low density and eco-friendly material. Their tensile properties are very good and can be used to replace the conventional fibers such as glass, carbon in reinforcing plastic materials. A major drawback of using natural fibers as reinforcement in plastics is the incompatibility, resulting in poor adhesion between natural fibers and matrix resins, subsequently lead to low tensile properties. In order to improve fiber-matrix interfacial bonding and enhance tensile properties of the composites, novel processing techniques, chemical and physical modification methods are developed. Also, it is obviously clear that the strength and stiffness of the natural fiber polymer composites is strongly dependent on fiber loading. The tensile strength and modulus increase with increasing fiber weight ratio up to a certain amount. If the fiber weight ratio increases below optimum value, load is distributed to more fibers, which are well bonded with resin matrix resulting in better tensile properties. Further increment in fiber weight ratio has resulted in decreased tensile strength as described in the main text. Mathematical models were also found to be an effective tool to predict the tensile properties of natural fibre reinforced composites. conventional fibers such as glass, carbon in reinforcing plastic materials. A major drawback of using natural fibers as reinforcement in plastics is the incompatibility, resulting in poor adhesion between natural fibers and matrix resins, subsequently lead to low tensile properties. In order to improve fiber-matrix interfacial bonding and enhance tensile properties of the composites, novel processing techniques, chemical and physical modification methods are developed. Also, it is obviously clear that the strength and stiffness of the natural fiber polymer composites is strongly dependent on fiber loading. The tensile strength and modulus increase with increasing fiber weight ratio up to a certain amount. If the fiber weight ratio increases below optimum value, load is distributed to more fibers, which are well bonded with resin matrix resulting in better tensile properties. Further increment in fiber weight ratio has resulted in decreased tensile strength as described in the main text. Mathematical models were also found to be an effective tool to predict the tensile properties of natural fibre reinforced composites. Finally, it can be found that the main weakness to predict the tensile properties of plant-based natural fibre composites by modelling was giving too optimistic values. The modelling has to be improved to allow improvements in the prediction of tensile properties of composites reinforced with both plant- and animal-based fibres.

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