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Effect of Chemical Treatment of Fibres on the Mechanical Properties of Natural Fibre Reinforced Polymer Composites

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Abstract—Composite materials consist of two or more constituent materials, the fibre and the matrix. Fibre reinforced polymer composites are becoming very popular and replacing conventional materials nowadays, because of their excellent properties suitable for various applications. The properties of fibre reinforced polymer composites are comparable to most conventional materials like metallic materials. Numbers of papers have been published in the field of high strength composites made up of glass fibres, carbon fibres, Kevlar fibres and carbon nanotubes. However, their utilizations are affected by their high cost, recyclability and biodegradability issues. The concern regarding environmental policies leads researchers to focus on natural fibre based composites. The works of researchers have shown that the drawbacks of natural fibres, such as incompatibility between fibres and matrices and reduced resistance to moisture, can be reduced by chemical modifications and treatments. This review paper summarized the papers that have been done within the last few years to determine the effect of chemical treatment of natural fibre on the mechanical property of natural fibre reinforced polymer composites (NFRPCs). Most of the papers agreed that chemically treating the natural fibres enhances the mechanical properties of natural fibre reinforced polymer composites whereas some papers identified that treating some natural fibres results in negative effect on the mechanical properties of NFRPCs.

Index Terms— Alkali treatment, Chemical Treatment, Composite materials, Mechanical Properties, Natural fibers, Polymer composites, Polymer matrix

1. Introduction

A composite material system is composed of two or more physically distinct phases whose combination produces aggregate properties that are different from those of its constituents. The increase in environmental regulations and unsustainable consumption of petroleum led thinking of use of environmentally friendly materials. Natural fibre is considered as one of the environmentally friendly materials which have good properties compared to synthetic fibres (1). The use of natural fibre, from both resources, renewable and nonrenewable, gained considerable attention in the last decades, so far. The plants, which produce cellulose fibres can be classified into bast fibres, seed fibres, leaf fibres, grass and reed fibres, and core fibres as well as all other kinds [2]. Natural fibre reinforced polymer composites are a composite material consisting of a polymer matrix embedded with high strength natural fibres, like jute, oil palm, sisal, kenaf, and flax [3]. Usually, polymers can be categorized into two categories, thermoplastics and thermosets. The application of natural cellulose fibres as reinforcements in composite materials has finding increased use over the last few years in different automotive applications by many automotive companies, in building and construction industry, sports, aerospace, and others, for example, panels, window frame, decking, and bicycle frame [4].

The attractive features of natural fibres have been their low cost, light weight, high specific modulus and health hazards of composites reinforced with synthetic fibres such as glass, carbon and aramid fibres. These advantages place the natural fibre composites among the high performance composites having economic and environmental advantages [5–7].Several natural fibres such as sisal, jute, coir, bagasse, flax and Luffa cylindrical fibres have been studied as a reinforcement and filler in polymer composites. Those studies were performed to improve the mechanical properties such as tensile strength, impact strength, flexural modulus and etc. Natural fibre-reinforced polymers can exhibit very different mechanical performances and environmental aging resistances depending on their interphase properties. The principal function of the interphase is to facilitate transfer of stress from fibre to fibre, across the matrix [8]. The interfacial bond between the reinforcing fibres and the resin matrix is an important element in realizing the mechanical properties of the composites. Because of the insufficient interface quality between the fibres and the polymer matrix is the first and the most important problem in natural fibre reinforced composites, the surface modification of the fibres by chemical treatments is one of the largest areas of current research. Several authors [1–4, 9] have focused their studies on the treatment of fibres to improve the bonding with resin matrix. A modification of the fibres can either increase or decrease the strength of the fibres, and thus an understanding of what occurs structurally is of great importance [10]. According to my record, there is no work done and it is a novel idea to review solely the papers that revealed the effect of chemical treatment of the mechanical properties of NFRPCs. The main objective of this review paper is to identify and summarize the works of different researchers that studied and revealed the effect of chemical treatment of natural fibre on the mechanical property of NFRPCs.

Chemical Treatments of NFRPCs

In a review of chemical treatments of natural fibres, Kabir et al. [11] concurred that chemical treatment is an important factor that has to be considered when processing natural fibres. They observed that fibres hydroxyl groups due to different chemical treatments, thereby reducing the hydrophilic behavior of the fibres and causing enhancement in mechanical strength as well as dimensional stability of natural fibre reinforced polymer composites. The effects of different chemical treatments on cellulosic fibres that were employed as reinforcements for thermoplastics and thermosets were also examined. The different kinds of chemical treatments used on natural fibres include silane [12], alkali[13], acrylation[14], benzoylation[15], maleated coupling agents [16], permanganate[17], acrylonitrile and acetylation grafting [18], stearic acid [19], peroxide [20], isocyanate [21], triazine [22], fatty acid derivate, sodium chloride and fungal [23]. Their general conclusion was that chemical treatment of natural fibres results in a remarkable improvement of the NFRPCs. This paper summarizes the works of several researchers regarding the effect of chemical treatment of some of the different types of natural fibre reinforced polymer composites.

2. Literature Review

Many researchers have studied about the effect of chemical treatment on mechanical properties of natural fibre reinforced polymer composite materials. The following are some of the papers that have been done within the last decades.

Isiaka Oluwole et al. [24] studied the effect of chemical treatment on the mechanical properties of sisal fibre reinforced polyester composites. Sisal (Agave Sisalana) is a plant that yields stiff fibres traditionally used for rope and twine. It is usually extracted by a process known as decortications. The materials and equipment used for this work were unsaturated polyester resin, Methyl Ethyl Ketone Peroxide (catalyst), Cobalt 2% in solution as accelerator, sisal fibre, KOH, H2O2, NaOH, Poly Vinyl Acetate, and Ethanol. Both chemically treated and untreated fibres were used to develop samples of sisal fibre reinforced polyester composites in a predetermined proportion after which they were tested for mechanical properties. Each treatment with NaOH, KOH, H2O2 and Ethanol respectively was carried out by using 2 molar solutions inside shaker water bath at 70°C for 2 hours. The fibres after treatments were allowed to dry naturally in the laboratory. Then, the fibre strands were cut into the length of the tensile test specimen mould which was 194mm before they are used. The tensile tests were performed on INSTRON 1195 at a fixed Cross head speed of 10mm/min. Samples were prepared according to ASTM D412 (ASTM D412 1983) and tensile strength of standard and conditioned samples were also determined. For the hardness determination, the sample was indented using Durometer following ASTM procedure No.D2240. The result reported that, KOH treated sample of 6g fibre weight has the highest ultimate tensile stress value of 29MPa where as for the untreated sample the highest value of 24MPa. Regarding the hardness testing, it was observed that KOH treated sample of 4% fibre weight has the highest hardness value of 258.59 BHN but for untreated samples the highest value was obtained at 7% fibre weights having the value of 201.28 HBN.

S. M. Sapuan et al. [25] conducted tensile test, flexural test, impact test and hardness test in their experiment and revealed the effect of the alkali (NaOH) treatment on the mechanical properties of pineapple leaf fibre (PALF) reinforced high impact polystyrene (HIPS) composites. The size of pineapple leaf fibre that was used in this research is a 10-40 mesh. The high impact polystyrene (HIPS) that has been utilized as the matrix polymer is the Idemitsu PS HT 50 that has density of 1.04 g/cm3. The short pineapple leaf fibre was soaked in different concentrations (0%, 2% and 4%) of NaOH solution in the water bath for 1 hour at room temperature. The ratio of the fibres and the solution was 1:20 (w/v). After treatment, the fibres were washed and rinsed several times with distilled water. Afterwards the fibres were dried in an oven at 80°C for 24 hours. The tensile test were

conducted following the standard of ASTM D638 type V using Instron (Model 4301) universal testing machine with load cell of 1 kN and using crosshead speed of 1mm/min. The similar Instron universal testing machine (Model 4301) was used for flexural testing. The applied load and the crosshead speed were specified at 1 kN and 1.3 mm/min, respectively, while the support span was 48 mm. For flexural test the samples were prepared and cut into rectangular specimens with 127 mm (L) x 12.7 mm (W) x and 3 mm (T) dimensions. The Izod impact test was carried out for the notched and unnotched samples with dimensions of 63.5 mm x 12.7 mm x 3 mm. This test was carried out according to ASTM D256-93. The result reported that alkali treated fibre composites improved the tensile strength and tensile modulus by 24% and 35%, respectively, compared with the untreated fibre composites. Also, flexural testing shows that increasing alkali concentration increases the flexural strength gradually. The flexural strength of untreated fibre was 31.661 MPa, whereas for the 2% and 4% of alkali concentration treatments 34.371 MPa and 40.789 MPa, respectively. It was also shown that the flexural modulus of untreated fibre is 4294.515 MPa, whereas for 2% and 4% NaOH concentration treatments about 4464.068 MPa and 4559.339 MPa, respectively. In addition to this, the Izod impact test result shows that untreated fibre has a lower value of notched and unnotched impact strength compared to 2% and 4% alkali-treated fibre. The increase of NaOH concentration from 2% to 4% increased the impact strength of composites from 47.164 to 52.417 J/m for notched impact and from 64.615 to 75.959 J/m for unnotched impact, respectively. Regarding the Rockwell hardness result, the experiment was carried out using the Rockwell Hardness Tester HA-101 in accordance to ASTM D 785 procedure B. The Rockwell hardness was measured using a 0.625 mm ball and 60 kg of indent force (Rockwell scale L). With alkali treatment of 2% and 4% NaOH, the hardness value of composites increased. The lowest result of the hardness test, 84.120, derived from the untreated short PALF composite; whereas for the short PALF treated with 2% and 4% NaOH, the obtained results for the hardness test had better values, 89.260 and 90.260.
Contrary results were found by George et al. [26] with the case of different alkali concentration to improve adhesion of PALF/PE composites. The PALF was treated with different concentrations of NaOH (1% and 5%) and when the PALF was treated with 1% NaOH, the mechanical properties of longitudinally oriented PALF/PE composites at different fibre loading were improved. Meanwhile when using 5% NaOH the tensile strength and tensile modulus were decreased. It was also concluded that using the higher concentration of alkali (5%) for treatment will cause the fibre to lose its characteristics. On the other hand, when using lower concentration of NaOH for treatment, the complete removal of lignin on the fibres is not possible. Fibres become thinner upon alkali treatment. This may be due to dissolution and leaching of fatty acids and some of the lignin component of the fibre. As the result, the surface of the fibre becomes rough, and will promote mechanical anchoring between fibre and matrix. Also Rout et al. [5] found that for treatment of coir fibre, increasing concentration of NaOH from 2% to 5% increased the impact strength of coir/polyester from 521.9 to 634.6 J/m. Meanwhile, the increased alkali concentration up to 10% decreased the impact strength of composites.

Mansour Rokbi et al. [27] studied about the effect of chemical treatments of fibres by alkalization on the flexural properties of polyester matrix composite reinforced with Alfa fibres. Alfa grass (Stipa tenacissima L.) is a tussock grass; it is constituted of stems with a cylindrical shape which have a maximum height of about 1m. Alfa fibres were subjected to alkali treatments with NaOH at 1, 5 and 10% for a period of 0, 24, and 48 h at 28°C. Flexural tests were performed using a three point bending set-up according to the ASTM D790 standard. The span length to the specimen thickness ratio is maintained at 16:1. The samples with dimensions of 12 x 1.5 x 0.8 cm3 were tested using ZWICK Z50 machine (1 mm/min). Results show that mechanical proprieties are strongly changed with fibres treatment. The flexural strength of composites treated with 10% NaOH for 24h showed the best flexural strength properties (57 MPa) which is nearly 60% more than that of untreated composite. The report also clearly shows that the material treated with 1% NaOH for 24h showed the highest improvement in terms of flexural strength; which increased by 62% compared to the untreated material. This may be due to bonding of the fibre with the polyester matrix thereby improving the fibre-matrix interaction [28]. On the other hand, it showed that the flexural modulus of the composite treated with 5% NaOH for 48h was considerably lower than those for the untreated Alfa/polyester composite about 20%. Compared to the untreated composite, the improvement in flexural modulus relative to the materials treated with 1% NaOH for 24h and 5% NaOH for 24h were about 45% and 55% respectively. The result also shows that the flexural modulus of the treated Alfa/polyester during 24h increases with an increase NaOH concentrations. However, the material treated with 5% NaOH for 24h is found to have lower values of flexural strength compared to the composite 1% NaOH for 24h. It is also observed that the flexural strength of the material treated with 5% NaOH for 48h was considerably lower than untreated composite about 22%. This remarkable reduction in the mechanical properties found in this composite may be mainly the result of the longer treatment time of Alfa fibres.

Benyahia et al. [29], studied the effect of alkali treatment of natural fibres on the mechanical behavior of the composite unsaturated Polyester-fibre Alfa. Alfa fibres were soaked in 1, 3, 5, and 7 (wt)% of NaOH solutions at 25°C for 24 h, maintaining a liquor ratio of 15:1. The fibres were washed for several times with water to remove any alkali solution sticking on their surface, neutralized with dilute acetic acid and then washed again with water. Tensile and flexural tests of the composite specimens were carried out at a cross head speed of 1mm.min-1. Flexural test was done by the three-point bending test. The tensile test result shows that the material with 7% Alkali treated Alfa fibres displayed the highest tensile strength (25.11MPa) compared to the untreated Alfa fibres composites (17.48MPa). This may be due to the bonding of the fibre with the polyester matrix thereby improving the fibre-matrix interaction [30]. The results have shown that the tensile strength and tensile modulus increased with the increasing of NaOH concentration. The alkali treatments of Alfa fibres have also a significant effect on flexural properties. The report shows that the material with 7% Alkali treated Alfa fibres showed the highest improvement in terms of flexural strength; which increased by 50% compared to the untreated material.

Julio César et al. [31] have reported that the effect of alkali treated banana fibre reinforced composites results in decreased mechanical properties than untreated fibres. The banana fibres were drawn from the banana plants' pseudo-stems by a defibring machine. These fibres were first washed with 2% detergent-water prior to alkali treatment and dried at 70ºC for 24h to remove external wax. They were then mercerised to remove fibre surface impurities, causing changes in the crystalline cellulose, and preparing the fibre for the effects of chemical treatment. Alkali treated fibre was then washed thoroughly with distilled water to remove excess NaOH from the surface and oven dried at 110ºC. A universal testing machine was used for assessing treated and untreated fibres' mechanical behaviour; 50 mm gauge and constant 4 mm/min crosshead speed were used for all tests. The tensile testing result reported that young's modulus value for untreated fibres was 6.6 to 25.6 GPa, ultimate tensile strength was 222.3 to 780.3 MPa and strain was 1.79% to 3.27%; whereas, young's modulus value for treated fibres was 9.73 to 21.6 Gpa, ultimate tensile strength was 148.1 to 536.2 MPa and strain was 1.38% to 2.57%. Comparing untreated fibres young's modulus, ultimate tensile strength and strain values to those for treated ones showed that all decreased in the ranges investigated here. The decrease in mechanical property after alkali treatment of banana fibre may be due to cellulose delignification and degradation during alkali treatment thereby disrupting bonding and leading to morphological changes, like increased surface roughness. Similar findings have been re-ported by Arifuzzaman et al. [32] in okra fibre. Such increased roughness acts as stress concentrator for decreasing fibre's mechanical behavior.

Hossain et al. [33], studied and reported the effect of chemicals on the mechanical properties of wood saw dust. Wood is a material that is widely used in numerous applications including agriculture, construction, transportation, furniture and so on. In order to know the effect of chemical treatment of wood particles on the composite properties, both the medium and coarse particles of the different woods were chemically treated separately with 10% NaOH solution. The treated and washed wood particle was dried for five hours in an oven at 1100C to remove all the moisture content absorbed. Samples were prepared as per ASTM standard. The final dimensions of the tensile test samples were 114mmx7mmx6mm. The tensile strength of the entire prepared specimen was measured using Instron Universal Testing Machine at a crosshead speed of 2mm/min according to ASTM D3039 standard. The results reported that the highest tensile strength of chemically treated (with % NaOH) medium size wood saw dust particle reinforced composite was increased by 6% when compared to the untreated counterpart.

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Elammaran et al. [34] studied the effect of fibre surface treatment on the mechanical, acoustical and thermal properties of Betelnut fibre polyester composites. Betelnut is the fruit of Areca palm tree (Areca catechu), a species of palm. The fruits of betelnut are in the form of round or oval in shape, with the color of golden yellow to orange. Betelnut fruits were soaked in water at room temperature for 5 days to loosen the fibre from the husk. Then the fibres were separated manually from the nut portion by a hand stripping method and washed thoroughly with distilled water before dried in an oven at 70oC for 24 h. Unsaturated polyester resin with the trade name of "Reversol P9509 was used as a matrix material. Betelnut fibres and unsaturated polyerster composites were prepared in the different ratio of 5:95, 10:90, 15:85, 20:80 (Betelnut fibre wt%:Unsaturated polyester wt%) using the cold press moulding technique. For the tensile test, a mould with a thickness of 5 mm and cross-sectional area of 72.5 mm2 was used. Tensile testing was performed with an LS-28011-50 Universal Testing Machine T-machine Technology Co., LTD, Taiwan using ASTM D638 as the control specimen. The mechanical testing reported that, the tensile strength of chemically treated fibre for a fibre loading of 5% and 10% increases by 50% and 42.86%, respectively, to that of the untreated one. However, when the fibre loading increases more than 10%; i.e, for 15% and 20% the tensile strength of treated fibre decreases by 22.22% and 41.18% respectively when compared to the untreated fibre. Similar investigations have also been reported by T. Yu et al. [35] stated that the reason behind the increasing tensile strength is due to the attribution presence of fibre which creates a dispersed matrix that allowed uniform distribution stress on the materials. Apart from that, the decrease in the tensile strength is due to over presence of fibre in the material that contributed in initiating the crack which can cause non-uniform stress transfer due to the fibre agglomeration within the jute matrix.

3. Conclusion

1. Alkali-treated banana fibres test result revealed that there is a decreased in mechanical properties when compared to the untreated banana fibre. This decrease in mechanical properties may be due to the removal of lignin and hemicellulose from the surface of the fibres, possibly causing weakening in the fibre's outer wall.
2. The study of mechanical properties of betelnut fibre reinforced composites show that composites prepared at 10% fibre loading and 5% NaOH treatment has optimum mechanical strength. This is primarily a result of improved adhesion and enhanced polar interactions at the fibre matrix interfaces.
3. Chemical treatment of wood saw dust substantially increases the strength of wood particle reinforced polymer matrix composites.
4. Chemical treatment of sisal fibre results in enhanced mechanical property.
5. The NaOH treatment of the short pineapple leaf fibre enhanced values of all of its mechanical properties such as tensile strength, tensile modulus, flexural strength, flexural modulus, notched and unnotched impact and hardness. The maximum improvement of the mechanical properties of short pineapple leaf fibre (PALF) reinforced high impact polystyrene (PALF) was obtained from the experiment of treated PALF with 4% NaOH.
6. The alkali treatment of Alfa fibres has also a significant effect on flexural properties. The report shows that the material with 7% Alkali treated Alfa fibres showed the highest improvement in terms of flexural strength; which is increased by 50% compared to the untreated material. Alfa fibre can be obtained at a relatively low cost compared to glass fibre reinforcements, and then these fibres have a very promising future in composite material.

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