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Review of Natural Fiber Composites

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Abstract—*Natural fibers are emerging as low cost, lightweight and environmental friendly alternatives to glass and other synthetic fibers in composite materials. In this paper we try to review various works carried out using different types of natural fibers, different manufacturing techniques used, relative mechanical strengths and various types of ageing studies done on natural fiber reinforced polymer composites. Natural fiber composites are likely to be environmentally superior to synthetic fiber composites in most cases for the following reasons: (1) natural fiber production has lower environmental impacts compared to glass fiber production (2) they have higher fiber content for equivalent performance, reducing more polluting base polymer content; (3) the light-weight natural fiber composites improve fuel efficiency and reduce emissions in the use phase of the component, especially in auto applications; and (4) end of life incineration of natural fibers results in recovered energy and carbon credits.*

1. INTRODUCTION

The use of natural fibres as the reinforcement in the composites is receiving increasing attention now a days. Composites are cohesive structures made by physically combining two or more compatible materials, different in composition and characteristics. Specific requirements of modern materials can be met mostly by reinforcing the natural fibers in manmade or synthetic resins.

The main advantages of natural fibres over man-made fibres are- low cost, low density, comparable specific tensile properties, low abrasiveness, non-irritation to the skin, reduced energy consumption, less health risk, renewability, recyclability and biodegradability. Also another feature of natural cellulose fibres is that they can be more easily modified than the relatively inert carbon, aramid and glass fibres.

However, certain drawbacks of natural fibre-polymer composites are the incompatibility between the hydrophilic natural fibres and the hydrophobic polymeric matrices. This leads to decrement in properties of the composites. It is therefore necessary to modify the fibre surface/structure by employing physical/chemical modifications to improve the adhesion between fibre and matrix.

Here we try to review the various research works carried out using different natural fibers, their processing into composites, mechanical characterization, possible improvement of properties, degradation and ageing related studies.

2. MECHANICAL PROPERTIES

Composites are one of the most advanced and adaptable engineering materials. Composites are defined by researchers [12,13,14,15] in many ways. Most of the studies on natural fiber composites involve study of mechanical properties as a function of fiber content, effect of various treatments of fibers, and the use of external coupling agents.

Both the matrix and fiber properties are important in improving mechanical properties of the composites. The tensile strength is more sensitive to the matrix properties, whereas the modulus is dependent on the fiber properties. To improve the tensile strength, a strong interface, low stress concentration, fiber orientation is required whereas fiber concentration, fiber wetting in the matrix phase, the aspect ratio is very important for determining the fracture properties.

In a study involving coconut fibres as reinforcement in low-density polyethylene,[1] it was observed that waxy layer provided good fibre matrix bond such that removal of the layer resulted in decrease of the fibre pullout stress, increase of the critical fibre length and corresponding decrease in tensile strength and modulus of the composites .

Flax and hemp-fibre reinforced polypropylenes have been tried for material characterisation [2], and concluded that the elastic properties of the composite material are dependent on the type of coupling agent. The experiments carried out on composites made by reinforcing jowar fibre into polyester found that jowar fibre has a tensile strength of 302 MPa, modulus of 6.99 GPa. [3].

Short randomly oriented banana/sisal hybrid fibre reinforced polyester composites were processed by varying the fibre volume fraction. There was good agreement obtained between thermal conductivity measurement values [4]. **The Young's modulus of sisal fibers used in a study was around 18 GPa**. The strain-to-failure decreased from approximately 5.2% to 2.6% when the gage length was increased from 10 mm to 40 mm [5]. The tensile strength, was found to be independent of the gage length.

The cross-sectional shape, the density and tensile properties of vakka, date and bamboo fibers, along with established fibers like sisal, banana, coconut and palm, were determined experimentally under similar conditions and compared. The mechanical properties of the modified coconut fibres were evaluated by tensile and fatigue tests [6]. Results presented a decrease in fatigue life of composites when applied greater tension, due to poor interfacial bonding.

Flax fibre was reinforced with polypropylene and the water absorption tests performed [8] showed that mechanical properties of specimens reduced due to the plasticization effect of water. A detailed study on water absorption characteristics was carried out on oil palm fibres [9]. It was found that the mechanical performance of the treated and untreated fibres decrease upon sorption and regains on desorption[10]. In a study on the effect of microstructure on the mechanical properties of injection moulded bio composites, an increasing trend of fracture toughness with fibre content was found for all sample orientations [11].

Flax fiber reinforced components are extensively used today in the automotive industry, but the fiber acts mainly as filler material in non-structural interior panels [16]. Natural fiber composites used for structural purposes do exist, but then usually with synthetic thermo-set matrices which limit the environmental benefits [17, 18]. In a recent study it was found that composites of poly-L-lactide acid (PLLA) reinforced by flax fibers showed specific tensile modulus equivalent to that of glass/polyester short fiber composites. The specific strength of flax/PLLA composites was lower than that of glass/polyester, but higher than that of flax/polyester. [19]

Wanjun Liu et.al [20] fabricated bio composites from kenaf fiber and soy based bio plastic by extrusion as well as compression molding. Compression molded samples showed higher heat deflection temperature (HDT) and impact strength. Wan Y Z et.al [21] had done research work on bacterial cellulose (BC) nanofibers reinforced biodegradable composites. The BC nanofibers were incorporated in the starch plasticized with glycerol and tensile properties of composite compared with those of the unreinforced starch. It was found that moisture absorption mechanism in the BC starch bio composites follows Fickan diffusion mode. The presence of BC nanofibers improves the tensile properties and the resistance to moisture and microorganism attacks. Murali Mohan Rao et al [22] carried out study on the properties of composites made by reinforcing vakka fiber into a polyester resin matrix. The composites were tested for tensile, flexural and dielectric properties and compared with those of established composites like sisal, bamboo and banana. It was observed that the tensile properties increased with volume fraction of fibers.

In short-fiber-reinforced composites, there exists a critical fiber length that is required to develop its full stressed condition in the polymer matrix. Fiber lengths shorter than this critical length lead to failure due to debonding at the interface at lower load. On the other hand, for fiber lengths greater than the critical length, the fiber is stressed under applied load and thus results in a higher strength of the composite.[62]

3. MANUFACTURING METHODS

The end properties of a composite produced from different materials is not only a function of indivisual properties of resin matrix but also of the way in which the material themselves are designed into the part and also the way in which they are processed. Few of such methods are-

3.1 Spray Lay Up- Fibre is chopped in a hand held gun and fed into a spray of catalyzed resin directed at the mould. The deposited materials are left to cure under standard atmospheric conditions.

3.2 Resin Transfer- Vacuum is applied to the mould cavity to assist resin in being drawn into the fabrics. This is Vacuum assisted resin injection. Once all the fabrics is wet out , the resin inlets are closed and the laminate is allowed to cure. Both injection and cure can take place at either ambient or elevated temperatures.

3.3 Pultrusion- Fibres are pulled out from a creel through a resin bath and then on through a heated die. The die completes the impregnation of the fibre, controls the resin content and cures the material into its final shape . This cured profile is then automatically cut into desired length. Fabrics may also may introduced into the die to provide the fibre direction.

4. CHEMICAL TREATMENTS

Chemical treatment of the natural fibres is carried out to improve its mechanical properties such as flexural strength, modulus and impact strength by varying its surface characteristics. Some of the chemical treatments carried out on fibres are alkylation, acetylation, benzylation etc. The chemical sources for the treatments include alkali, silane, acetylation, benzoylation, acrylation and acrylonitrile grafting, maleated coupling agents, permanganate, peroxide, isocyanate, stearic acid, sodium chlorite, triazine, fatty acid derivate (oleoyl chloride) and fungal. The chemical treatments used in various researchs are summarized below.

4.1 Alkylation

Alkylation is the transfer of an alkyl group from one molecule to another. Alkylating agents are widely used in chemical treatment of fibres because the alkyl group is the most common group encountered in organic molecules. Fibres are treated with Sodium Hydroxide solutions of different strengths in this process.

Prasad S V et al [29] treated the coir fiber with alkali solution to improve the wettability of fibers with polyester. Fibers were soaked in 5% aqueous solution of Sodium hydroxide (NaOH) at 28±1° C for 72 to 76 hours and treatment of fibers increased the tensile strength by 15%. The debonding stress of treated fibers from matrix was 90% higher than that of untreated fibers from the same matrix. Treatment of fibers increased the mechanical strengths such as flexural strength, modulus and impact strength of composites by 40%. Valadez Gonzalez et al [30] treated the henequen fibers with a NaOH aqueous solution (2% w/v) for an hour at 25°C. The treatment of fibers increased the surface roughness that resulted in a better mechanical interlocking and incremented the amount of cellulose exposed on the fiber surface. Morphological and silane chemical modification of the fiber surface improved interfacial shear strength between fibers and thermoplastic matrix. Jochen Gassan et al [31] improved the mechanical properties of jute epoxy composites by the NaOH treatment process. Alkali treatment was done by treating the fiber samples with different solutions of NaOH at concentrations up to 28 wt% for a maximum of 30 min at a temperature of 20°C. The Young's modulus. of composites with treated and untreated fibers were approximately 30% and 50% respectively, lower than comparable glass fiber epoxy composites. Srinivasababu et al [32] exploited new composite material made up of okra fiber reinforced with polyester resin. Extracted fibers treated with 0.125M NaOH for 6 hours and 45 minutes. The fibers treated for 45 minutes reinforced composite material showed the highest tensile strength and modulus of 64.41 MPa and 946.44 MPa respectively. Specific tensile strength and modulus of untreated and treated okra FRP composites is 34.31% and 39.84% higher than pure polyester specimen. Min Zin Rong et al [33] investigated effect of fiber treatment on the mechanical properties of unidirectional sisal reinforced epoxy composites. Sisal fiber was immersed in a solution of 2% NaOH for 4hr at 60°C, in which fiber to solution weight ratio was 1:25. Van de Weyenberg I et al [34] modified flax fiber with NaOH, dilute epoxy, silane and acetone. Fibers were dipped in NaOH solution of different concentration (1, 2 or 3%) for 20 minute at room temperature. A treatment consisting of a combination of alkali and dilute epoxy showed the highest flexural properties. The longitudinal properties of unidirectional composites enhanced with 40%, transverse strength increased with 250%. Varada Rajulu et.al [35] reinforced short natural fiber belonging to the species Hildegardia Populifolia in styrenated polyester matrix. Alkali treatment of the fibers enhanced the tensile modulus by 3.5% and compressive strength by 7.5%. Threepopnatkul [36] studied the properties of pineapple leaf fiber reinforced polycarbonate composites. Surfaces of fiber were pre-treated with sodium hydroxide at 30°C for 5h. Material with 20% fiber content showed high tensile strength and Young's modulus.

4.2 Potassium Treatment

A study was conducted to determine the effects of potassium silicate treatment [41] on newspaper and unbleached kraft fibers. Treated wood fiber was better than the untreated wood fiber as Silane and K silicate treatments had generally higher average normalized toughness values than N-silicate-treated newspaper fiber –cement composites.

The effect of potassium permanganate treatment on the mechanical (tensile), properties of okra (Abelmoschus esculentus) fibres has been studied [42]. The treatment makes the fibre more permeable to resin when used as composite reinforcement. Fracture surfaces present an increased level of twisting as an effect of chemical modification and of reduced adhesion force between the fibrils.

A study was conducted to improve the interfacial bond between Sansevieria cylindrica fibres (SCFs) and polyester matrix [43]. Potassium permanganate treatment was carried out to modify the fibre surface. Potassium-permanganate-treated S. cylindrica fibre/polyester (PSCFP) composites showed optimum mechanical properties among the treated S. cylindrica fibre/polyester (SCFP) composites. The SEM micrographs reveal that interfacial bonding between potassium-permanganate-treated SCF (PSCF) and polyester matrix has significantly improved, suggesting that better dispersion of PSCF into the matrix has occurred upon potassium permanganate treatment of SCF.

Permanganate treatment on natural fibres was conducted by potassium permanganate ($KMnO_4$) in acetone solution[44]. This treatment enhances chemical interlocking at the interface and provides better adhesion with the matrix wall. It reduces the hydrophilic nature of the fibre[45]. Higher concentrations of KMnO4 (more than 1%) cause excess delignification within the cellulosic structure and degrade fibre properties [46,47]. $KMnO_4$ etches the fibre surface and makes it physically rougher to improve mechanical interlocking with the matrix. Flexural strength and modulus properties were increased by 5% and 10% for the treated banana fibre polypropylene composites. Li et al. [48] applied 0.2% potassium permanganate ($KMnO_4$) solution (in 2% acetone) on alkali (2% NaOH for 1 h) pre-treated flax fibre and reported treated fibre-LLDPE and HDPE composites had higher tensile strength properties compared to the untreated fibre composites.

4.3 Acrylation

Acrylic acid (CH2=CHCOOH) is used to enhance interfacial bonding between the fibre and matrix (polypropylene). CH2=CHCOOH reacts with the cellulosic hydroxyl groups of the fibre and provides more access of reactive cellulose macroradicals to the polymerization medium.

In a study, rice straw fiber (RSF) was modified by suspension polymerization of butyl acrylate (BA) monomer [49]. Mechanical properties showed that the tensile strength of poly(lactic acid) (PLA) composites (W(%) = 7.98) increased by 6 MPa compared with blank sample. The water absorption of the PLA composites was lower than PLA/RSF composites. The differential scanning calorimetry (DSC) data showed that RSF played a role as a nucleating agent and PBA(poly butyl acrylate) made crystallization of PLA more difficult and incomplete.

A test was carried out by fiber surface modification by ethylene dimethylacrylate (EMA) and cured under UV radiation [50]. The grafting of alkali treated fiber shows an increase of polymer loading (about 56% higher) and tensile strength (about 27%) than 50% EMA grafted fiber. The best improvement was observed for the 20% alkali treated followed by grafting with 50% EMA.

5. SURFACE TREATMENTS

The primary objective of surface treatments on natural fibres is to maximize the bonding strength so as the stress transferability in the composite Acrylic acid (CH=CHCOOH) is used to enhance interfacial bonding between the fibre and matrix (polypropylene). It reduces hydrophilic hydroxyl groups from the fibre structure and improves moisture resistance properties. This coupling mechanism between the fibre and matrix by acrylic acid enhances the stress transfer capacity at the interface and thus improves composite properties.[51,52]

The overall mechanical properties of natural fibre reinforced polymer composites are highly dependent on the morphology, aspect ratio, hydrophilic tendency and dimensional stability of the fibres used.

In a study, jute fibres were corona discharge and ultraviolet (UV) treated to improve the mechanical properties of natural-fibre/epoxy composites [54]. The UV treatment of the single fibres and yarns led to significantly higher gains in polarity in comparison with those observed in relation to corona-treated materials. Increasing treatment time at a constant bulb-sample distance or alternatively decreasing the distance significantly increased the polarity and decreased yarn tenacity. An increase in the composite flexural strength of about 30% was achieved. Significant improvements in the mechanical properties of the composites are reported by using different chemical treatment processes on the reinforcing fibre.

6. AGEING STUDIES

The effect of three different aging methods (immersion in hot water, freeze–thaw cycles and wet–dry cycles) on the mechanical properties of GRC were studied and compared [55]. A new aging method, mixing freeze–thaw cycles and wet–dry cycles, was the most accurate simulation of weather conditions that produce a noticeable change in GRC mechanical properties.

This study investigated the effect of accelerated weathering on the visual appearance and on mechanical properties of high impact polystyrene (HIPS) as well as HIPS reinforced with mercerized and bleached sugarcane bagasse fibers composites [56]. After accelerated weathering period of 900 h, under UV-B radiation and moisture regular cycles, changes in mechanical properties were investigated by tensile tests. It was observed that composites reinforced with bleached fibers are less susceptible to accelerated weathering exposure than composites reinforced with mercerized fibers, which is explained by the higher amount of lignin present in mercerized fibers.

The effects of ageing on the mechanical properties and dimensional stability of cardanol derivative of toluene diisocyanate (CTDZC) treated and untreated sBal/LDPE composites have been studied [57]. Samples were immersed in boiling water for 7 h under atmospheric pressure; in the second case, samples were heated at 70°C in an air circulating oven for 7 days. It has been demonstrated that CTDIC-treated composites showed superior mechanical properties and dimensional stability as compared to untreated composites due to the existence of an efficient interfacial bond between the fiber and the polymer matrix.

A study on the influence of water ageing on mechanical properties and damage events of flax–fibre composites, compared with glass–fibre composites [58] showed that failure stress decreased with the increasing of immersion time for both glass–fibre and flax–fibre composites. Elastic properties of flax–fibre composites were hardly affected by water ageing, whereas only the tensile stress was affected in the case of glass–fibre composites.

7. CONCLUSIONS

Composite Materials are being used and will play a major role in various fields of engineering applications. Apart from lightness, such materials are also bio degradable if these are prepared using the natural fibers and resins. The goal of degradation can be achieved at least partly, by reinforcing with natural fibers while retaining the synthetic resins for very severe applications. The tensile properties of natural fibre reinforced polymers (both thermoplastics and thermosets) are mainly influenced by the interfacial adhesion between the matrix and the fibres. Usage of natural fibers as raw materials while developing new composite materials which can find applications in various fields would be one prominent solution for protecting the planet Earth. Composite properties can be moderately improved by subjecting them to chemical and surface treatments.

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