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1 author:

Asim Shahzad
Swansea University
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Article

Hemp fiber and its composites – a review

Asim Shahzad

Abstract
The use of hemp fibers as reinforcement in composite materials has increased in recent years as a response to the increasing demand for developing biodegradable, sustainable, and recyclable materials. Hemp fibers are found in the stem of the plant which makes them strong and stiff, a primary requirement for the reinforcement of composite materials. The mechanical properties of hemp fibers are comparable to those of glass fibers. However their biggest disadvantage is the variability in their properties. Composites made of hemp fibers with thermoplastic, thermoset, and biodegradable matrices have exhibited good mechanical properties. A number of hemp fiber surface treatments, used to improve the fiber/matrix interfacial bonding, have resulted in considerable improvements in the composites' mechanical properties.

Introduction

The invention of polymers in early 20th century laid the foundation of a revolution in materials. This revolution had an inherent drawback with respect to the environment because of poor biodegradability of these materials. This meant that disposal of this material put extra burden on already dwindling landfill resources in most countries. The annual global disposal of millions of tons of plastics, especially from packaging, has raised the demands for looking for new means of managing this non-biodegradable waste. These environmental issues have resulted in considerable interest in the development of new composite materials based on biodegradable resources, such as natural fibers as low-cost and environment-friendly alternative for synthetic fibers. Hemp, sisal, and flax are some examples of the natural fibers being used in composite materials. Synthetic polymers have been conventionally used as matrices in composite materials which are not eco-friendly. Now new matrix materials are also being developed based on natural and renewable resources, for the development of 'green' biocomposites. Polyactic acid, soy oil, and lignophenolic resins are some examples of such biodegradable matrix materials.

Hemp

After sisal, hemp is the most widely used natural fiber as reinforcement in composites. Hemp is naturally one of the most ecologically friendly fibers and also the oldest. The Columbia History of the World states that the oldest relics of human industry are bits of hemp fabric discovered in tombs dating back to approximately 8,000 BC. The flowering tops and, to a lesser extent, leaves of hemp produce resin secretions containing the narcotic 9-Δ tetrahydrocannabinol (THC) for which marijuana and hashish are famous. Since industrial hemp produces less than 0.2% THC, it cannot be used as a narcotic.

Hemp is an annual plant native to central Asia and known to have been grown for more than 12,000 years. It probably reached central Europe in the Iron Age and there is evidence of its growth in the UK by the Anglo-Saxons (800-1000 AD). It is now grown mostly in the EU, Central Asia, Philippines, and China. According to Food and Agriculture Organisation (FAO), almost half of the world's industrial hemp supply is grown in China, with most of the remainder being cultivated in Chile, France, the Democratic People's Republic of Korea, and Spain.[1]

In hemp plant, fibers are contained within the tissues of the stems which help to hold the plant erect. In doing so these fibers impart strength and stiffness to

Materials Research Centre, School of Engineering, Swansea University, Swansea, UK.

Corresponding author:
Asim Shahzad, Materials Research Centre, School of Engineering, Swansea University, Swansea SA2 8PP, UK
Email: mr_asim_shahzad@yahoo.com

the tree. This high strength and stiffness of hemp fibers makes them a useful material to be used as reinforcement in composite materials. There has been an exponential increase in the use of hemp for various applications in recent years. According to FAO, world production of hemp fiber grew from 50,000 tons in 2000 to almost 90,000 tons in 2005.[1] Hemp currently accounts for less than 0.5% of total world production of natural fibers.

As Annie Kelly wrote in the Guardian in 2006,[2] only fifteen years ago, any farmer trying to grow hemp in Britain could be arrested. But in 2006, grown under license, more than 3,500 acres of hemp was harvested as an industrial crop, processed, and made into a plethora of natural products, including insulation, horse bedding, fabric, bio diesel, and paper. 'Hemp is back and is throwing off its hippy shackles to emerge as one of the UK's fastest growing sustainable industries', she wrote. The National Non-Food Crops Centre (www.nnfcc.co.uk) recognizes hemp as an established minor crop. The ban on hemp cultivation, imposed in 1971 under the Misuse of Drugs Act, was finally overturned in 1993. Campaigners successfully argued that although industrial hemp was a variety of the cannabis plant, it could be grown as a legitimate crop as it contained practically no THC. Still permission of the Home Office is required to grow hemp. Only one company in Britain has the license to grow hemp. Hemcore, the UK's first large-scale hemp company, has seen rapid growth over the last few years. It now owns the only hemp processing plant in the UK and currently contracts 40 farmers to grow 3,500 acres of hemp a year, which it converts into industrial materials. It currently provides all BMW 5 Series cars with hemp door panels, as well as making high-quality horse bedding.

The main constituents of hemp fibers are cellulose, hemi-cellulose, lignin, and pectin as shown in Table 1.

Cellulose, hemi-cellulose, and lignin are basic components that define physical properties of fibers. Cellulose is the stiffest and the strongest organic constituent in the fibers. Perhaps the biggest drawback of hemp fibers is the variability in their composition as shown in Table 1. This, invariably results in variability in their physical and mechanical properties. Diameters and properties of natural fibers vary significantly depending upon factors such as source, age, retting and separating techniques, geographic origin, rainfall during growth, and constituents' content.[4]

Nishino[5] has identified the following factors that can cause the variability in the physical and mechanical properties of natural fibers:

Materials	Microscopic: crystallinity; microfibril angle; crystal modifications
	Macroscopic: fineness; porosity; size and shape of lumen
	History: source; age; retting and separating conditions; geographical origin; rainfall during growth
Measurement conditions	tensile speed; initial gauge length; moisture; temperature; different cross-section of fibers at different points

Morvan et al.[17] showed that the diameter of flax fiber bundles is greater in the middle part of the stem than in the bottom or top regions. Although the cross section of natural fiber bundles is polygonal in shape, the major dimension is referred to as the diameter in literature and this convention has been followed in this review. The diameter was also found to vary depending on the development stage of the plant, the diameter being lower during flowering/capsulation stage than

Table 1. Chemical composition of hemp fibers as reported by different authors

Cellulose (%)	Hemicellulose (%)	Pectin (%)	Lignin (%)	Other (%)	Reference
67.0	16.1	0.8	3.3	2.8	[6]
74.4	17.9	0.9	3.7	0.8	[7]
74.0	18.0	1.0	4.0		[8]
55.0	16.0	18.0	4.0	7.0	[9]
76.0	11.5	1.3	3.2		[10]
57–77			9–13		[11]
75.1	<2		8.0		[12]
70–74	17.9–22.4	0.9	3.7–5.7	0.8	[13]
75.6	10.7		6.6		[14]
78.3			2.9		[15]
76.1	12.3	1.6	5.7	3.3	[16]

mature capsule or seed maturation stages. It is natural to expect that this variability in diameter will also impart variability to the mechanical properties of the fibers.

Catling and Grayson[18] measured the diameters of hemp fiber bundles and found the average to be 30 µm with a range of 11.68–31.96 µm. The average hemp fiber length was found to be 8.46 mm with a range of 1–34 mm. Olsen and Plackett[19] found the average hemp fiber bundle diameter to be 25 µm and average hemp fiber bundle length was found to be 25 mm with a range of 5–55 mm.

Table 2 shows typical physical and mechanical properties of hemp fiber and Table 3 shows tensile properties of hemp fibers as reported by different authors. Again these are representative values for these

Table 2. Typical physical and mechanical properties of hemp fiber

Properties	Values
Length (ultimate) (mm)	8.3–14
Diameter (ultimate) (µm)	17–23
Aspect ratio (length / diameter)	549
Specific apparent density (gravity)	1500
Microfibril angel (θ)	6.2
Moisture content (%)	12
Cellulose content (%)	90
Tensile strength (MPa)	310–750
Specific tensile strength (MPa)	210–510
Young's modulus (GPa)	30–60
Specific Young's modulus (GPa)	20–41
Failure strain (%)	2–4

Table 3. Tensile properties of hemp fibers as reported by different authors

Tensile strength (MPa)	Tensile modulus (GPa)	Elongation at break (%)	Reference
690		1.6	[4]
1235		4.2	[6]
310–750	30–60	2–4	[21]
550–900	70	1.6	[22]
690		1.6	[23]
895	25		[8]
500–1040	32–70	1.6	[10]
920	70		[24]
690–1000	50	1.0–1.6	[25]
920	70	1.7	[26]
270–900	20–70	1.6	[27]

properties with considerable scope for variation. Apart from their high tensile strength and stiffness, their high aspect ratio (length/diameter ratio) and lower density make hemp fibers a good material to be used as reinforcements in composite materials. Their disadvantages for use in composite materials, like most other natural fibers, are their non-uniform and non-smooth surfaces, variability of properties, and low resistance to water absorption and decay.

Pickering et al.[20] studied the effect of growth period on tensile properties of hemp fibers. During the flowering stage of male plants (99–124 days of growth) the tensile strength of fiber increased gradually. Changes in chemical composition of the fibers as the plant aged were attributed to the increase in tensile properties. They also found that fibers with gauge length of 1.5 mm have higher tensile strength than fibers with gauge length of 10 mm.

Good mechanical properties of hemp fibers notwithstanding, the variability in their properties will continue to pose challenges to researchers to come up with ways of reducing these variabilities.

Evolution of hemp fiber composites

In 1941, hemp fibers (and flax) were used in resin matrix composites for the bodywork of a Henry Ford car which was able to withstand ten-times the impact on an equivalent metal panel. Unfortunately the car did not make into general production due to economic limitations at the time. The commercial success of glass fiber, having superior properties over natural fibers, and concurrent development of synthetic resins such as unsaturated polyester resins and epoxies during and just after the Second World War led to the mass production of synthetic composites and the corresponding decline in use of natural fibers in these composites. Ecological concerns of society in issues such as sustainability, recyclability, and environmental safety in 1990s resulted in renewed interest in natural fiber composites.

Germany has been at the forefront of using natural fibers in composite materials for automobile applications. A recent survey[28] found that the use of natural fibers (excluding wood and cotton) in automotive composites almost doubled from 9,600 tons in 1999 to 19,000 tons in 2005. Flax fibers had the biggest market share at 65%, followed by hemp fibers at 10%. According to authors the market share of hemp fibers can be increased by establishing further processing capacities or by reduction in hemp insulation material market. Similarly the use of natural fiber reinforced composites doubled from 15,000 tons in 1999 to 30,000 tons in 2005. On average the 5.4 million passenger cars produced in 2005 used 3.6 kg of natural fibers per car.

The consumption of hemp fibers in European Union also increased in this time. Another survey[29] found the market share of hemp fiber in automotive industry in EU countries increased from 1% in 1996 to 15% in 2002.

In 2008, the British company Lotus (www.lotuscars. com) unveiled its environment friendly car Elise Eco. The car uses hemp, sisal, and wool fibers in the manufacture of interior trim, roof, seat covers, and hard top.

Window frames and floor coverings made of hemp fiber reinforced polymer composites were used during the 2008 Beijing Olympics.[30]

Sustainable Composites Ltd (www.suscomp.com) is a British company that was established in 2003 to develop a range of eco-friendly manufacturing materials made from sustainable crops such as hemp and castor oil. The company has successfully made surfboard and a dinghy from these materials. They have also developed a linseed oil based resin that contains 96% vegetable oil.

A Dutch company NPSP Composieten BV (www. npsp.nl) is manufacturing diverse products using hemp and flax fibers. Examples of products are mushroom-shaped guideposts for bicycle paths, housings of radar units (glass fibers disturb the radar rays), boats, furniture, and loudspeakers.

The 4th International Conference of the European Industrial Hemp Association (EIHA), held in November, 2006 in Germany, revealed a globally increasing interest in hemp raw materials due to worldwide raw material shortages.[30] It was also pointed out that because of increasing wood prices worldwide, manufacturers have started to use hemp as a replacement material in lightweight chip boards. The conference concluded that the demand of hemp fiber was bound to increase in coming years.

Hemp fiber reinforced thermoplastics

Thermoplastics offer many advantages over thermoset polymers for use with natural fibers like low processing cost, design flexibility, and ease of molding complex parts. Their biggest disadvantage is that their processing temperature is restricted to below 230°C to avoid thermal degradation of natural fibers. Only those thermoplastics are useable for natural fibers whose processing temperature does not exceed 230°C, for example, polyethylene and polypropylene (PP). Other thermoplastics like polyamides, polyesters, and polycarbonates, that require processing temperatures of greater than 250°C, cannot be used with natural fibers.

Natural fibers generally have higher modulus than thermoplastics, thus resulting in higher modulus composites[31]. Natural fiber reinforced thermoplastic composites are flexible, tough, and show good mechanical properties. However the fiber orientation in these composites is random and accordingly property improvement is not as high as in thermoset composites. Thermoplastics have significant viscosity and therefore their good wetting with natural fibers is difficult to achieve. Increasing their temperature to reduce viscosity can damage the natural fibers.

PP is the most extensively used thermoplastic in natural fiber composites owing to its low density, excellent processibility, good mechanical properties, high temperature resistance, good dimensional stability, and good impact strength. Poor fiber/matrix interfacial bonding can be an issue while using natural fibers with PP. Mutje et al.[32] determined polarities of hemp fibers, glass fibers, and PP and observed hydrophobic behavior of glass and PP in contrast to very hydrophilic nature of hemp fibers. However it was found that irregular surface morphology of hemp could still improve the fiber-matrix adhesion. The tensile strength of hemp-PP composites was 50% of that of glass-PP composites and tensile modulus was 80% of glass-PP composites at 40% fiber weight fraction. Using maleated polypropylene (MAPP) as compatibility agent resulted in considerable increase in tensile properties of hemp-PP composites. Mechanical properties of some hemp fiber-PP composites and glass fiber-PP composites reported by various researchers are shown in Table 4.

Yamamoto et al.[33] studied the processing and mechanical properties of natural fiber composites. They used three kinds of kenaf-hemp non-woven materials impregnated with acrylic matrix. The mixing ratio of kenaf/hemp was 50:50. The prepregs were compression molded. The tensile strength of these composites varied from 28 MPa to 75 MPa for molding pressure of 1.5 MPa to 6 MPa. The tensile modulus varied form 3.9 GPa to 11 GPa. Elongation to fracture varied from 0.8% to 1.4%. The fractographic study of fractured samples was carried out which showed good adhesion between fiber and matrix.

Khoathane et al.[34] studied the mechanical and thermal properties of hemp fiber reinforced 1-pentene/PP copolymer composites. At fiber weight fraction of 30%, tensile strength was measured at 25 MPa, tensile modulus at 2.5 GPa, flexural strength at 3.3 GPa, and impact strength at 1.3 kJ/m^2. The thermal stability of the composites was found to be better than that of the fibers or the matrix as individual entities.

Wambua et al.[35] studied the mechanical properties of natural fiber/PP composites, using kenaf, coir, sisal, hemp, and jute fibers, all at 40% fiber weight fraction. Hemp fiber composites showed the highest tensile strength of 52 MPa while coir showed the lowest (10 MPa). The strengths of kenaf, sisal and jute composites were approximately 30 MPa. Similar results for flexural properties of these composite were achieved.

Table 4. Mechanical properties of (untreated) hemp fiber reinforced polypropylene composites

Fiber/fraction (%)	Tensile strength (MPa)	Tensile modulus (GPa)	Flexural strength (MPa)	Flexural Modulus (GPa)	Impact strength (J/m^2)	Reference
None	20–40	1.0–1.4	42.1	1.1	16	[36]
Hemp/30 (wt)	22.1	10.2				[37]
Hemp/40 (wt)	40.2	3.55	73.3	4.1	29	[38]
Hemp/50 (wt)	50	6.5	85	4	53 (kJ/m^2)	[39]
Hemp/30 (wt)	32.3	1.53	48.5	1.67	15.1 (kJ/m^2)	[40]
Hemp/30–70 (wt)	55	6.7	85	4.7	46 (kJ/m^2)	[41]
Hemp/64 (wt)	42		63		152	[42]
Hemp	34	2.7	41.5	2.3	3.89 (kJ/m^2)	[43]
Glass/22 (vol)	88.6	6.2	60	4.4	54.12 (kJ/m^2)	[44]

Hemp composites showed the best flexural strength properties (54 MPa) which is comparable with glass fiber composites with propylene matrix (60 MPa). But, more importantly, the specific flexural strength of hemp composite (36.5) was higher than that of glass fiber composite (24). Similarly tensile and flexural moduli also showed similar trends. Hemp and kenaf composites registered highest tensile modulus of 6.8 GPa, which was comparable to that of glass fiber composites (6.2 GPa). It was lowest for coir composites with 1.3 GPa. Again the specific moduli of hemp and kenaf composite (4.6) were higher than that of glass fiber composite (2.5). The flexural modulus was again highest for hemp composite at 5 GPa and it was lowest for coir composite at 0.5 GPa. Finally Charpy impact strength of these composites was investigated. The impact strength of these composites was found to be quite low compared to glass fiber composites. The maximum impact strength was registered for hemp and sisal composites which was 25 kJ/m^2 whereas it was 54 kJ/m^2 for glass fiber composites. The mechanical properties of hemp fiber composites proved them as promising candidate to replace glass fiber. The specific properties of natural fiber composites were better than those of glass. It was concluded that natural fibers reinforced PP composites have potential to replace glass fiber composites in many applications that do not require very high load bearing capabilities.

However the use of hemp fibers with thermoplastic matrices has been increasing quite slowly, an indication of the issue of poor wetting of the two. More research needs to be done to overcome this issue if the use of hemp fibers with thermoplastic matrices is to match the corresponding use with thermoset matrices.

Hemp fiber reinforced thermosets

Although the use of thermoplastics in natural fiber composites is increasing, natural fiber reinforced thermosets still constitute the majority of the composites made from natural fibers. The most commonly used thermosets are polyester, epoxy, and vinylester. The natural fiber composites made with thermosets are highly solvent resistant, tough, and creep resistant. The fibers can carry as much as 80% of the load in these composites.[45]

The mechanical properties of thermoset biocomposites depend on the mechanical properties of the matrix and the fiber as well as the interfacial bonding between the two. Interfacial bonding between the matrix and the fiber depends on three factors: mechanical anchoring, physical attractive forces (van der Waals force and hydrogen bond), and chemical bonding between the matrix and the fiber. The surface of the natural fiber contains many hydroxyl groups in its chemical structure which make hydrogen bonds with the hydroxyl groups in the main backbone chain of the matrix. The polyester resin, having no hydroxyl group in its backbone chain, has generally the weakest bonding with the natural fiber compared to epoxy and vinylester resins.[46] Considerable shrinkage (up to 8%) during curing of polyester resin also weakens this bonding.

Unsaturated polyester resin is still by far the most widely used thermoset for natural fiber composites. Table 5 shows the summary of mechanical properties of various hemp fiber composites with unsaturated polyester resin reported by various researchers. For comparison the properties of glass fiber composites have also been given.

Hautala et al.[47] fabricated plywood-type composites from hemp fiber strips and epoxy resin. The flexural strength was found to be comparable to that of traditional plywood. The appearance, manufacturing properties, and workability was also found to be suitable for floor and furniture applications. By using 48 layers of hemp or flax fibers, composites of even greater strength were manufactured. The flexural strength of the composites at 50-60% fiber weight faction was determined at 140 MPa and flexural modulus at 6 GPa.

Table 5. Mechanical properties of (untreated) hemp fiber reinforced unsaturated polyester composites

Fiber	Fiber wt/vol fraction (%)	Tensile Strength (MPa)	Tensile Modulus (GPa)	Flexural Strength (MPa)	Flexural Modulus (GPa)	Impact Strength (kJ/m²)	Reference
Hemp	35 (vol)	60.2	1.74	112.9	6.40	14.2	[50]
Hemp	36 (vol)	–	–	110.0	7.50	13.0	[45]
Hemp	30 (vol)	38.0	6.0	100.0	6.50	20 (J/m²)	[53]
Hemp	20 (vol)	33.0	1.42	54.0	5.00	4.8	[54]
Glass (CSM)	7 (vol)	–	–	108.0	5.60	34	[45]
Glass (CSM)	20 (vol)	73.4	7.9	233.8	9.28	80.4	[55]

Aziz and Ansell[48] studied the mechanical properties of hemp and kenaf-fiber reinforced polyester composites, untreated and with alkali treatment. The alkali-treated fibers of both types of composites showed superior flexural strength and flexural modulus values compared to untreated fibers. The improvement in properties was observed for short, long, and random mat fibers. They also studied the effect of using specially formulated polyester resins for natural fibers. These resins are more polar in nature, making them hydrophilic so that they can bond better with the OH-groups on the surface of natural fibers. It was observed that these polyesters made a positive impact to the strength of the composites. The flexural stiffness of these composites was found to be close to that of glass fiber composites. A considerable improvement was also observed in flexural strength of these composites.

Richardson and Zhang[49] studied the effects of non-woven hemp on mechanical properties of phenolics and their micro structural features. They found a significant increase in flexural strength and modulus in phenolics with the introduction of non-woven hemp. Impact toughness was substantially improved. This improvement was attributed to two factors. First that hemp is tougher and stronger mechanically than phenolic matrix and leads to increase in mechanical properties. The second is that presence of hemp has the capability of reducing void (defect) numbers and dimensions, which significantly contributes to improvement in mechanical properties.

Rouison et al.[50] also studied the optimization of Resin Transfer Molding (RTM) process in manufacturing hemp fiber composites and their mechanical properties. The tensile strength of hemp fiber/polyester composites increased linearly with increasing fiber content above 11% fiber volume fraction. A maximum tensile strength of 60 MPa was achieved for fiber volume fraction of 35%. The results for tensile modulus also showed similar trend. Again a maximum elastic modulus of 1.7 GPa was achieved for fiber volume fraction of 35%. The flexural strength and flexural modulus also increased linearly with increasing fiber volume fraction. The highest values attained for flexural strength and flexural modulus were 113 MPa and 6.4 GPa respectively. The impact strength of these composites also increased with increasing fiber content. The highest impact strength of 14.2 kJ/m² was achieved again for fiber volume fraction of 35%. Finally flexural creep properties of 21% fiber content were investigated. It was found that these materials are not suitable for use under high fatigue load conditions.

Sebe et al.[51] studied the flexural and impact properties of hemp fiber-reinforced polyester composites made by Resin Transfer Molding technique. Flexural stress at break and flexural modulus showed an increasing trend with increasing fiber content. Impact strength was found to decrease at low fiber content, and then gradually increase with further addition of fibers.

Hemp fiber reinforced thermosets can rival glass fiber composites in terms of their mechanical properties. Their main weakness is their poorer impact strength compared to glass fiber composites. Most of the research done so far has concentrated on studying the tensile, flexural, and impact properties of hemp fiber composites. The study of fatigue properties of hemp fiber composites is still in preliminary stages[52] and more work needs to be done to understand the fatigue properties of these composites.

Hemp fiber reinforced biodegradable polymers

The natural fibers reinforced with biodegradable polymers result in completely 'green' composites. The advantages of using such composites with respect to environmental pollution are obvious and the research in such composites has increased substantially in recent years. Examples of biodegradable polymers are thermoplastic starch, polyhydroalkanoates (PHA), polylactides (PLA), lignin based epoxy, soy based resins, and epoxidised linseed and soyabean oil.

Although produced from natural sources, these polymers are not necessarily biodegradable because they depend mainly on the chemical structure (degree of cross linking) of the polymers. One of the major drawbacks of these polymers is their cost. Most

biodegradable resins currently cost three to five times the commonly used resins such as PP, LDPE, HDPE, and PVC.[4] Some examples of studies done on these composites are shown in Table 6. These studies have shown promising properties of these composites, although in most cases these properties are lower than those of natural fiber reinforced thermosets and thermoplastics.

Hemp fiber hybrid composites

One method of increasing the mechanical properties of natural fiber composites is by hybridizing them with another synthetic or natural fiber of superior mechanical properties. The synthetic fiber mostly used for this purpose is glass fiber.[56,57] Although the biodegrability of the composite is reduced, this can offset the advantages gained by the increase in mechanical properties.

Hemp fiber surface modification

A strong degree of adhesion is needed for effective transfer of stress from matrix to fibers in composite materials. This requires surface modification of fiber surfaces. Strongly polarized cellulose fibers are not inherently compatible with the hydrophobic non-polar polymers matrices. Also the poor resistance to moisture absorption makes natural fibers less attractive for making composites to be used in outdoor applications. Additionally for several technically oriented applications, the fibers have to be specially prepared or modified regarding homogenization of the fibers' properties, degrees of elementarization and degumming, degrees of polymerization and crystallization, moisture repellence, and flame retardant properties.[67]

There are two methods of improving the fiber-matrix interface: physical methods and chemical methods.

Physical methods do not change chemical composition of fibers, but improve mechanical bonding to polymers. Commonly used methods are stretching, clantering, thermotreatment, production of hybrid yarns, electric discharge, corona treatment, cold plasma treatment, and alkalization.

Chemical modification is defined as a chemical reaction between some reactive constituents of the natural fibers and chemical reagent, with or without catalyst, to form a covalent bond between the two. In chemical methods, coupling agents are used to bring about compatibility between strongly polarized cellulose fibers that are inherently incompatible with hydrophobic polymers.[68] These coupling agents react with the hydroxyl groups of cellulose and the functional groups of the matrix. General methods used are change of surface tension, impregnation of fibers, and chemical coupling. The surface modifications due to coupling agents cause noticeable improvements of the characteristic values of composites. The most commonly used chemical methods are liquid ammonia, esterification, silane coupling method, isocynates, permengnates, and graft copolymerization. A number of studies have been carried out on the effects of different fiber surface treatments on the properties of hemp fiber composites.

Sebe et al.[51] modified the surface of hemp fiber composites, with polyester as the matrix, via esterification of hemp hydroxyl groups using mathacrylic anhydride. Increased bonding between fiber and matrix due to this treatment did not vary the flexural stress at break but was detrimental to toughness. This behavior was ascribed to a change in the mode of failure, from fiber pull-out to fiber fracture, resulting in a marked reduction in the energy involved in the failure of the composite, leading to a more brittle material.

Rouison et al.[54] studied the effect of various surface modifications on the mechanical properties of hemp fiber/polyester composites. Chemicals used for paper sizing (AKD, ASA, Rosin Acid, and SMA) as well as silane compound and sodium hydroxide were used to modify the fibers' surface. The tensile, flexural, and impact properties were investigated. All the chemicals, except alkali solution, improved substantially the hydrophobic behavior of fibers. The paper sizing chemicals decreased the flexural and tensile properties by creating a weaker fiber-matrix interface. But the silane, SMA, and alkali treatments improved these properties slightly. These improvements were attributed to better interface interaction due to enhanced hydrophobic behavior of fibers and, for alkali treatment, to better the strength properties of fibers. The lack of strong adhesion between fiber and matrix was confirmed by the slight increase in impact strength of these samples as well as the analysis of their tensile fracture surface. A slight improvement in mechanical

Table 6. Hemp fiber reinforced biodegradable polymer composites

Fiber	Matrix	Reference
Hemp	Cashew nut shell liquid	[58]
Flax, hemp and wool	Expoxidised soy bean oil	[59]
Flax, hemp, ramie	Starch, lactic acid	[60]
Manila hemp	Starch	[61]
Hemp, flax	Soy oil	[62]
Hemp	Cellulose ester	[63]
Flax, cellulose, pulp, hemp	Acrylated epoxized soyabean oil	[64]
Hemp	Polylactic acid	[65]
Hemp	Euphorbia oil	[66]

properties was observed for SMA, silane, and alkali treated specimens. However close examination of these tests and of the fracture surface of the samples showed no improvement in fiber-matrix adhesion.

Tserki et al.[69] studied the effect of acetylation and propionylation treatments on flax, hemp, and wood fibers. The highest effect of esterification was achieved for woof fibers due to their high lignin/hemicellulose content, followed by flax and hemp fibers. Crucially, both the acetylation and propionylation treatments reduced water adsorption in fibers, rendering them more hydrophobic. The SEM examination showed the surfaces of the esterified materials to be smoother than untreated material.

Oujai et al.[70] studied the effect of solvent extraction, alkalization, and acrylonitrile (AN) grafting of hemp fibers on surface properties and mechanical properties of hemp fibers. Spectroscopic studies and diffraction techniques showed slight decrease of crystallinity index. The structural transformation of fibers from cellulose 1 to cellulose 2 was observed at high NaOH concentration of 10–20% wt/vol. The AN grafted fibers had no transformation of crystalline structure as observed after mercerization. Only a variation of X-ray crystallinity index with grafting amount was observed. Moisture regain of pre-treated and modified fibers depended on the structure of the fiber and amount of grafting.

Mwaikambu and Bisanda[71] studied alkalization and acetylation techniques for hemp, sisal, jute, and kapok fibers. Both treatments successfully modified the structure of natural fibers as well as increase in the crystillinity index of the fibers. A high crystallinity index resulted in stiff/strong fibers and hemp fared the best in this respect. By observing the first exothermic peak (DSC), hemp had the highest stability after acetylation. This implied that acetylation did not result in degradation of the crystalline cellulose. Fourier Transform Infrared (FTIR) spectroscopy showed hemp to be not much reactive. But SEM results indicated that after chemical treatment, all the fibers except kapok possessed rough surfaces which increased mechanical interlocking with resins. It was concluded that hemp had the highest crystallinity index and thermal stability when alkalized and acetylated with and without acid catalyst, followed by jute, sisal, and kapok fibers.

Mwaikambo and Ansell[72] undertook a thorough analysis of physical and fine structure of hemp fiber bundles, namely surface topography, diameter, cellulose content, and crystillinity index. The fiber bundles were alkalized and their physical and mechanical properties analyzed. Alkalization was found to change the surface topography of fiber bundles and the diameter decreased with increased concentration of caustic soda. Cellulose content increased slightly at lower NaOH concentrations and decreased at higher NaOH concentrations. The crystallinity index decreased with increase in caustic soda concentration up to 0.24% NaOH beyond which, it decreased with increase in NaOH concentration.

It was also found that the tensile strength and stiffness increased with increase in the concentration of NaOH up to a limit. Tensile strength and Young's modulus increased with decrease in cellulose content, while crystalline cellulose decreased slightly but with improved crystalline packing order resulting in increased mechanical properties. Similar observations were elucidated by the crystallinity index. Alkalized hemp fiber bundles were found to exhibit a similar specific stiffness to steel, E-glass, and Kevlar 29 fibers. The results also showed that crystallinity index obtained following alkalization had a reverse correlation to the mechanical properties. Stiffer alkalized hemp fiber bundles were suitable candidates as reinforcements to replace synthetic fibers. The improvement in mechanical properties of alkali treated hemp fiber bundles confirmed their use as reinforcement.

Kostic et al.[16] studied the effect of alkalization of varying concentrations at different temperatures on hemp fibers. The treated hemp fibers were finer, with lower lignin content, increased flexibility, and in some cases tensile properties were improved.

FTIR analysis of 8% wt/vol NaOH treated hemp fibers by Ouajai and Shanks[73] indicated the removal of pectin and hemicellulose by this treatment. The IR lateral crystallinity index exhibited slight decrease following this treatment indicating the presence of less ordered cellulose structure. X-ray diffraction studies of untreated hemp fibers showed the characteristics of cellulose 1. Following NaOH treatment of up to 20% wt/vol, the crystalline transformation to cellulose 2 was observed which resulted in decrease of crystallinity.

It is generally accepted that interfacial adhesion can be best described in terms of dispersion forces and acid–base interactions. Therefore, there is a need for quantitative determination of acid–base character of natural cellulosic fibers. In their study, Gulati and Sain[74] determined acid base characteristics and dispersion component of surface energy of hemp fibers using inverse gas chromatography. Effect of alkalization and acetylation on acid–base characteristics was also examined. The results indicated that alkalization and acetylation made the hemp fiber amphoteric, thereby improving their potential to interact with both acidic and basic resins. A parallel was drawn between the changes in fiber-matrix acid–base interactions and the actual improvement in the mechanical properties of the composites manufactured using resin transfer molding process.

Wang et al.[14] exposed hemp fibers of nano-scale (30–100 nm width) to 12% NaOH solution. The

cellulose content was found to increase significantly from 76% for untreated fibers to 94% for treated fibers. There was corresponding decrease in hemicelluloses of 10.7% for untreated fibers to 1.9% for treated fibers. X-ray crystallography of fibers showed that the crystallinity of fibers increased after the treatment, thus affirming the increase in cellulose content.

The FTIR analysis of 5% alkalized hemp, kenaf, flax ad henequen fibers by Sgriccia et al.[75] showed complete removal of hemicellulose and partial removal of lignin following the treatment. Pietak et al.[76] observed that hemp fibers treated to 18% alkali solution for only 30 min resulted in removal of non-cellulosic components and more cellulose rich surface.

Beckermann and Pickering[77] used 10% NaOH and 5% NaOH/2% Na_2SO_3 solutions for treatment of hemp fibers. NaOH treatment was more effective in removal of lignin and increased the crytallinity index of fibers following the treatment. The treated fibers were also more thermally stable than untreated fibers. Both the treatments resulted in increase in tensile properties of hemp-PP composites.

Mwaikambo and Ansell[58] also observed increase in tensile properties of hemp fiber reinforced cashew nut shell liquid (CNSL) composites following alkalization of fibers. The increase was observed for both unidirectional and non-woven fiber composites.

Bledzki et al.[78] used 22% NaOH solution on hemp fibers and studied the properties of unidirectional hemp yarn-epoxy composites. The flexural strength was increased by 45% and flexural modulus was increased by 100% following the treatment. The use of maleic-anhydride-polypropylene copolymer (MAH-PP) coupling agent was also studied for unidirectional hemp-PP and flax-PP composites. The transverse tensile strength of hemp-PP composites increased by 75% but there was no increase in longitudinal strength following the treatment. Flax-PP composites showed 150% increase in flexural strength and 65% increase in transverse tensile strength following the treatment.

Mehta et al.[53] studied the effect of various fiber surface treatments on the mechanical properties of hemp/polyester composites. The surface treatments used were: 5% alkalization, silane, and acrylonitrile. All treatments resulted in increase in mechanical properties of the composites. The only exception was impact strength which was found to decrease following alkalization treatment, and there was only marginal increase in impact strength for other treatments. This was consistent with the fact that improved fiber/surface adhesion will reduce the impact strength of the composites.

Thygesen et al.[79] studied the effects of steam explosion, wet oxidation, and enzymatic defibration on chemical composition, crystallinity, and cellulose

chain length of hemp fibers. The degree of crystallization of cellulose decreased slightly by enzymatic defibration but not by other treatments. The cellulose chain length decreased significantly only by steam explosion. The cellulose content was highest in the fiber bundles after wet oxidation.

Mehta et al.[53] exposed hemp fibers to 5% NaOH solution for 1 h. They reported increase in thermal stability of hemp fibers after treatment. After the treatment, the carbon content within the fibers was found to increase while oxygen and nitrogen contents were found to decrease.

This review shows that considerable improvements can be gained in the mechanical properties of hemp fiber composites by using the suitable surface treatment. The choice of treatment will depend on cost, the range of improvement required, and the applications of the composites following the treatment.

Environmental properties of hemp fiber composites

Hemp fibers are hydrophilic and absorb moisture. The moisture content of fibers can vary between 5%–10% and sometimes even exceed this value. This can lead to dimensional variations in composites and also affects mechanical properties of composites. It can also lead to poor processibility and porous products. It can cause fibers to swell and ultimately rot through attack by fungi. A possible solution is to improve fiber-matrix interface by using compatibilizers and adhesion promoters. With better adhesion, the moisture sensitivity is usually reduced[9]. Also surface treatments of fibers with silanes can make the fibers more hydrophobic.

Ho and Ngo[80] studied the water sorption characteristics of hemp and coir fibers. Both fibers displayed two-stage water sorption curves: the first stage obeyed Fick's law, whereas the second stage represented non-Fickian diffusion. Hemp fiber displayed saturation water uptake of 66% at 23°C which increased to 80% at 80°C. At 23°C the following sorption parameters for hemp fibers were determined: Diffusion coefficient 0.56×10^{-9} m^2/s, sorption coefficient 0.66, and permeability coefficient 3.7×10^{-10} m^2/s.

Jimenez and Bismarck[12] exposed different natural fiber to 100% relative humidity and determined the equilibrium moisture uptake. Cornhusk, hemp, and sisal fibers had uptake values of 33% whereas abaca, flax, luffa, henequen, lechuguilla, and lyocell fibers had equilibrium moisture uptake of 23–30%.

In their studies on water absorption behavior of hemp fiber reinforced PP composites, Hargitai et al.[39] found the saturation water uptake to be strongly dependent on fiber weight fraction. Whereas the saturation water uptake for composites with 30% fiber weight

fraction was only 7%, the value for composites with fiber weight fraction of 70% was as high as 53%.

Sgriccia et al.[75] reported that hemp fiber reinforced epoxy composites had a saturation weight gain of 18.4% following immersion in water for 1,500 h. This is a considerably high value considering that the composites had a fiber weight fraction of 15%. At the same fiber weight fraction, henequen-epoxy composites had the highest weight gain of 28% while flax-epoxy and kenaf-epoxy composites had similar weight gain as hemp-epoxy composites.

In another study, Aghedo and Baillie[81] reported a saturation mass gain of 10% for hemp fiber reinforced recycled linear low-density polyethylene composites following 2,000 h of immersion in water.

Mehta et al.[82] reported saturation moisture uptake of 0.7% for hemp fiber reinforced polyester composites placed in humidity chamber having 90% RH. For acrylonitrile treated hemp fiber composites, the equilibrium moisture uptake was 0.3%.

Mishra et al.[83] reported water uptake of 19.5% after 30 h of immersion for hemp fiber reinforced novolac composites having fiber weigh fraction of 40%. For fibers treated with maleic anhydride, the composites' water uptake was reduced to 14.5% for the same time of immersion.

Another study[10] showed that composites reinforced with 30% hemp fibers in PP matrix immersed in water absorbed approximately 7% of their weight. On the other hand, under standard hygroscopicity and temperature conditions, after saturation the part released all the water adsorbed, and no hysteresis was observed in the sorption/desorption curves. The mechanical properties of the composites decreased slightly following the absorption of water, although the original properties were recovered as soon as the water was desorbed.

Various coupling agents have been used to reduce the moisture absorption of natural fibers. The improved moisture resistance of natural fiber composites because of coupling agents can be explained by an improved fiber-matrix adhesion. The coupling agent builds chemical bonds (silanol bonds) and hydrogen bonds which reduce the moisture absorption caused by fiber-matrix debonding.

Mishra et al.[83] studied the swelling properties of natural and maleic anhydride esterified fibers of banana, hemp, and sisal reinforced in novolac resin. Amongst all the fibers tested, the maximum absorption of water was found in hemp fiber composite and the minimum in surface treated sisal fiber composite. The surface treatment reduced the water absorption in all the composites. Steam absorption was also found to be maximum for hemp fiber composites. The surface treatment of fibers again reduced the steam absorption in all composites.

Rouison et al.[84] studied the water absorption of hemp/unsaturated polyester composites. The water absorption increased with increasing fiber content. The moisture absorption process was shown to follow diffusion mechanism and was more prominent in longitudinal than transverse direction. Composite samples immersed in water reached saturation levels after about 8 months and showed no signs of cracking due to swelling. Various fiber treatments were tested (sizing, alkali, silane), but none resulted in substantial increase in resistance to water absorption. The most effective technique to increase moisture resistance was to properly enclose all the fibers within the matrix. The samples were immersed in water for 1 month and then tested for flexural properties. The flexural strength was found to decrease by 11% while flexural modulus was found to decrease by 34%.

The moisture absorption of hemp fiber composites is a clear disadvantage compared to glass fiber composites. This precludes the use of these composites in outdoor applications. The surface treatment of fibers seems to offer a solution.

Hemp fiber composites in comparison to glass fiber composites

The performance of natural fiber composites will inevitably be judged against the performance of glass fiber composites because the primary reason of using natural fibers is their potential to replace glass fibers. Various studies have been undertaken in this regard.

Table 7 shows the values of different environmental parameters in the production of 1 kg of hemp fibers and 1 kg of glass fibers. The lower power consumption and lower emission of greenhouse gases gives natural fibers a clear edge compared to glass fiber. However to compare the environmental impact of composites made from hemp and glass fibers, life cycle analysis of both composites is a more reliable scientific tool. Various researchers have undertaken studies to compare life cycle assessment of hemp and glass fiber composites.

Wotzel et al.[85] undertook life cycle assessment (LCA) of a side panel of Audi A3 car made form

Table 7. Environmental parameters in production of 1 kg of fibers [10]

Parameters	Hemp fiber	Glass fiber
Power consumption (MJ)	3.4	48.3
CO_2 emission (kg)	0.64	20.4
SO_x emission (g)	1.2	8.8
NO_x emission (g)	0.95	2.9
BOD (mg)	0.265	1.75
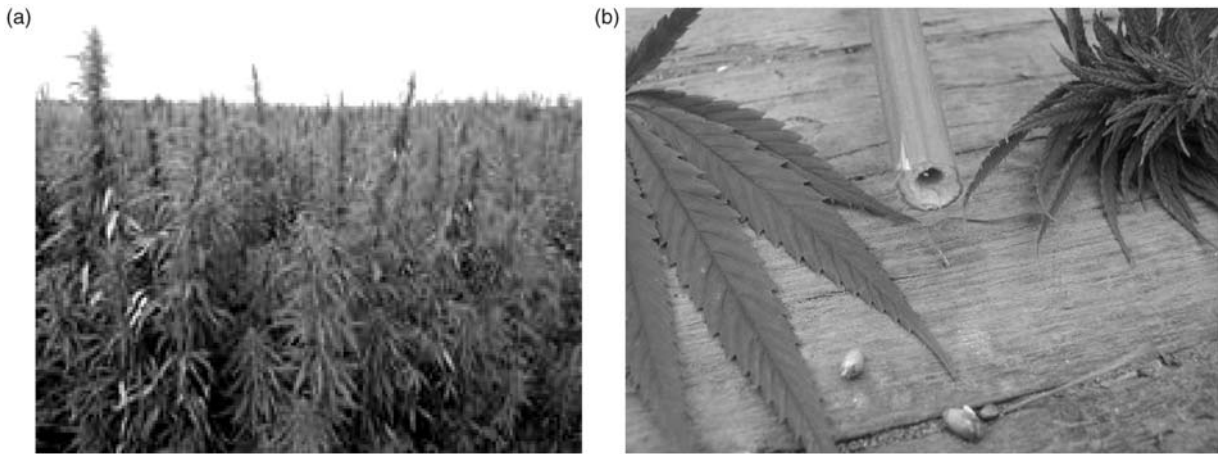
Figure 1. Hemp crop (a) and hemp leaves and stem (b).[3]

ABS co-polymer and an alternate design made from hemp fiber (66 volume %)-epoxy resin composite. The study modelled inputs, energy use, and emissions up to the component manufacturing stage. For the hemp fiber composite components, cultivation of hemp, hemp fiber extraction, and component manufacture stages were modelled. Their study concluded that hemp fiber components used 45% less energy and produced lower air emissions. However water emissions of nitrates and phosphates and nitrogen oxide emissions to air were higher due to fertiliser use in hemp cultivation. Overall Eco-indicator point score were about 8% less for hemp components. The environmental impacts for hemp-epoxy composite were dominated by the energy and emissions from epoxy production. Although hemp fiber accounted for 66% of the component volume, it contributed to only 5.3% of the cumulative energy demand.

Schmidt and Beyer[86] conducted simplified LCAs of two designs of an insulation component of a Ford car. The reference component was made from ethylene propylene diene copolymer (EPDM), PP, and reinforced with glass fibers. The alternative component was also made from EPDM and PP but reinforced with 30 wt % hemp fibers. They considered the main inputs and emissions for glass fiber production, EPDM production, PP production, hemp fiber production, component molding, use phase on the car, collection, and disposal. They presented their final results in the form of net benefits of switching to the hemp fiber component from glass fiber component. The hemp fiber components showed net benefits of 88.9 MJ in cumulative energy demand, 8.18 kg of CO_2 emissions, 0.056 kg of sulphur dioxide emissions, 0.002 kg of phosphate emissions, and 0.018 kg of nitrate emissions.

Murphy[87] studied the environmental impacts of a composite garage door system manufactured containing a door skin based either on glass fiber or non-woven hemp fiber mat using LCA. The results showed that there was little difference between the two door types in terms of impact categories, like ozone depletion potential, ecotoxicity potential, global warming potential, acidification potential, human toxicity potential, etc. Hemp mat door performed slightly better than the glass fiber equivalent in only one category (solid waste).

Pervaiz and Sain[42] studied the environmental performance of hemp-PP composites by quantifying carbon storage potential and CO_2 emissions and comparing the results with commercially available glass fiber composites. A value of 325 kg carbon per metric ton of hemp based composite is estimated that can be stored by the product during its useful life. A net saving of 50,000 MJ (~3 tons CO_2 emissions) per ton of thermoplastic can be achieved by replacing 30% glass fiber reinforcement with 65% hemp fiber. Further 3.07 million tons CO_2 emissions and 1.19 million m^3 crude oil consumption can be saved by substituting 50% glass fiber composites with natural fiber composites in North American automotive applications.

Various studies done on recycling and reprocessing of natural fiber composites, especially with thermoplastic matrices, have revealed the benefits of these materials over synthetic fiber composites. Bourmaud and Baley[88] studied the effect of multiple reprocessing on mechanical, thermal, and rheological properties of hemp-PP and sisal-PP composites. Seven cycles of reprocessing did not show significant effect on tensile properties of the composites. Comparatively the glass-PP composites showed considerable degradation in tensile properties following the same number of reprocessing cycles. The fiber length and the composite viscosity were found to decrease following reprocessing.

The LCA studies have revealed potentially significant benefits of hemp fiber composites compared to glass fiber composites, especially in automotive and

transport applications. These benefits are often in the order of 15% to 50% savings of environmental impact. However the LCA results for natural fiber composites can be very sensitive to the specific applications and it is difficult to draw broad and generic conclusions.

Conclusions

Hemp fibers have properties that make them a suitable material to replace glass fibers as reinforcements in composite materials. Their main disadvantage is the variability in their properties. Extensive research has been carried out on hemp fiber composites using thermoplastic, thermoset, and biodegradable polymer matrices. These composites have shown promising mechanical properties which, in some cases, surpass even those of glass fiber composites. Various fiber surface treatments have been shown to improve the hemp fiber/matrix interfacial bonding, resulting in improved mechanical properties. Another disadvantage of hemp fiber composites, their moisture absorption, can also be overcome by using suitable fiber surface treatment.

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