

*Review***NATURAL FIBER-REINFORCED POLYMER COMPOSITES****Saira Taj¹, Munawar Ali Munawar², and Shafiullah Khan³**^{1,3}*Applied Chemistry Research Centre, PCSIR Labs Complex, Lahore-54600, Pakistan, and* ²*Institute of Chemistry, University of the Punjab, Lahore, Pakistan*

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Summary: Natural fibers have been used to reinforce materials for over 3,000 years. More recently they have been employed in combination with plastics. Many types of natural fibers have been investigated for use in plastics including Flax, hemp, jute, straw, wood fiber, rice husks, wheat, barley, oats, rye, cane (sugar and bamboo), grass reeds, kenaf, ramie, oil palm empty fruit bunch, sisal, coir, water hyacinth, pennywort, kapok, paper-mulberry, raphia, banana fiber, pineapple leaf fiber and papyrus. Natural fibers have the advantage that they are renewable resources and have marketing appeal. The Asian markets have been using natural fibers for many years e.g., jute is a common reinforcement in India. Natural fibers are increasingly used in automotive and packaging materials. Pakistan is an agricultural country and it is the main stay of Pakistan's economy. Thousands of tons of different crops are produced but most of their wastes do not have any useful utilization. Agricultural wastes include wheat husk, rice husk, and their straw, hemp fiber and shells of various dry fruits. These agricultural wastes can be used to prepare fiber reinforced polymer composites for commercial use. This report examines the different types of fibers available and the current status of research. Many references to the latest work on properties, processing and application have been cited in this review.

Introduction

Composites are materials that comprise strong load carrying material (known as reinforcement) imbedded in weaker material (known as matrix). Reinforcement provides strength and rigidity, helping to support structural load. The matrix, or binder (organic or inorganic) maintains the position and orientation of the reinforcement. Significantly, constituents of the composites retain their individual, physical and chemical properties; yet together they produce a combination of qualities which individual constituents would be incapable of producing alone [1].

Wood [2] is natural three-dimensional polymeric composite and consists primarily of cellulose, hemicellulose and lignin. In addition, wood is an original and natural composite. The

biological world offers other examples of composites in bone and teeth, which are essentially composed of hard inorganic crystals in a matrix of tough organic collagen.

Historical examples of composites [3] are abundant in literature. Significant examples include the use of reinforcing mud walls in houses with bamboo shoots, glued laminated wood by Egyptians (1500 BC) and laminated metals in the forging of swords (1800 AD). In the 20th century, modern composites were used in 1930s, where glass fibers reinforced resins. Boats and aircrafts were built out of these glass composites, commonly called fiberglass. Since the 1970s, the application of composites has widely increased due to development of new fibers such as carbon, boron and aramids, and new composite systems with matrices made of metal and ceramics.

Types of Composites

For the sake of simplicity, however, composites can be grouped [4] into categories based on the nature of the matrix each type possesses. Methods of fabrication also vary according to physical and chemical properties of the matrices and reinforcing fibers.

Polymer Matrix Composites (PMCs)

The most common advanced composites are polymer matrix composites. These composites consist of a polymer thermoplastic or thermosetting reinforced by fiber (natural carbon or boron). These materials can be fashioned into a variety of shapes and sizes. They provide great strength and stiffness along with resistance to corrosion. The reason for these being most common is their low cost, high strength and simple manufacturing principles.

Metal Matrix Composites (MMCs)

Metal matrix composites, as the name implies, have a metal matrix. Examples of matrices in such composites include aluminum, magnesium and titanium. The typical fiber includes carbon and silicon carbide. Metals are mainly reinforced to suit the needs of design. For example, the elastic stiffness and strength of metals can be increased, while large coefficient of thermal expansion, and thermal and electrical conductivities of metals can be reduced by the addition of fibers such as silicon carbide.

Ceramic Matrix Composites (CMCs)

Ceramic matrix composites have ceramic matrix such as alumina, calcium, aluminosilicate reinforced by silicon carbide. The advantages of CMC include high strength, hardness, high service temperature limits for ceramics, chemical inertness and low density.

Naturally resistant to high temperature, ceramic materials have a tendency to become brittle and to fracture. Composites successfully made with ceramic matrices are reinforced with silicon carbide fibers. These composites offer the same high temperature tolerance of super alloys but without such a high density. The brittle nature of ceramics makes composite fabrication difficult. Usually most CMC production procedures involve starting materials in powder form.

There are four classes of ceramics matrices: glass (easy to fabricate because of low softening temperatures, include borosilicate and aluminosilicates), conventional ceramics (silicon carbide, silicon nitride, aluminum oxide and zirconium oxide are fully crystalline), cement and concretion carbon components.

Carbon-carbon composites (CCMs)

CCMs use carbon fibers in a carbon matrix. Carbon-carbon composites are used in very high temperature environments of up to 6000 °F, and are twenty times stronger and thirty times lighter than graphite fibers.

Constituents of composites

1. **Matrices**
2. **Reinforcing Fibers**

Matrices

The role of matrix in a fiber-reinforced composite is to transfer stress between the fibers, to provide a barrier against an adverse environment and to protect the surface of the fibers from mechanical abrasion. The matrix plays a major role in the tensile load carrying capacity of a composite structure. The binding agent or matrix in the composite is of critical importance. Four major types of matrices have

been reported: Polymeric, Metallic, Ceramic and Carbon. Most of the composites used in the industry today are based on polymer matrices. Polymer resins have been divided broadly into two categories: Thermosetting and Thermoplastics.

Thermosetting

Thermoset [5] is a hard and stiff cross-linked material that does not soften or become moldable when heated. Thermosets are stiff and do not stretch the way that elastomers and thermoplastics do. Several types of polymers have been used as matrices for natural fiber composites. Most commonly used thermoset polymers are epoxy resins and other resins (Unsaturated polyester resins (as in fiberglass) Vinyl Ester, Phenolic Epoxy, Novolac and Polyamide) [1-3].

Unsaturated polyesters [6] are extremely versatile in properties and applications and have been a popular thermoset used as the polymer matrix in composites. They are widely produced industrially as they possess many advantages compared to other thermosetting resins including room temperature cure capability, good mechanical properties and transparency. The reinforcement of polyesters with cellulosic fibers has been widely reported. Polyester-jute [7,8], Polyester-sisal [9], polyester-coir [10] polyester-banana-cotton [11], polyester-straw [12], polyester-pineapple leaf [13], and polyester-cotton-kapok [14], are some of the promising systems.

Thermoplastics

Thermoplastics [5] are polymers that require heat to make them processable. After cooling, such materials retain their shape. In addition, these polymers may be reheated and reformed, often without significant changes in their properties. The thermoplastics which have

been used as matrix for natural fiber reinforced composites are as follows:

High density polyethene (HDPE)	[15, 16]
Low density polyethene (LDPE)	[17, 18]
Chlorinated polyethylene (CPE)	[19]
Polypropylene (PP)	[16, 20, 21]
Normal polystyrene (PS)	[22, 23]
Poly (Vinyl chloride) PVC)	[24, 25]
Mixtures of polymers	[26]
Recycled Thermoplastics	[27, 28]

Only those thermoplastics are useable for natural fiber reinforced composites, whose processing temperature (temperature at which fiber is incorporated into polymer matrix) does not exceed 230°C. These are, most of all, polyolefines, like polyethylene and polypropylene. Technical thermoplastics, like polyamides, polyesters and polycarbonates require processing temperatures > 250°C and are therefore not useable for such composite processing without fiber degradation.

Reinforcing fibers

The three most common types of reinforcing fibers include fiberglass, carbon and Aramid.

Carbon fibers

Carbon fibers [29] are used for reinforcing certain matrix materials to form composites. Carbon fibers are unidirectional reinforcements and can be arranged in such a way in the composite that it is stronger in the direction, which must bear loads. The physical properties of carbon fiber reinforced composite materials depend considerably on the nature of the matrix, the fiber alignment, the volume fraction of the fiber and matrix, and on the molding conditions. Several types of matrix materials such as glass and ceramics, metal and plastics have been used as matrices for reinforcement by carbon fiber.

Carbon fiber composites, particularly those with polymer matrices, have become the dominant advanced composite materials for aerospace, automobile, sporting goods and other applications due to their high strength, high modulus, low density, and reasonable cost for application requiring high temperature resistance as in the case of spacecrafts.

Glass fibers

Glass fibers are the most common of all reinforcing fibers for polymeric (plastic) matrix composites (PMCs). The principal advantages of glass fiber are low cost, high tensile strength, high chemical resistance and excellent insulating properties. The two types of glass fibers commonly used in the fiber reinforced plastics industries are E-glass and S-glass. Another type known as C-glass is used in chemical applications requiring greater corrosion resistance to acids than is provided by E-glass.

Kevlar fibers

Kevlar belongs to a group of highly crystalline aramid (aromatic amide) fibers that have the lowest specific gravity and the highest tensile strength to weight ratio among the current reinforcing fibers. They are being used as reinforcement in many marine and aerospace applications.

Boron fiber

The most prominent feature of boron fiber is their extremely high tensile modulus. Boron fibers offer excellent resistance to buckling, which in turn contributes to high compressive strength for boron fiber reinforced composites.

Natural Fibers

The use of natural fiber for the reinforcement of the composites has received increasing attention both by the academic sector and the in-

dustry. Natural fibers have many significant advantages over synthetic fibers. Currently, many types of natural fibers [30] have been investigated for use in plastics including flax, hemp, jute straw, wood, rice husk, wheat, barley, oats, rye, cane (sugar and bamboo), grass, reeds, kenaf, ramie, oil palm empty fruit bunch, sisal, coir, water hyacinth, pennywort, kapok, paper mulberry, raphia, banana fiber, pineapple leaf fiber and papyrus. Thermoplastics reinforced with special wood [2] fillers are enjoying rapid growth due to their many advantages; lightweight reasonable strength and stiffness. Some plant proteins are interesting renewable materials, because of their thermoplastic properties. Wheat gluten [31] is unique among cereal and other plant proteins in its ability to form a cohesive blend with viscoelastic properties once plasticized. For these reasons, wheat gluten has been utilized to process edible or biodegradable films or packing materials. Hemp [32] is a bast lingocellulosic fiber, comes from the plant *Cannabis sativa* and has been used as reinforcement in biodegradable composites.

Composites based on biologically degradable polyester amide and plant fiber (flax and cottons) [33] with good mechanical properties, such as sufficient water resistance and biodegradability, have also been investigated.

Kenaf, *Hibiscus cannabinus* L, a member of hibiscus family is also a biodegradable and environmentally friendly crop. It has been found to be an important source of fiber for composites and other industrial applications. Aziz *et al.* [6] studied the mechanical properties of composites manufactured from polyester resin with Kenaf fiber that blows to a height of at least 10 meter. Traditionally, hemp has been used to make ropes but these days its fiber is used to make items such as clothing, toys and shoes. The fiber is fully biodegradable, is non-toxic and may be recycled.

Flax fibers [34] are potentially outstanding reinforcing fillers in thermoplastic biocomposites. These biocomposites could have a great potential in lowering the usage of petroleum-based plastics. Automotive, building and appliance industries are increasing the utilization of flax fibers day by day due to cost saving, non-abrasiveness and the green movement. Biocomposites containing thermoplastics and modified flax fiber have mechanical properties comparable with those of glass fiber-based thermoplastic (LLDPE/HDPE) composites. Bos *et al.* [35] have investigated the mechanical properties of flax/polypropylene composites, manufactured both with batch kneading and an extrusion process, and compared with the properties of natural fiber mat thermoplastic composites.

Yang *et al.* [36-37] have studied the effect of compatibilizing agents on the mechanical properties and morphology of thermoplastic polymer composites filled with rice husk flour. As the filler loading increases, the composites made without any compatibilizing agent show decreased tensile strength and more brittleness, but greatly improved mechanical properties by incorporation of the compatibilizing agent. The poor interfacial binding between the filler and the polymer matrix causes the composites to have decreased tensile strength, but the tensile strength and modulus improve with the addition of compatibilizing agent. Wheat straw has been used for making composites, panel boards and anion exchangers where the straw is used in powder form rather than in the fibrous form. A limited number of studies have reported the use of wheat straw fibers for production of composites [38-39]. Panthapulakkal *et al.* [40] has processed and characterized wheat straw fibers to evaluate their potential as reinforcing material for thermoplastic composites. Jute is also one of the most common agro fibers used as a reinforcing component for thermoplastics and thermosetting matrices [41,42]. Bamboo is an abundant natural source in Asia and South

America and has been used to develop bamboo reinforced thermosetting plastic (epoxy and polyester) [43,44]. Thwe *et al.* [45] have investigated the effect of environmental aging on the mechanical properties of bamboo-glass fiber reinforced polymer matrix hybrid composite. Okubo *et al.* [46] have fabricated bamboo fiber eco-composites for ecological purposes with the conventional hot press method. They studied their static strength and internal state after their fabrication, and concluded that high weight content of bamboo fiber enabled the bamboo composites to increase their strength in the most effective way, when the bamboo fiber was modified into the cotton shape.

Natural fibers and countries of origin

Natural fibers [4] are generally lignocellulosic in nature, consisting of helically wound cellulose microfibrils in a matrix of lignin and hemicellulose. According to a Food and Agricultural Organization survey, Tanzania and Brazil produce the largest amount of sisal. Hennequen is grown in Mexico. Abaca and hemp are grown in the Philippines. The largest producers of jute are India, China, and Bangladesh. Presently, the annual production of natural fibers in India is about 6 million tons as compared to worldwide production of about 25 million tons.

Table 1. Fibers and countries of origin.

Flax	: Borneo
Hemp	: Yugoslavia, China
Sun Hemp	: Nigeria, Guyana, Sierra Leone, India
Ramie	: Honduras, Mauritius
Jute	: India, Egypt, Guyana, Jamaica, Ghana, Malawi, Sudan, Tanzania
Kenaf	: Iraq, Tanzania, Jamaica, South Africa, Cuba, Togo
Roselle	: Borneo, Guyana, Malaysia, Sri Lanka, Togo, Indonesia, Tanzania
Sisal	: East Africa, Bahamas, Antigua, Kenya, Tanzania, India
Abaca	: Malaysia, Uganda, Philippines, Bolivia
Coir	: India, Sri Lanka, Philippines, Malaysia

Table 2. Annual production of natural fibers and sources.

Fiber Source	World Production 10³ Tons	Origin	Fiber Source	World Production 10³ tons	Origin
Abaca	70	Leaf	Nettles	Abundant	Stem
Bamboo	10,000	Stem	Oil Palm Fruit	Abundant	Fruit
Banana	200	Stem	Palm rah	Abundant	Stem
Broom	Abundant	Stem	Ramie	100	Stem
Coir	100	Fruit	Roselle	250	Stem
Cotton Lint	18,500	Stem	Rice Husk	Abundant	Fruit/grain
Elephant Grass	Abundant	Stem	Rice Straw	Abundant	Stem
Flax	810	Stem	Sisal	380	Stem
Hemp	215	Stem	Sun Hemp	70	Stem
Jute	2,500	Stem	Wheat Straw	Abundant	Stem
Kenaf	770	Stem	Wood	1,750,000	Stem
Linseed	Abundant	Fruit			

Chemical composition of natural fiber

The chemical composition of natural fibers varies depending upon the type of fibers. The chemical composition (Table 3) as well as the structure of the plant fibers is fairly complicated [4]. Plant fibers are a composite material designed by nature. The fibers are basically a rigid, crystalline cellulose microfibril-reinforced amorphous lignin and/or with hemicellulosic matrix. Most plant fibers, except for cotton, are composed of cellulose, hemicellulose, lignin, waxes, and some water-soluble compounds, where cellulose, hemicelluloses, and lignin are the major constituents [30]. The properties of

the constituents contribute to the overall properties of the fiber. Hemicellulose is responsible for the biodegradation, microabsorption and thermal degradation of the fiber as it shows least resistance, whereas lignin is thermally stable but prone to UV degradation. The percentage composition of each of these components varies for different fibers. Generally, the fiber contains 60-80 % cellulose, 5-20 % lignin and up to 20 % moisture. The cell wall of the fibers undergoes pyrolysis with increasing processing temperature and contributes to char formation. These charred layers help to insulate the ligno-cellulose from further thermal degradation.

Mechanical properties of natural fibers

The mechanical properties and physical properties of natural fibers vary considerably depending on the chemical and structural composition, fiber type and growth conditions. Mechanical properties [4] of plant fibers are much

lower when compared to those of the most widely used competing reinforcing glass fibers (Table 4). However, because of their low density, the specific properties (property-to-density ratio), strength, and stiffness of plant fibers are comparable to the values of glass fibers [47].

Table 3. Chemical composition, moisture content, and microfibrillar angle of vegetable fibers.

Fiber	Cellulose (Wt %)	Hemicelluloses (Wt %)	Lignin (Wt %)	Pectin (Wt %)	Moisture Content (Wt %)	Waxes	Microfibrillar Angle (Deg)
Flax	71	18.6-20.6	2.2	2.3	8-12	1.7	5-10
Hemp	70-74	17.9-22.4	3.7-5.7	0.9	6.2-12	0.8	2.6.2
Jute	611-71.5	13.6-20.4	12-13	0.2	12.5-13.7	0.5	8
Kenaf	45-57	21.5	8-13	3-5			
Ramie	68.6-76.2	13.1-16.7	0.6-0.7	1.9	7.5-17	0.3	7.5
Nettle	86				11-17		
Sisal	66-78	10-14	10-14	10	10-22	2	10-22
Henequen	77.6	4-8	13.1				
PALF	70-82		5-12.7		11.8		14
Banana	63-64	10	5		10-12		
Abaca	56-63		12-13	1	5-10		
Oil palm EFB	65		19				42
Oil palm Mesocarp	60		11				46
Cotton	85-90	5.7		0-1	7.85-8.5	0.6	--
Coir	32-43	0.15-0.25	40-45	3-4	8		30-49
Cereal Straw	38-45	15-31	12-20	8			

Table 4. Mechanical properties of natural fibers as compared to conventional reinforcing fibers

Fiber	Density (g/cm ³)	Elongation (%)	Tensile Strength (MPa)	Young's Modulus (Gpa)
Cotton	1.5 – 1.6	7.0 – 8.0	287 – 597	5.5 – 12.6
Jute	1.3	1.5 – 1.8	393 – 773	26.5
Flax	1.5	2.7 – 3.2	345 – 1035	27.6
Hemp	---	1.6	690	---
Ramie	---	3.6 – 3.8	400 – 938	61.4 – 128
Sisal	1.5	2.0 – 2.5	511 – 635	9.4 – 22.0
Coir	1.2	30.0	175	4.0 – 6.0
Viscose (cord)	---	11.4	593	11.0
Soft Wood Kraft	1.5	---	1000	40.0
E-glass	2.5	2.5	2000 – 3500	70.0
S-glass	2.5	2.8	4570	86.0
Aramid (normal)	1.4	3.3 – 3.7	3000 – 3150	63.0 – 67.0
Carbon (standard)	1.4	1.4 – 1.8	4000	230 – 240

Advantages of natural fiber

Natural fibers, as reinforcement, have recently attracted the attention of researchers because of their advantages over other established materials. They are environmentally friendly, fully biodegradable, abundantly available, renewable and cheap and have low density. Plant fibers are light compared to glass, carbon and aramid fibers. The biodegradability of plant fibers can contribute to a healthy ecosystem while their low cost and high performance fulfils the economic interest of industry.

When natural fiber-reinforced plastics are subjected, at the end of their life cycle, to combustion process or landfill, the released amount of CO₂ of the fibers is neutral with respect to the assimilated amount during their growth [47]. The abrasive nature of fiber is much lower which leads to advantages in regard to technical process and recycling process of the composite materi-

als in general. Natural fiber-reinforced plastics, by using biodegradable polymers as matrices, are the most environmental friendly materials, which can be composed at the end of their life cycle. Natural fiber composites are used in place of glass mostly in non-structural applications. A number of automotive components previously made with glass fiber composites are now being manufactured using environmentally friendly composites [48]. Although natural fibers and their composites are environmental friendly and renewable (unlike traditional sources of energy, i.e., coal, oil and gas), these have several bottlenecks. These have: poor wettability, incompatibility with some polymeric matrices and high moisture absorption [49]. Composite materials made with the use of unmodified plant fibers frequently exhibit unsatisfactory mechanical properties. To overcome this, in many cases, a surface treatment or compatibilizing agents need to be used prior to composite fabrication. The properties can be improved both by physi-

cal treatments (cold plasma treatment, corona treatment) and chemical treatments (maleic anhydride organosilanes, isocyanates, sodium hydroxide permanganate and peroxide) [50]. Mechanical properties [51] of natural fibers are much lower than those of glass fibers but their specific properties, especially stiffness, are comparable to the glass fibers.

Chemical modification of natural fibers

One of the major problems associated with the use of natural fibers in composites is their high moisture sensitivity leading to severe reduction of mechanical properties and delamination. The reduction in mechanical properties may be due to poor interfacial bonding between resin matrices and fibers. It is therefore necessary to modify the fiber surface to render it more hydrophobic and also more compatible with resin matrices. An effective method of chemical modification of natural fibers is graft copolymerisation. The resulting co-polymer displays the characteristic properties of both fibrous cellulose and grafted polymer [52,53]. One of the most explored chemical modifications is the acetylation-esterification of cellulose-OH, by reaction with acetic anhydride. This reaction reduces hydrophilicity and swelling of lignocellulosics and their composites [54].

The effect of chemical treatment of natural fibres with sodium alginate and sodium hydroxide has also been reported for coir, banana and sisal fibres by Mani *et al.* [55]. This modification results in an increase in adhesive bonding and thus improves ultimate tensile strength up to 30%. Mitra *et al.* [56] have reported that treatment of jute with polycondensates such as phenol-formaldehyde, melamine-formaldehyde and cashew nut shell with liquid-formaldehyde improves the wettability of jute fibres and reduces water regain properties. Samal and Ray [57] have studied the chemical modification of pineapple leaf fibers using alkali treatment,

diazo coupling with aniline and cross-linking with formaldehyde. These chemical treatments result in significant improvements in mechanical properties, chemical resistance and reduced moisture regain. Finally, Joseph *et al.* [58] have investigated the influence of chemical treatment with sodium hydroxide, isocyanate and peroxide on the properties of sisal/polyethylene composites. The observed enhancement in properties of the composites and attributed this to the strong bonding between sisal and polyethylene matrix.

In an effort to improve the mechanical properties of recycled HDPE/wood fiber composites, Selke *et al.* [59] investigated the use of several additives with possible effect on the fibre/matrix adhesion or fibre dispersion into the matrix. They found that maleic anhydride-modified polypropylene appears especially promising, since its use at a concentration of 5% in composites with 30% wood fibre results in an increase in tensile strength and elongation at break. Similar results have been obtained by Dalvag *et al.* [60], who have reported that the composite's elastic modulus remains unchanged. Zadorecki and Flodin [61] have found that some coupling agents, namely trichloro-s-triazine and di-methylol melamine can produce covalent bonds between cellulosic materials and polymer matrices, leading to modified performance and reduced sensitivity to water. This approach has been further explored by Maldas and Kokta [62], who used phthalic anhydride as coupling agent for wood fiber/polystyrene composites. In addition to the chemical affinity of the benzene rings of phthalic anhydride with those of polystyrene, the anhydride group can directly attack the -OH group of cellulose. Furthermore, Razi *et al.* [63] found that the treatment of wood with sodium hydroxide followed by drying with vinyltrimethoxysilane is superior, for obtaining maximum bonding strength at the wood/polymer interface that yields improved mechanical properties.

Physical surface treatment methods

The use of different kinds of physical surface treatment methods (i.e. corona discharge, cold plasma) leads to changes in the surface structure of the fibres, as well as to changes in the surface energy. The treatment of rayon fibers with oxygen plasma results in increasing the total and polar part of the free surface energy with increasing treatment time, because of the increased O/C ratio. Similar results were observed by Belgacem *et al.* [64] for an increasing corona current level with cellulose fibres. Also, Sakata *et al.* [65] found improved wettability of wood with increasing level of corona treatment. In the case of wood, surface activation increases the amount of aldehyde groups according to the report of Wang *et al.* [66].

Surface modification through coupling agents

Many other coupling agents have also been investigated, namely anhydrides, maleated polymer [67,68], isocyanates [69], and alkoxy-silanes [70-72]. Among these different reagents, maleated polypropylene (MaPP) or polyethylene (MaPE) gives significant enhancement in tensile and flexural strength, ranging from 40 up to 80%, when they are blended with cellulose fibres before mixing with matrix [67,68].

Silane coupling

Silane chemical coupling presents three main advantages: (i) they are commercially available in large scale, (ii) at one end, they bear alkoxy-silane groups capable of reacting with OH-rich surface, and (iii) at the second end, they have a large number of functional groups which can be tailored as a function of the matrix to be used. The last feature ensures, at least, a good compatibility between the reinforcing element and the polymer matrix or even covalent bonds between them. The reaction of silane coupling agents with ligno-cellulose fi-

bers (mainly: cellulose and lignin) has been found to be quite different in comparison with that observed between them and glass surface, in the sense that with cellulose macromolecules, only prehydrolyzed silanes undergo the reaction with cellulose surface [73]. Besides the chemical bonding theory, other theories such as the interpenetrating networks theory have also been proposed. This theory states that the matrix diffuses inside the silane interphase to form an entangled network [74-76].

A number of attempts have been carried out to understand the silane-cellulose system [77-79]. Thus, the interaction of silane coupling agents with cellulosic fibers and the effect of some parameters, such as pH, the initial amount of silane with respect to cellulose and the adsorption contact time, on their anchoring capability onto the fiber surface have been ascertained. Different spectroscopic techniques have been used to show the presence of silane and to quantify its amounts on the substrate and to elucidate the structure of the anchored siloxane network on the fiber's surface [78]. Then, using epoxy and unsaturated polyester resins Abdelmouleh *et al.* [79] have shown that the fiber treatment with silane coupling agents, bearing functional group able to react with the matrix, enhances significantly the mechanical strength of the final composite [28]. Recently, Abdelmouleh *et al.* [80] have studied three silane agents (different by the functionality of the radical moiety appended to silicon atoms) to treat delignified cellulose fibers in order to improve their adhesion low density polyethylene and nitrile rubber matrix. The effect of these treatments on the mechanical properties of the ensuing composites and on their water uptake ability has been investigated accordingly. Poor adhesion [47] at the interface means that the full compatibilities of the composite cannot be exploited and which leaves it vulnerable to environmental attacks that may weaken it, thus reducing its life

span. Insufficient adhesion between hydrophobic polymers and hydrophilic fibers results in poor mechanical properties of natural fiber reinforced polymer composites. Gassan *et al.* [81] have improved the tensile, flexural strength and stiffness of jute-epoxy composites by treating the fibers with silane. Tripathy *et al.* [82] have found that delignification by bleaching produces better interfacial bond between the jute fiber and the polyester matrix, and hence results in better mechanical properties of the composites. The absorption of steam by sisal, hemp and banana fiber/Novolac resin composites has been found to reduce after esterification of the $-OH$ groups with the maleic anhydride [83]. The tensile strength of maleic anhydride treated fiber composites is higher than that of the untreated fiber composites. Luo *et al.* [38] found an increase in the mechanical properties of green composites prepared from the pineapple leaf fibers and polyhydroxy butyrate-co-valerate resin (a biodegradable polymer) [84] with the fiber in longitudinal direction. However, a negative effect of the fibers on properties in transverse direction has been observed. Gauthier *et al.* [85] reported that adhesion may be improved by using coupling agents like maleic anhydride to incorporate hydroxyl groups on the matrix through hydrophilization and consequently enhancing the wetting effect of the resin on the fibers. Mwaikambo and Ansell [86] treated hemp, jute, sisal and kapok fibers with various concentrations of NaOH and found 6% to be the optimized concentration in terms of cleaning the fiber bundle surfaces, yet retaining a high index of crystallinity.

Sharifah *et al.* [87] studied the effect of the chemically modified polyester resin on the mechanical properties of the natural fiber composites and its compatibility with natural fibers. Natural fiber properties are highly variable and depend on conditions of growth. It is therefore very difficult to get the same mechanical properties after repeated testing.

Special types of composites based on natural fibers

In general, the mechanical and physical properties of natural fiber reinforced plastics only conditionally reach the characteristic values of glass-fiber reinforced systems. By using hybrid composites, made of natural fibers and carbon fibers or natural fibers and glass fibers, the properties of natural fiber reinforced composites can be improved further for compression strength [30].

Natural fiber composites have been evaluated with regard to their anti-ballistic characteristics. Flax, hemp and jute reinforced polypropylene composites, with or without mild steel backing or facing, have been studied under ballistic impact test conditions [87]. Many researchers have investigated the response of composite materials to ballistic impact. Recently, D'Almeida *et al.* [88] investigated ballistic impact damage of glass fiber reinforced epoxy composites, while Hasur *et al.* [90] reported on the response of carbon/epoxy composites under high velocity impact. Lee *et al.* [90] studied ballistic impact on armour grade spectra and aramid reinforced composites, whereas Chou *et al.* [91] worked on damage of S2 glass reinforced plastic structural armour. Hine *et al.* [92] studied the energy absorption of woven nylon and aramid composites and UHMWPE (ultra high molecular weight polyethylene). Cantwel and Villanueva [93] investigated the failure of fiber-metal laminate (FML) reinforced aluminium foam sandwich structures at high velocity impact.

Research on ballistic impact has been focused only on the high performance fibers, metal and ceramics and now attempts have been made to study the behavior of natural fiber composite under ballistic impact. Wambua *et al.* [87] bridged the gap and investigated the response of flax, hemp, and jute fabric reinforced

polypropylene composites to ballistic impact by fragment simulating projectiles.

Technical applications of natural fiber reinforced composites

Natural fibers are replacing synthetic fibers as reinforcement in various matrices. The composites so prepared can effectively be used as substitute for wood and also in various other technical fields, e.g. automotive parts.

Seventy years ago, nearly all resources for the production of commodities and many technical products were materials derived from natural textiles. Textiles, ropes, canvas and also paper, were made of local natural fibers, such as flax and hemp. Some of these are still used today. As early as 1908, the first composite materials were applied for the fabrication of large quantities of sheets, tubes and pipes for electronic purposes (paper or cotton to reinforce sheets, made of phenol or melamine-formaldehyde resins). For example in 1996, aeroplane seats and fuel tanks were made of natural fibers with small content of polymeric binders. The last decade has seen a multiplicity of applications of natural fiber composites due to their impressive properties such as biodegradability and high specific properties. Currently, a revolution in the use of natural fibers, as reinforcements in technical application, is taking place mainly in the automobile and packaging industries (e.g., egg boxes). In the automotive industry, textile waste has been used for years to reinforce plastics used in cars, especially in the Trabant.

The use of natural fibers within composite applications is being pursued extensively throughout the world. Consequently, natural fiber composite materials are being used for making many components in the automotive sector. These materials are based largely on polypropylene or polyester matrices, incorporating fibers such as flax, hemp, and jute. Thus in the

future cars may be moulded from cashew nut oil and hemp. Even golf clubs may be built around jute fibers, and tennis racket may be stiffened with coconut hair. Bicycle frames may derive their strength from any one of the 2000 other suitable plants. The high-tech revolution in use of natural fibers could end in replacement of synthetic materials.

The diverse range of products now being produced, utilizing natural fibers and biobased resins derived from soybeans, is giving life to a new generation of biobased composites for a number of applications. These include not only automotive vehicles (including trucking) but also hurricane-resistant housing and structures, especially in the United States [94]. The construction sector and the leisure industry are some of the other areas where these novel materials are finding a market. In Germany, car manufactures are aiming to make every component of their vehicles either recyclable or biodegradable [95].

Future Outlook

In order to be environment friendly, automotive engineers have now developed a concept car, the Eco Car. It is expected to be the sustainable vehicle for the future, running on biofuels. It uses natural fiber composite panels

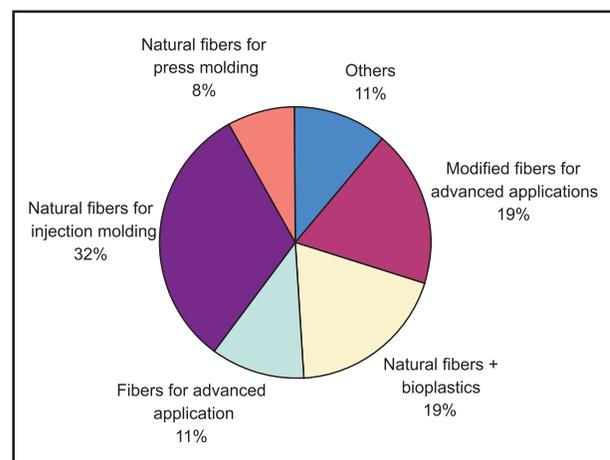


Figure 1. Future Outlook (after [97]).

where biodegradable resins have been incorporated as the matrix material [96]

It has been recently predicted that the most important technologies of the future that incorporate natural fiber composite materials will be natural fibers for injection moulded products (32%) followed by natural fibers with a bioplastic matrix (19%) and modified fibers for use in advanced applications (19%)(Fig.1 [97]).

Concluding remarks

Natural fibers, when used as reinforcement, compete with such technical fibers as glass fiber. The advantages of technical fibers are good mechanical properties; which vary only little, while their disadvantage is difficulty in recycling. Several natural fiber composites reach the mechanical properties of glass fiber composites, and they are already applied, e.g., in automobile and furniture industries. Till date, the most important natural fibers are Jute, flax and coir. Natural Fibers are renewable raw materials and they are recyclable.

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