11
Natural Fibers

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11.1
Introduction

The term “natural fibers” covers a broad range of vegetable, animal, and mineral fibers. However, in the composites industry, it usually refers to wood fiber and plant-based bast, leaf, seed, and stem fibers. These fibers often contribute greatly to the structural performance of the plant and, when used in plastic composites, can provide significant reinforcement. Below is a brief introduction to some of the natural fibers used in plastics. More detailed information can be found elsewhere [1-4].

Although natural fibers have been used in composites for many years, interest in these fibers has waned with the development of synthetic fibers such as glass and carbon fibers. However, recently there has been a resurgence of interest, largely because of ecological considerations, legislative directives, and technological advances. One of the largest areas of recent growth in natural fiber plastic composites is the automotive industry, particularly in Europe, where the low density of the natural fibers and increasing environmental pressures are giving natural fibers an advantage. Most of the composites currently made with natural fibers are press-molded although a wide range of processes have been investigated [1, 5].

Flax is the most used natural fiber (excluding wood) in the European automotive industry, most of which is obtained as a by-product of the textile industry [5]. However, other natural fibers such as jute, kenaf, sisal, coir, hemp, and abaca are also used. Natural fibers are typically combined with polypropylene, polyester, or polyurethane to produce such components as door and trunk liners, parcel shelves, seat backs, interior sunroof shields, and headrests [6].

European consumption of natural fibers in automotive composites was estimated at 26000 tons in 2003 and is expected to grow by 10% per year [7]. Worldwide consumption in all applications by 2010 has been estimated at 110 000–120000 tons per annum in a variety of applications including automotive, building, appliances and business equipment, and consumer products.

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11.2
Structure and Production Methods

The major steps in producing natural fibers for use in plastics include harvesting of the fiber-bearing plants, extraction of the fibers, and further processing of the raw fiber to meet required purity and performance aspects for use in plastic composites.

Methods exist for harvesting most natural fibers since they are used in manufacturing of products other than composites. For example, fibers derived from wood are used in the paper and forest products industries, flax fiber is used to make linen and cigarette papers, and jute fiber is used in making rope and burlap [4]. Since many natural fibers are an annual crop, issues such as storage and variability in the growing season need to be considered. Europe is making large investments in new harvesting and fiber separation technologies for natural fibers such as flax [8].

Fiber extraction procedures will depend on the type and portion of plant the fibers are derived from (e.g., bast, leaves, wood) as well as the required fiber performance and economics. Fiber-bearing plants have very different anatomies (e.g., tree versus dicotyledonous plants) and often fibers are derived from agricultural residues or by-products from industry [8]. Consequently, the processing needs can differ greatly.

Wood is primarily composed of hollow, elongated, spindle-shaped cells (called tracheids or fibers) that are arranged parallel to each other along the trunk of the tree [9]. These fibers are firmly cemented together and form the structural component of wood tissue. Fibers are extracted from wood by mechanical or chemical means during the pulping process.

Bast fibers such as flax or kenaf have considerably different structure than wood and, consequently, are processed quite differently. They exist in the inner bark of the stems of dicotyledons, which are typically less than 30% of the stem [8]. Inside the inner bark is a woody core (called the “shive”) with much shorter fibers [4]. Fiber strands are removed from the bast. These fiber strands are several meters long and are actually fiber bundles of overlapping single ultimate fibers. Bast fibers are processed by various means that may include retting, breaking, scrutching, hackling, and combing [4, 8]. The exact process depends, in a large part, on the type of plant and fiber source. For example, flax fiber can be obtained from different flax plants or from by-products from linen of flax seed production [4]. Useful natural fibers have also been derived from other parts of the plant including leaves (e.g., sisal), seeds (e.g., cotton, coir). or grass stems [3]. The production of these fibers varies greatly depending on fiber type. Some natural fibers can be spun into continuous yarns or made into nonwoven mats that allow expanded processing options for composite production. For example, much of the natural fibers used in automotive composites are currently made into fiber mats that are often needled, thermally fixed with small amounts of polymeric fibers, or otherwise modified to improve handling, and then press molded [3]. However, this additional processing comes with increased cost. The use of short fibers in more conventional processes such as injection molding is projected to increase in the future opening up new markets as new technologies overcome processing hurdles [10].

11.3
Properties

11.3.1
Chemical Components

The structure and chemical makeup of natural fibers varies greatly and depends on the source and many processing variables. However, some generalizations are possible. Natural fibers are complex, three-dimensional, polymer composites made up primarily of cellulose, hemicellulose, pectins, and lignin [12]. These hydroxyl-containing polymers are distributed throughout the fiber wall. The major chemical components of selected natural fibers are listed in Table 11.1.

Cellulose varies the least in chemical structure of the three major components and can be considered the major framework component of the fiber. It is a highly crystalline, linear polymer of anhydroglucose molecules with a degree of polymerization (*n*) around 10 000. It is the main component providing the strength, stiffness, and structural stability. Hemicelluloses are branched polymers containing 5- and 6-carbon sugars of varied chemical structure and whose molecular weights are well below those of cellulose but which still contribute as a structural component of wood [13]. Portions of the hemicelluloses are polymers of 5-carbon sugars and are called pentosans [1].

Lignin is an amorphous, cross-linked polymer network consisting of an irregular array of variously bonded hydroxy- and methoxy-substituted phenylpropane units [13]. The chemical structure varies depending on its source. Lignin is less polar than cellulose and acts as a chemical adhesive within and between fibers.

Pectins are complex polysaccharides whose main chain is a modified polymer of glucuronic acid and residues of rhamnose [3]. Side chains are rich in rhamnose, galactose, and arabinose sugars. Chains are often cross-linked by calcium ions improving structural integrity in pectin-rich areas [3]. Pectins are important in nonwood fibers especially bast fibers. The lignin, hemicelluloses, and the pectins collectively function as matrix and adhesive, helping to hold together the cellulosic framework structure of the natural composite fiber.

Natural fibers also contain lesser amounts of additional extraneous components including low molecular weight organic components (extractives) and inorganic matter (ash). Though often small in quantity, extractives can have large influences on

Table 11.1 Chemical composition of selected natural fibers [3].

Species	Cellulose	Lignin	Pectin
Flax	65–85	1-4	5–12
Kenaf	45–57	8–13	3–5
Sisal	50-64	—	—
Jute	45–63	12–25	4–10
Hardwood	40-50	20–30	0-1
Softwood	40-45	36–34	0-1

properties such as color, odor, and decay resistance [13]. The high ash content of some natural materials, such as rice hulls, causes some concern about their abrasive nature.

11.3.2
Fiber Dimensions, Density, and Mechanical Performance

Due to different species, a natural variability within species, and differences in climates and growing seasons, natural fiber dimensions as well as physical and mechanical performance can be highly variable. Methods of producing fibers with more reproducible properties are a major research effort [14].

Most natural fibers have a maximum density of about 1.5 g/cm^3. Though some natural fibers, such as wood, are hollow and have low densities in their native state, they are often densified during processing. Nevertheless, even the maximum density of these fibers is considerably less than that of inorganic fibers such as glass fibers. As such, their low density makes them attractive as reinforcement in applications where weight is a consideration.

Table 11.2 summarizes the dimensions and Table 11.3 the mechanical properties of selected natural fibers. Though variable, high aspect ratios are found especially for flax and hemp. The mechanical performance of the fibers is good but not as good as synthetic fibers such as glass. Variability in mechanical properties can be large and is due to influences such as species effects, growing conditions, and fiber harvesting or processing methods. However, their densities are considerably lower. The balance of significant reinforcing potential, low cost, and low density is part of the reason that they are attractive to industries such as the automotive industry.

11.3.3
Moisture and Durability

The major chemical constituents of natural fibers contain hydroxyl and other oxygen containing groups that attract moisture through hydrogen bonding [16]. The

Table 11.2 Dimensions of selected natural fibers.

	Length (mm)		Width (µm)		
Fiber type	Average	Range	Average	Range	References
Flax	33	9–70	19	5–38	[23]
Hemp[a)]	25	5–55	25	10-51	[23]
Kenaf	5	2–6	21	14–33	[23]
Sisal	3	1–8	20	8-41	[23]
Jute	2	2–5	20	10-25	[23]
Hardwood	1	—	—	13–45	[9]
Softwood	—	3–8	—	15–45	[8]

a) Industrial hemp is listed as a controlled substance in the United States and cannot be used in commercial production of composites.

Table 11.3 Mechanical properties of selected organic and inorganic fibers

Fiber/fiber bundles	Density (g/cm^3)	Stiffness (GPa)	Strength (MPa)	Elongation at break (%)	References
Glass	2.49	70	2700		[24]
Kevlar	1.44	124	2800	2.5	[25]
Nylon 6	1.14	1.8–2.3	503–690	1745	[25]
Polypropylene	0.91	1.6-2.4	170–325	80–100	[25]
Polyester (staple)	1.38	1.5–2.1	270–730	12–55	[25]
Flax[a)]	1.4–1.5	50–70	500–900	1.5-4.0	[3]
Hemp[a),b)]	1.48	30–60	300–800	2-4	[3]
Jute[a)]	1.3–1.5	20–55	200–500	2–3	[3]
Softwood	1.4	10–50	100–170	—	[3]
Hardwood	1.4	10–70	90–180	—	[3]

a) Fiber bundles.
b) Industrial hemp is listed as a controlled substance in the United States and cannot be used in commercial production of composites.

moisture content of these fibers can vary greatly depending on fiber type. The processing of the fiber can also have a large effect on moisture sorption. Table 11.4 shows the wide range of moisture contents for different natural fibers at several relative humidities.

This hygroscopicity can create challenges both in composite fabrication and in the performance of the end product. If natural fibers are used, a process that is insensitive to moisture must be used or the fibers must be dried before or during processing. Natural fibers absorb less moisture in the final composites since they are at least partially encapsulated by the polymer matrix. However, even small quantities of absorbed moisture can affect performance. Moisture can plasticize the fiber, altering the composite's performance. Additionally, volume changes in the fiber associated with moisture sorption can reduce fiber–matrix adhesion and damage the matrix [15]. Methods of reducing moisture sorption include adequately dispersing the fibers in

Table 11.4 Equilibrium moisture content at 27 °C of selected natural fibers [23].

	Equilibrium moisture content (%)		
Fiber	**30% Relative humidity**	**65% Relative humidity**	**90% Relative humidity**
Bamboo	4.5	8.9	14.7
Bagasse	4.4	8.8	15.8
Jute	4.6	9.9	16.3
Aspen	4.9	11.1	21.5
Southern pine	5.8	12.0	21.7
Water hyacinth	6.2	16.7	36.2
Pennywort	6.6	18.3	56.8

the matrix, limiting fiber content, improving fiber–matrix bonding, chemically modifying the fiber, or simply protecting the composite from moisture exposure.

Natural fibers undergo photochemical degradation when exposed to UV radiation [16]. They are degraded biologically because organisms recognize the chemical constituents in the cell wall and can hydrolyze them into digestible units using specific enzyme systems [16]. Though the degradability of natural fibers can be a disadvantage in durable applications where composites are exposed to harsh environments, it can also be an advantage when degradability is desired.

Due to their low thermal stability, natural fibers are generally processed with plastics where high temperatures are not required (less than about 200 "C). Above these temperatures, many of the polymeric constituents in natural fibers begin to decompose. Since cellulose is more thermally stable than other chemical constituents, highly pulped fibers that are nearly all cellulose have been used to extend this processing window [11, 17].

The release ofvolatile gases can, before, during, and after processing, lead to odor issues in applications where the composite is in an enclosed environment such as in many automotive applications and especially when moisture is present [18].

11.4
Suppliers

Natural fibers are used to manufacture a variety of products - linen, geotextiles, packaging, and specialty papers, for example. Natural fibers can be obtained from growers, distributors, importers, and as by-products from other manufacturing processes. Additionally, some companies sell semifinished products made from natural fibers (e.g., nonwoven mats) that can be further processed into composites. Due to the huge variety and diverse nature of natural fibers, there are currently few good resources that list a wide number of manufacturers. However, with the growing use of natural fibers in plastics, they are beginning to be listed in plastics industry resources. For example, the following is a list of some of the major suppliers ofnatural fibers to the North American composite industry from one industry resource [19][1]:

- Danforth Technologies (Point Pleasant, NJ, USA)
- JRS Rettenmaier (Schoolcraft, MI, USA)
- Kenaf Industries of South Texas (Lasara, TX, USA)
- Creafil Fibers (Chestertown, MD, USA)
- Rice Hull Specialty Products (Stuttgart, AR, USA)
- Stemergy (Delaware, Ontario, Canada)

Some of the major European fiber suppliers listed by another online database [20] are

- AGRO-Dienst GmbH, Germany
- Badische Naturfaseraufbereitung GmbH, Germany

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1) Private communications with Principia Partners, Exton, PA

- K.E.F.I. - Kenaf Eco Fibers Italia S.p.A., Guastalla, Italy
- Holstein Flachs GmbH, Mielsdorf, Germany
- Procotex SA Corporation, Belgium
- SANECO, France

11.5
Cost/Availability

Cost and availability of various natural fibers depend greatly on locale, region, import markets, and competing applications. For example, jute is commonly grown in India and Bangladesh, flax is prevalent in Europe, and many nonwood, natural fibers have to be imported in the United States. Although nonwood agricultural fibers and agricultural fiber wastes are abundant worldwide, their source can be diffuse and infrastructure for collection, purification, and delivery is sometimes limiting. Although there is increasing interest in commercial uses of industrial hemp worldwide, it is listed as a controlled substance in the United States and cannot be used in the commercial production of composites.

11.6
Environmental/Toxicity Considerations

The environmental benefits of wood and other natural fibers have been an important influence on their use, particularly in Europe. Natural fibers are derived from a renewable resource, do not have a large energy requirement to process, and are biodegradable [21].

Generally speaking, natural fibers are not particularly hazardous. However, natural fibers have low thermal stability relative to other reinforcing fibers and can degrade, release volatile components, and burn. Some basic precautions include avoiding high processing temperatures, using well-ventilated equipment, eliminating ignition sources, and using good dust protection, prevention, and control measures. Due to the wide variety of fibers classified as natural fibers, it is difficult to make specific comments, For information on environmental and health risks, users should consult their suppliers.

11.7
Applications (Primary and Secondary Functions)

Recently, there has been a resurgence of interest in the use of natural fibers as reinforcements in plastics due to their good mechanical performance, increasing ecological considerations in selecting materials, and legislative direction. Considerable funds have been expended on research trying to introduce flax, hemp, kenaf, and other natural fibers especially in the automotive industry with the greatest success in the use of mat technologies in panel applications. Considerable research and development is being undertaken to overcome existing limitations and expand natural fiber use into other areas such as injection-molded products.

The mechanical performance of natural fiber reinforced plastics varies greatly depending on the type of natural fibers, fiber treatments, type of plastics, additives, and processing methods. Natural fibers are added to plastics to improve mechanical performance such as stiffness and strength without increasing the density or cost too much. Though lower in mechanical performance than glass, the balance of properties of natural fibers along with other advantages such as lower density, aesthetics, and low abrasiveness during processing offer advantages in some applications. However, the generally low impact performance of natural fiber composites tends to limit their use [1].

Natural fibers are hydrophilic and do not tend to be easily wetted or bond well with many matrix materials, particularly the commodity thermoplastics. Coupling agents, such as maleated polyolefins, silanes, and isocyanates, are often necessary for adequate performance. A wide variety of coupling agents, fiber surface modifications, and treatments has been investigated for use in natural fiber plastic composites and are reviewed elsewhere [22].

Table 11.5 shows the mechanical performance of polypropylene composites made with several different natural fibers. Not surprisingly, the fibers (i.e., pulp, kenaf) are more effective reinforcements than the particulate (i.e., wood flour–low aspect ratio wood fiber bundles). Wood fibers are an order of magnitude stronger than the wood from which they derive [12] and the higher aspect ratio improves stress transfer efficiency, particularly when a coupling agent is used. Adding a maleated polypropylene coupling agent improves performance, especially flexural and tensile strengths and unnotched impact strength. Fiber preparation methods have a large effect on reinforcing ability. The high-performance dissolving pulp fibers have nearly all noncellulose components removed and are more effective than the lower cost, thermomechanical pulp fibers.

Recently, new compounding methods have been investigated to produce long, natural fiber-reinforced thermoplastic pellets and improve composite mechanical properties [31, 32]. For example, pellets have been formed by melt impregnation of continuous natural fiber yarns by pultrusion followed by cooling and chopping. Another method involves commingling of continuous forms of natural and synthetic fibers that are then heated, consolidated, and chopped.

Many reasons such as lack offamiliarity and the current, limited availability have so far prevented large-scale penetration of these materials into broader injection-molded markets. However, double-digit growth is still expected as large companies enter the market and technological advances are made [32].

Environmental considerations are also driving increased use of wood and other natural fibers since they are derived from renewable resources, do not have a large energy requirement to process, and are biodegradable [6]. They are lighter than inorganic reinforcements, which can lead to benefits such as fuel savings when their composites are used in transportation and packaging applications. A comparison of the environmental impact of natural fibers versus glass fibers is presented in

Table 11.5 Effect of selected natural fibers on the mechanical performance of several polypropylenes (all composites contain 40 wt% fiber).

		Izod impact a)		Flexural properties b)		Tensile properties')			
Filler type	Coupling agent	Notched (j/m)	Unnotched (j/m)	Maximum strength (MPa)	Modulus (GPa)	Maximum strength (MPa)	Modulus (GPa)	Elongation at break (%)	References
PP-1 d)									
None	No	20.9	656	38.3	1.19	28.5	1.53	5.9	[26]
Wood flour e)	No	22.2	73	44.2	3.03	25.4	3.87	1.9	[26]
Wood flour	Yes f)	21.2	78	53.1	3.08	32.3	4.10	1.9	[26]
Thermomechanical pulp (softwood) g)	No	22.2	90	48.9	3.10	29.7	3.68	2.1	[26]
Thermomechanical pulp (softwood)	Yes f)	21.3	150	76.5	3.50	50.2	3.89	3.2	[26]
PP-2 h)									
None	No	24	—	41	1.4	33	1.7	≫10	[27]
Dissolving pulp (softwood)')	Yes f)	—	—	82.7	3.43	60.4	4.67	4.5	j)
Kenaf k)	Yes l)	28	160	82	5.9	56	6	1.9	[27]

a) ASTM D-256 [28].
b) ASTM D-790 [29].
c) ASTM D-638 [30].
d) Fortilene 3907, polypropylene homopolymer, melt flow index = 36.5 g/10 min, Solvay Polymers, Deer Park, TX, USA.
e) Grade 4020 (American Wood Fibers, Schofield, WI, USA).
f) Maleated polypropylene (MP880, Aristech, Pittsburgh, PA, USA).
g) Laboratory produced from a mixture of pines.
h) Fortilene 1602, polypropylene homopolymer, melt flow index = 12 g/10 min, Deer Park, TX, USA.
i) High purity cellulose pulp (Ultranier-J, Rayonier Inc., Jessup, GA, USA).
j) Clemons, C.M. Unpublished data.
k) Kenaf strands obtained from AgFibers, Inc., Bakersfield, CA, USA.
l) Maleated polypropylene, G-3002. Eastman Chemical Company, Longview, TX, USA.

Chapter 7. With changing consumer perceptions, some manufacturers use the natural look of these composites as a marketing tool. Others have added natural fibers to increase bio-based material content. Natural fibers are often a preferred choice when reinforcing biodegradable polymers since natural fibers themselves are biodegradable.

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