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 agro-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.

One of the largest areas of recent growth in natural fiber plastic composites is the automotive industry, particularly in Europe, where natural fibers are advantageously used as a result of their low density and increasing environmental pressures. Most of the composites currently made with natural fibers are press-molded, although a wide range of processes have been investigated [1,5]. Table 11-1 shows the consumption of natural fibers by the European automotive industry and projections of future total consumption [1]. Flax is the most widely used natural fiber in the European automotive industry, comprising 71% of the natural fibers consumed in 2000. Most of this is short-fiber flax obtained as a by-product of the textile industry [5]. 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].

Increased technical innovations, identification of new applications, continuing political and environmental pressures, and government investment in new methods for fiber harvesting and processing are leading to projections of continued growth in the use of natural fibers in composites, with expectation of reaching 100,000 tonnes per annum by 2010 [1,6].

Functional Fillers for Plastics. Edited by M. Xanthos

ISBN 3-527-31054-1

Tab. 11-1 Consumption of natural fibers tonnes in the European automotive industry. (Reproduced with permission from ref. [1]).

Fiber type	1996	1999	2000	2005	2010
Flax	2,100	15,900	20,000	–[a]	–
Hemp[b]	0	1,700	3,500	–	–
Jute	1,100	2,100	1,700	–	–
Sisal	1,100	500	100	–	–
Kenaf	0	1,100	2,000	–	–
Coir	0	0	1,000	–	–
Total	4,300	21,300	28,300	50,000–70,000	>100,000

[a] Not estimated.
[b] Industrial hemp is listed as a controlled substance in the U.S. and cannot be used in commercial production of composites.

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 the harvesting of most natural fibers since they are used in the manufacture 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 [7].

Fiber extraction procedures depend on the type of plant and portion thereof from which the fibers are derived (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 [7]. 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 [8]. 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 structures compared to wood and, consequently, are processed quite differently. They are found in the inner bark of the stems of dicotyledons, and typically account for less than 30% of the stem [7]. 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, scutching, hackling, and combing [4,7]. The exact process depends, to a large extent, on the type of plant and fiber source. For example, flax fiber can be obtained from different flax plants or from by-products of linen or flax seed production [4]. Useful natural fibers have also been derived from other parts of plants, including leaves (e.g., sisal), seeds (e.g., coir), or grass stems [3]. The production ofthese fibers varies greatly depending on the fiber type.

Much of the natural fiber used in composites is currently made into fiber mats, which are often needled, thermally fixed with small amounts of polymer fibers, or otherwise modified to improve handling, and then press molded [3]. However, this additional step comes with increased cost. The use of short fibers in more conventional processes such as injection molding is projected to increase in the future [9]. When using natural fibers, practical processing issues such as feeding and metering of the low bulk-density fibers, as well as fiber bridging, must be addressed. Some progress has been reported on methods for pelletizing fibers to facilitate their introduction into polymer compounding equipment [10].

11.3 Properties

11.3.1 Chemical Components

The structure and chemical make-up 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 [11]. These hydroxyl-containing polymers are distributed throughout the fiber wall. The major chemical components of selected natural fibers are listed in Table 11-2, reproduced with permission from [3].

Tab. 11-2 Chemical compositions (%) 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

Of the three major components, cellulose shows the least variation in chemical structure 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 poly-

merization (n) of around 10,000, It is the main component providing the strength, stiffness, and structural stability. Hemicelluloses are branched polymers containing five- and six-carbon sugars of varied chemical structure, the molecular weights of which are well below those of cellulose but which still contribute as a structural component of wood [12]. Portions of the hemicelluloses are polymers of five-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 [12]. 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, the main chains of which consist of a modified polymer of glucuronic acid and residues of rhamnose [3]. Their side chains are rich in rhamnose, galactose, and arabinose sugars. The chains are often cross-linked by calcium ions, improving structural integrity in pectin-rich areas [3]. Pectins are important in non-wood fibers, especially bast fibers. The lignin, hemicelluloses, and 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 properties such as color, odor, and decay resistance [12]. 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 the goal of a major research effort [13].

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 ofthese fibers is considerably less than that ofinorganic fibers such as glass fibers. As such, their low density makes them attractive as reinforcements in applications where weight is a consideration.

Table 11-3 summarizes the dimensions and Table 11-4 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 that of synthetic fibers such as glass. However, their densities are considerably lower. The balance of significant reinforcing potential at low cost and low density is part of the reason why they are attractive to industries such as the automotive industry.

Tab. 11-3 Dimensions of selected natural fibers

Fiber type	Length (mm)		Width (μm)		Ref.
	Avg.	Range	Avg.	Range	
Flax	33	9–70	19	5–38	[22]
Hemp[a]	25	5–55	25	10–51	[22]
Kenaf	5	2–6	21	14–33	[22]
Sisal	3	1–8	20	8–41	[22]
Jute	2	2–5	20	10–25	[22]
Hardwood	1	–	–	15–45	[8]
Softwood	–	3–8	–	15–45	[8]

[a] Industrial hemp is listed as a controlled substance in the U.S. and cannot be used in commercial production of composites.

Tab. 11-4 Mechanical properties of selected organic and inorganic fibers.

Fiber/fiber bundles	Density ($g\,cm^{-3}$)	Stiffness (GPa)	Strength (MPa)	Strain (%)	Ref.
Glass	2.49	70	2700	–	[23]
Kevlar	1.44	124	2800	2.5	[24]
Nylon-6	1.14	1.8–2.3	503–690	17–45	[24]
Polypropylene	0.91	1.6–2.4	170–325	80–100	[24]
Polyester (staple)	1.38	1.5–2.1	270–730	12–55	[24]
Flax[a]	1.4–1.5	50–70	500–900	1.5–4.0	[3]
Hemp[a,b]	1.48	30–60	300–son	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 U.S. and cannot be used in commercial production of composites.

11.3.3 Moisture and Durability

The major chemical constituents ofnatural fibers contain hydroxyl and other oxygen-containing groups that attract moisture through hydrogen bonding [15]. The moisture content of these fibers can vary greatly depending on the fiber type. The processing of the fiber can also have a large effect on moisture sorption. Table 11-5 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 ab-

Tab. 11-5 Equilibrium moisture content at 27 °C of selected natural fibers [22]

Fiber	Equilibrium moisture content (%)		
	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

sorbed 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 [14]. Methods of reducing moisture sorption include adequately dispersing and encapsulating the fibers in the matrix during compounding, 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 [15]. 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 [15]. 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 for which high temperatures are not required (less than about 200 "C). Above such 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 [16].

The release of volatile 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 [17].

11.4 Suppliers

Natural fibers are used to manufacture a variety ofproducts –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 semi-finished products made from natural fibers (e.g., non-woven 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 large 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 of natural fibers to the North American composite industry from one industry resource [18].

- Danforth Technologies (Point Pleasant, New Jersey, USA)
- Flaxcraft (Cresskill, New Jersey, USA)
- JRS Rettenmaier (Schoolcraft, Michigan, USA)
- Kenaf Industries of South Texas (Lasara, Texas, USA)
- Rayonier (Jesup, Georgia, USA)
- Creafill Fibers (Chestertown, MD, USA)
- Rice Hull Specialty Products (Stuttgart, Arkansas, USA)

Some of the major European fiber suppliers listed by another, online database [19] are:

- AGRO-Dienst GmbH, Germany
- Badische Naturfaseraufbereitung GmbH, Germany
- 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, and import markets. For example, jute is commonly grown in India and Bangladesh, flax is prevalent in Europe, and many non-wood natural fibers have to be imported into the United States. Although non-wood agricultural fibers and agricultural fiber wastes are abundant worldwide, their source can be diffuse and infrastructure for their 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 U.S. and cannot be used in the commercial production of composites.

The cost ofnatural fibers greatly depends on such factors as fiber type, location, degree of refinement, other markets for natural fibers, etc. However, some pricing for natural fibers of interest in North America is given in Table 11-6 [25].

Tab. 11-6 Pricing of selected natural fibers (Reprinted with permission from ref. [25]).

Fiber type	Price (FoB, US$/kg)
Flax: core (6.4 mm and less)	0.44
Flax: long (>6.4 mm)	>0.73
Kenaf	0.68
Sisal (–3.2 mm)	1.03
Hemp:[a] core (6.4 mm and less)	0.55
Hemp:[a] long (>6.4 mm)	>0.88

[a] Industrial hemp is listed as a controlled substance in the U.S. and cannot be used in commercial production of composites.

11.6 Environmental/Toxicity Considerations

The environmental benefits of wood fibers 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 [20].

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 and perceived environmental advantages. Considerable funds have been expended on research aimed at introducing flax, hemp, kenaf, and other natural fibers, especially in the automotive industry, with the greatest success being achieved in the use of mat technologies in panel applications. However, issues such as: 1) lack of established delivery channels, 2) processing complications due to the low density of the fibers (feeding, metering, and bridging), and 3) performance issues such as odor control, are limiting the wider use of natural fibers as reinforcements in thermoplastics. These are active areas of research and development.

11.7.1
Mechanical Property Modification (Primary Function)

The mechanical performance of natural fiber reinforced plastics varies greatly depending on the type of natural fibers, fiber treatments, the type of plastic, 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. The generally low impact performance of natural fiber composites tends to limit their use and addressing this issue is an active area ofresearch [1].

Natural fibers are hydrophilic and do not tend to be easily wetted or to 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 and fiber surface modifications and treatments has been investigated for use in natural fiber plastic composites and these are reviewed elsewhere [21].

Table 11-7 shows the mechanical performances of polypropylene composites made with several different natural fibers. Not surprisingly, the fibers (i.e., pulp fibers, kenaf fiber) are more effective reinforcements than the particulates (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 [11] and the higher aspect ratio improves stress transfer efficiency, particularly when a coupling agent is used. The addition of a coupling agent, namely maleated polypropylene, was found to improve performance, especially flexural and tensile strengths and unnotched impact energy. Fiber preparation methods proved to have a large effect on reinforcing ability. The high performance dissolving pulp fibers have nearly all of their non-cellulose components removed and are more effective than the lower cost, thermomechanical pulp fibers.

11.7.2
Environmental Preference and Biodegradability (Secondary Function)

The environmental advantages of natural fibers are an important influence, particularly in Europe. Natural fibers are derived from renewable resources and often from industrial by-products. Wood and other natural fibers 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 applications. Natural fibers can be used to reinforce biodegradable polymers since natural fibers themselves are biodegradable.

Tab. 11-7 Effect ofselected natural fibers on the mechanical performance of several polypropylenes (all composites contain 40 wt. % fiber).

		Izod Impact[b]		Flexural Propertles[c]		Tensile Properties[d]			
Filler Type	**Coupling Agent**	**Notched ($J\ m^{-1}$)**	**Un-notched ($J\ m^{-1}$)**	**Max. strength (MPa)**	**Modulus (GPa)**	**Max. strength (MPa)**	**Modulus (GPa)**	**Elongation at break (%)**	**Ref.**
PP-1[a]									
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[g]	21.2	78	53.1	3.08	32.3	4.10	1.9	[26]
Thermomechanical pulp (softwood)[f]	No	22.2	90	48.9	3.10	29.7	3.68	2.1	[26]
Thermomechanical pulp (softwood)	Yes[g]	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	[28]
Dissolving pulp (softwood)[i]	Yes[g]	–	–	82.7	3.43	60.4	4.67	4.5	[27]
Kenaf[j]	Yes[k]	28	160	82	5.9	56	6	1.9	[28]

[a] Fortilene 3907, polypropylene homopolymer, melt flow index = 36.5 g/10 min., Solvay Polymers, Deer Park, TX, USA.
[b] ASTM D-256 [29].
[c] ASTM D-790 [30].
[d] ASTM D-638 [31].
[e] Grade 4020 (American Wood Fibers, Schofield, WI, USA).
[f] Laboratory produced from a mixture of pines.
[g] Maleated polypropylene (MP880, Aristech, Pittsburgh, PA, USA).
[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] Kenaf strands obtained from AgFibers, Inc., Bakersfield, CA, USA.
[k] Maleated polypropylene, G-3002, Eastman Chemical Company, Longview, TX, USA.

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Functional Fillers for Plastics

Edited by Marino Xanthos

WILEY-VCH Verlag GmbH & Co. KGaA

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Library of congress Card No.: Applied for

British Library Cataloguing-in-Publication Data:
A catalogue record for this book is available from the British Library.

Bibliographic information published by Die Deutsche Bibliothek
Die Deutsche Bibliothek lists this publication in the Deutsche Nationalbibliografie: detailed bibliographic data is available in the Internet at <http://dnb.ddb.de>.

Printed in the Federal Republic of Germany

Printed on acid-free paper

Composition TypoDesign Hecker GmbH, Leimen
Printing Strauss GmbH, Mörlenbach
Bookbinding Litges & Dopf Buchbinderei GmbH, Heppenheim

ISBN-13 978-3-527-31054-8
ISBN-10 3-527-31054-1