# Natural Fiber Composites

## edited by R.D.S.G. Campilho



## Natural Fiber Composites

# Natural Fiber Composites

## edited by R.D.S.G. Campilho



CRC Press is an imprint of the Taylor & Francis Group, an **informa** business CRC Press Taylor & Francis Group 6000 Broken Sound Parkway NW, Suite 300 Boca Raton, FL 33487-2742

© 2016 by Taylor & Francis Group, LLC CRC Press is an imprint of Taylor & Francis Group, an Informa business

No claim to original U.S. Government works Version Date: 20150914

International Standard Book Number-13: 978-1-4822-3901-0 (eBook - PDF)

This book contains information obtained from authentic and highly regarded sources. Reasonable efforts have been made to publish reliable data and information, but the author and publisher cannot assume responsibility for the validity of all materials or the consequences of their use. The authors and publishers have attempted to trace the copyright holders of all material reproduced in this publication and apologize to copyright holders if permission to publish in this form has not been obtained. If any copyright material has not been acknowledged please write and let us know so we may rectify in any future reprint.

Except as permitted under U.S. Copyright Law, no part of this book may be reprinted, reproduced, transmitted, or utilized in any form by any electronic, mechanical, or other means, now known or hereafter invented, including photocopying, microfilming, and recording, or in any information storage or retrieval system, without written permission from the publishers.

For permission to photocopy or use material electronically from this work, please access www.copyright.com (http://www.copyright.com/) or contact the Copyright Clearance Center, Inc. (CCC), 222 Rosewood Drive, Danvers, MA 01923, 978-750-8400. CCC is a not-for-profit organization that provides licenses and registration for a variety of users. For organizations that have been granted a photocopy license by the CCC, a separate system of payment has been arranged.

**Trademark Notice:** Product or corporate names may be trademarks or registered trademarks, and are used only for identification and explanation without intent to infringe.

Visit the Taylor & Francis Web site at http://www.taylorandfrancis.com

and the CRC Press Web site at http://www.crcpress.com

## Contents

Preface	vii
Editor	ix
Contributor	sXi
Chapter 1	Introduction to Natural Fiber Composites1
	R.D.S.G. Campilho
Chapter 2	Natural Fibers and Their Characterization
	Z.N. Azwa, B.F. Yousif, A.C. Manalo, and W. Karunasena
Chapter 3	Alternative Solutions for Reinforcement of Thermoplastic Composites
	Nadir Ayrilmis and Alireza Ashori
Chapter 4	Matrices for Natural Fiber Composites
	Juan Carlos Domínguez, Mercedes Oliet, María Virginia Alonso, and Bo Madsen
Chapter 5	Interfacial Compatibility and Adhesion in Natural Fiber Composites
	Le Quan Ngoc Tran, Carlos A. Fuentes, Ignace Verpoest, and Aart Willem Van Vuure
Chapter 6	Processing of Natural Fiber Composites
	Shinichi Shibata
Chapter 7	Testing and Characterization of Natural Fiber-Reinforced Composites
	H.N. Dhakal, J. MacMullen, and Z.Y. Zhang
Chapter 8	Environment-Related Issues
	Samrat Mukhopadhyay

Chapter 9	Modeling of Natural Fiber Composites	221
	Liva Pupure, Janis Varna, and Roberts Joffe	
Chapter 10	Design of Natural Fiber-Reinforced Composite Structures S.M. Sapuan and M.R. Mansor	255
Chapter 11	Joint Design in Natural Fiber Composites R.D.S.G. Campilho and Lucas F.M. da Silva	279
Chapter 12	State-of-the-Art Applications of Natural Fiber Composites in the Industry Dipa Ray	319
Index		341

### Preface

Composite materials have come to the fore a few decades ago because of their superior specific mechanical properties as a result of the increasing demand of both consumers and industries for highly performing materials and structures. However, the combination of the fibers with the aggregating material or matrix highly increases the complexity of the design process and usually leads to challenges in the composite engineering and, correspondingly, to more conservative solutions for a given application. Although the success of these materials is obvious, recently, a general consensus all around the world was reached regarding the negative influence of human beings on global warming and the environment. The best way in which the environment could be conserved is through the use of renewable and nontoxic natural materials, and all efforts should be undertaken to make them competitive. Actually, environmental awareness all around the world has led to the research and development of cheap and biodegradable materials that are concurrently available from nature. This triggered interest in more sustainable materials that could be processed with lower energy consumption, such as natural fiber composites. Recycling of natural fiber composites and natural fiber reinforcement of waste materials are other steps used for saving resources and the environment. Although the use of these materials dates back to civilization itself, it is clear that renewed incentives for their use are emerging. Thus, scientists and engineers have become more interested in the study of natural fibers and their composites. The replacement of conventional materials and artificial composites with natural fiber composites can thus become a reality, contributing towards the creation of a sustainable economy. On the other hand, concerns on the availability of petrochemicals in the future can also trigger the use of natural fiber composites. On account of large research efforts in fiber extraction and chemical treatments, fiber-matrix adhesion, or processing conditions, natural fiber composites are currently a viable replacement for glass composites in many applications in terms of both mechanical strength and a lower price. Actually, by treating the fibers with coupling agents, engineering the fiber orientation of the natural fiber components, devising extraction techniques to increase the fiber length, and combining with the best possible matrix, very interesting characteristics have been found. These achievements and the superior environmental performance are important drivers for the growing use of natural fiber composites in the near future. Despite all these advantages, some features still prevent a more widespread use of these materials, such as the strength prediction during structural loading and uncertainties about long-term performance. However, it is expected that a lot of useful information previously gathered for artificial composites can be applied to these materials.

This book is comprised of 12 independently written chapters covering the most relevant topics related to introductory knowledge on natural fiber composites, material properties, treatment and processing, modeling, design, and applications. The first chapter is introductory, giving an overview of natural fiber composites, and each of the next few chapters deals with a specific issue of paramount importance that is required to understand and to be able to analyze and design structural components in such materials. The initial chapters discuss issues such as the characterization of natural fibers, matrices, and respective composites. At this stage, relevant information is provided on how to choose the best possible set of materials for a specific application given the design requirements. Methods that enhance the performance and processing techniques follow these initial discussions, enabling us to understand how to improve the strength of the fabricated composites and also which is the most suitable processing technique, respectively. Testing should always be considered during design as a safeguard against design mistakes and to study how the structures behave under service conditions. Environmental issues are not forgotten, and many related aspects are discussed. The last chapters focus on modeling, design issues, and applications. Modeling aims at providing the necessary tools to design natural fiber composites back at the office, as well as at reducing prototype testing to a minimum. Design is related to the overall design process and tools that are used to bring the product to life. Joint design is also included as structures in general usually require some means of joining either because of their dimensions or due to their complex shape, which prevents construction in a single piece. The chapter on applications overviews past, present, and potential applications of these materials based on their characteristics showing cases of success that substantiate the future bet on natural fiber composites. Together, this set of subjects aims at enabling the reader to analyze and design natural fiber composite structures in a scientifically supported manner with the assurance of using state-of-the-art information and methods.

This book has an internationally recognized team of contributors with each one writing about their specific field of knowledge and, thus, providing the best overview of each particular subject. As the editor of this book, it was a great pleasure for me to work with the expert contributors in this book.

R.D.S.G. Campilho ISEP Portugal

## Editor

**R.D.S.G. Campilho** was born in 1979. In 2003, he graduated in mechanical engineering at the Instituto Superior de Engenharia do Porto (ISEP), Porto, Portugal. He completed his MS degree in 2006 and his doctoral degree in 2009, both at Faculdade de Engenharia da Universidade do Porto, Porto, Portugal. He currently serves as an assistant professor at ISEP, where he teaches mechanical engineering. He is an active researcher in numerical modeling, finite element methods, cohesive zone models for fracture behavior, natural and artificial composite materials, and adhesive joint design.

## Contributors

#### María Virginia Alonso

Department of Chemical Engineering Complutense University of Madrid Madrid, Spain

#### Alireza Ashori

Department of Chemical Technologies Iranian Research Organization for Science and Technology (IROST) Tehran, Iran

#### Nadir Ayrilmis

Department of Wood Mechanics and Technology Faculty of Forestry Istanbul University Istanbul, Turkey

#### Z.N. Azwa

Faculty of Health, Engineering and Sciences University of Southern Queensland Toowoomba, Australia

#### R.D.S.G. Campilho

Departamento de Engenharia Mecânica Instituto Superior de Engenharia do Porto Instituto Politécnico do Porto Porto, Portugal

#### Lucas F.M. da Silva

Departamento de Engenharia Mecânica Faculdade de Engenharia Universidade do Porto Porto, Portugal

#### H.N. Dhakal

Advanced Polymer and Composites (APC) Research Group School of Engineering University of Portsmouth Portsmouth, United Kingdom

#### Juan Carlos Domínguez

Department of Chemical Engineering Complutense University of Madrid Madrid, Spain

#### **Carlos A. Fuentes**

Department of Materials Engineering (MTM) KU Leuven Leuven, Belgium

#### **Roberts Joffe**

Division of Material Sciences Lulea University of Technology Lulea, Sweden

#### W. Karunasena

Faculty of Health, Engineering and Sciences University of Southern Queensland Toowoomba, Australia

#### J. MacMullen

Advanced Polymer and Composites (APC) Research Group School of Engineering University of Portsmouth Portsmouth, United Kingdom

#### **Bo Madsen**

Department of Wind Energy Section of Composites and Materials Mechanics Technical University of Denmark Roskilde, Denmark

#### A.C. Manalo

Faculty of Health, Engineering and Sciences University of Southern Queensland Toowoomba, Australia

#### M.R. Mansor

Faculty of Mechanical Engineering Universiti Teknikal Malaysia Melaka Melaka, Malaysia

#### Samrat Mukhopadhyay

Department of Textile Technology IIT Delhi New Delhi, India

#### **Mercedes Oliet**

Department of Chemical Engineering Complutense University of Madrid Madrid, Spain

#### Liva Pupure

Division of Material Sciences Lulea University of Technology Lulea, Sweden

#### Dipa Ray

Irish Centre for Composites Research (ICOMP) Mechanical, Aeronautical and Biomedical Engineering Department Materials and Surface Science Institute University of Limerick Limerick, Ireland

#### S.M. Sapuan

Department of Mechanical and Manufacturing Engineering Universiti Putra Malaysia Selangor, Malaysia

#### Shinichi Shibata

University of the Ryukyus Nishihara, Japan

#### Le Quan Ngoc Tran

Singapore Institute of Manufacturing Technology Agency for Science, Technology and Research (A\*STAR) Singapore

#### Aart Willem Van Vuure

Department of Materials Engineering (MTM) KU Leuven Leuven, Belgium

#### Janis Varna Division of Material Sciences Lulea University of Technology Lulea, Sweden

#### Ignace Verpoest

Department of Materials Engineering (MTM) KU Leuven Leuven, Belgium

#### **B.F. Yousif** Faculty of Health, Engineering and Sciences University of Southern Queensland Toowoomba, Australia

#### Z.Y. Zhang

Advance Polymer and Composites (APC) Research Group School of Engineering University of Portsmouth Portsmouth, United Kingdom

## 1 Introduction to Natural Fiber Composites

R.D.S.G. Campilho

#### CONTENTS

1.1	Introduction	1
1.2	Natural Fibers	4
1.3	Biopolymers	10
1.4	Biocomposites	11
1.5	Benefits and Applications of Natural Fiber Composites	15
1.6	Life Cycle Assessment of Natural Fiber Composites	16
1.7	Potential of Natural Fiber Composites and Drivers for Change	22
	1.7.1 Mechanical Properties	22
	1.7.2 Environment	23
	1.7.3 Cost-Effectiveness	24
1.8	Challenges in the Use of Natural Fiber Composites	24
1.9	Conclusions	27
Refe	rences	

#### 1.1 INTRODUCTION

Composite materials have come to the fore a few decades ago because of their superior specific mechanical properties, as a result of the increasing demand of both consumers and industries for highly performing materials and structures. However, the combination of the fibers with the aggregating material or matrix highly increases the complexity of the design process and usually leads to challenges in the composite engineering and, correspondingly, to more conservative solutions for a given application. Although the success of these materials is obvious, recently, a general consensus all around the world was reached regarding the negative influence of the human being on global warming and the environment. The best way in which the environment could be conserved is by using renewable and nontoxic natural materials, and all efforts should be undertaken to make them competitive. Actually, the environmental consciousness all around the world has led to the research and development of the next generation of materials, products, and processes [1]. Within this scope, it is necessary to develop cheap and biodegradable materials that are concurrently available from nature. This awareness triggered interest in more sustainable materials that are able to be processed with lower energy consumption, such as natural fiber composites. As a result, natural fiber composites are under intense investigation because of their potential as alternatives for synthetic fibers. This century, in particular, has witnessed major improvements in sustainable technology and biocomposites, and the interest in these issues is still increasing. Recycling of natural fiber composites and natural fiber reinforcement of waste materials are other steps for conserving resources and the environment. Because of these issues, biocomposites are gaining industrial interest in a world focused on environmental outcomes.

The use of these materials dates back to civilization itself and, for many centuries, natural fibers have been used as raw material. Natural fibers were initially used around 3000 years ago along with clay in Egypt, and they have been used ever since. Recently, it is clear that renewed incentives for their use are emerging. Thus, scientists and engineers have become more interested in the study of natural fibers and their composites. The replacement of conventional materials and synthetic composites with natural fiber composites can thus become a reality, contributing to the creation of a sustainable economy. On the other hand, concerns on the availability of petrochemicals in the future could also trigger the use of natural fiber composites due to the induced pressure from the global market. Because of this, natural polymers are also gaining ground as matrix materials and are taking their market share. It should, however, be noticed that biodegradability is not the sole attribute of natural materials: some synthetic materials can be biodegradable, whereas some natural materials may not be. Obviously, an ideal natural fiber composite is fully biodegradable under controlled conditions and is composed only of short-cycle renewable plants. On account of large research efforts in fiber extraction and chemical treatments, fiber-matrix adhesion, or processing conditions, natural fiber composites are currently considered a viable replacement for glass composites in many applications in terms of both mechanical strength and a lower price. Actually, by treating the fibers with coupling agents, engineering the fiber orientation of the natural fiber components, devising extraction techniques to increase the fiber length, and combining with the best possible matrix, very interesting characteristics are achievable. Other advantages include the large availability, renewability of raw materials, flexibility during processing, low cost, low density, and, because of this, high specific strength and stiffness. Compared with synthetic fibers, energy requirements for processing are much lower, and energy recovery is also possible. Kim et al. [2] showed that natural fiber composites have a higher energy absorption rate under impact loadings than glass-reinforced composites. These achievements and the superior environmental performance are important drivers for the growing use of natural fiber composites in the near future, and they enable these materials to be attractive to industrial companies. Despite all these advantages, some features still prevent a more widespread use of these materials, such as the strength prediction during structural loading, uncertainties about the long-term performance, moisture absorption, lower fire resistance, lower mechanical properties and durability, limited processing temperatures, larger scatter in the cost and properties than synthetic composites, and some difficulties in the use of well-known fabrication processes [3]. However, it is expected that a lot of useful information previously gathered for synthetic composites can be applied to these materials. In fact, many efforts are being made to address the mentioned limitations, with attention to surface treatments for the fibers and interfacial improvement with the matrix. Natural fiber composites with a thermoplastic matrix (e.g., polyethylene [PE], polypropylene [PP], or polyvinyl chloride [PVC]) are also recent solutions. There is equal potential for biodegradable polymers to replace synthetic ones in the near future, at least in applications that do not require a long lifespan, and these matrices have recently seen an important increase in industrial applications. Regarding the production volumes, the main products are starch-based plastics, poly(lactic acid) (PLA), and microbial synthesis polymers or polyhydroxyalkanoates (PHA) [4]. As a result of intensive research and development, these materials became competitors with conventional engineering materials in some fields of application, with new compositions and manufacturing processes emerging. The research interest in natural fiber composites has been consistent over the past two decades, but this has not yet translated into a large range of industrial applications.

In the industry, several companies are becoming increasingly interested in using materials that weigh less, are durable and ecologically efficient, and present interesting mechanical properties. Within this scope, natural fibers are highly valued since they come at a low cost, are recyclable and biodegradable, can be easily processed, and have a very low density. Because of this, the use of natural fiber reinforcement will likely highly increase over the next few years. According to the technical report by Lucintel [5], the global market on natural fiber composites had reached US\$289.3 million in 2010, with a compound annual growth rate (CAGR) of 15% from 2005 onward. By 2016, the natural fiber market should reach nearly US\$550 million, with CAGR being reduced to around 11%. In terms of applications, the global market for natural fibers is mainly divided into two: wood and nonwood fibers. Wood fibers are typically used in the construction industry, and this application is more widespread in North America. On the other hand, nonwood natural fiber applications thrive in Europe, with tremendous growth mainly in the automotive industry, by using thermoplastic and thermoset-based natural fiber composites, because of issues such as raw material renewability, environmentally sound materials, good sound insulation properties, and fuel saving, on account of the smaller component weight. This usage was made possible by large investments and development in using compression molding as the adopted process in the European automotive industry. Automotive applications include door interior panels, package trays, trunk liners, and seat backs. More specific examples are interior vehicle parts such as door trim panels made of natural composites with polypropylene matrix, or exterior parts, for example, engine or transmission covers, with polyester reinforced with natural fibers [6]. This change was triggered by the European Union End-of-Life Vehicle Directive (2000), stipulating that 80 wt.% of a waste vehicle should be reused or recycled. On account of this directive, the use of these materials has been increasing in the past years. For vehicle applications, using thermoplastic matrices rather than thermoset gives some advantages, such as increased design possibilities, since fabrication by injection molding and extrusion become feasible, in addition to the possibility of recycling. In civil engineering, natural fiber composites can also play an important role because of the lower weight and cost of natural fiber reinforcement plates, compared with carbonor glass-based composites. Natural fiber fabrics are easier to handle, with advantages in column wrapping for posterior cure with temperature, and are acoustic insulators.

However, according to Dittenber and GangaRao [3], there is a major ecological benefit of using natural fiber composites in construction, since these materials enable the fabrication of large and biodegradable structures only with natural resources and with a reduced amount of embodied energy. Extruded natural fiber composites for decking applications are used in the United States because of the generous thickness of the plates, which allows overcoming limitations with regard to the mechanical properties. Regarding the global usage of natural fibers, Europe is the largest consumer, and Asia is becoming a big market for natural fibers because of the increasing demand in both China and India. In the near future, a fragmentation of the natural fibers market is expected because of emerging economies [5]. Bio-based plastics also follow this increasing tendency of natural fiber composites, with past growth rates of 38% between 2003 and 2007 (worldwide), reaching 48% in Europe alone. The fabrication capacity of bio-based plastics increased from 0.36 million tons in 2003 to 2.33 million tons in 2013, and it is expected to increase further to 3.45 million tons in 2020 [7]. Global markets both now and in the future should be very competitive, striving to get the best possible materials, and those companies that show innovation in this area will perform the best. On account of their potential, natural materials can play a very important role in the near future for the success of industries.

#### 1.2 NATURAL FIBERS

Nonrenewable resources are becoming scarcer on the planet, and a generalized awareness exists regarding renewable resources and products. Because of this, different natural fibers, or species of natural plants that can result in natural reinforcement fibers, are always appearing. There are three ways in which natural fibers can be used: in textiles, paper, and fabrics; for biofuel; and as reinforcement material for composites. As for reinforcement, natural fibers can eventually be used to replace glass fibers in some applications, providing composite parts to be used in the automotive industry, construction, and packaging. Natural fibers can be categorized according to their origin (Figure 1.1): lignocellulosic materials, animals, or minerals [6,8–9]. Lignocellulosic fibers, also known as cellulose-based fibers, can be divided into wood and nonwood or plant fibers. Wood fibers are undoubtedly the most abundant. Plant fibers also have an important market share, and these consist of cellulose, hemicellulose, lignin, and pectin [6]. Many of the fiber properties can be approximated by the relative content of these constituents. Nonwood lignocellulosic fibers are divided into seed fibers, leaf fibers, bast or stem fibers, fruit fibers, and stalk fibers. Most industrial fibers are from bast (e.g., hemp, flax, kenaf, and jute). These fibers are collected from the phloem that surrounds the stem and exist in plants of a certain required height; this enables fibers with high stiffness to maintain stability. Fibers from leafs (e.g., sisal) are also common as raw materials but generally suffer from lower stiffness. Figure 1.2 shows some examples of natural fibers and natural fiber fabrics [9].

The plants that originate fibers can be viewed as primary or secondary, as a function or role of the fiber in the plant. Primary plants, such as jute, hemp, kenaf, or sisal, are grown with the sole objective of providing fibers for industrial usage. Secondary plants (e.g., pineapple, oil palm, or coir) have a different main purpose, such as that



**FIGURE 1.1** Classification of natural fibers. (From Saxena, M. et al., *Advances in composite materials – Analysis of natural and man-made materials*, Rijeka: InTech, 121–162, 2011; Technologies and products of natural fiber composites. CIP-EIP-Eco-Innovation-2008: Pilot and market replication projects – ID: ECO/10/277331; Majeed, K. et al., *Materials and Design*, 46, 391–410, 2013 [6,8–9].)

of a human food source. In general, lignocellulosic natural fibers such as flax, hemp, henequen, sisal, coconut, jute straw, palm, bamboo, rice husk, wheat, barley, oats, rye, cane (sugar and bamboo), reeds, kenaf, ramie, oil palm, coir, banana fiber, pineapple leaf, papyrus, wood, or paper have been used as reinforcement in thermosetting and thermoplastic resin composites [10]. Fabricated products in natural composites include door and trunk liners, parcel shelves, seat backs, interior sunroof shields, and headrests [11]. Table 1.1 details the most commercially used natural fibers, in terms of annual worldwide production. Natural fibers usually have a diameter on the order of 10  $\mu$ m and are, by themselves, a composite material, since they are composed by a primary cell wall and three secondary cell walls. The cell walls include microfibrils that are randomly oriented. The angle of the microfibrils with respect to the fiber axis has a major role in the fiber properties, given that smaller angles give high strength and stiffness, whereas larger angles provide ductility [12]. Since fibers are bundled together by lignin and fixed to the stem by pectin (both of which are weaker than cellulose), these constituents must be removed for the fibers to attain the maximum reinforcement effect. Fibers are still used in bundles connected by lignin, since this is less time consuming, but the overall strength is smaller than using the isolated fibers. The length of the fibers also plays an important role in the composite strength, especially when the interfacial adhesion is weak. Compared with glass or carbon



**FIGURE 1.2** Natural fibers: banana (a), sugarcane bagasse (b), curaua (c), flax (d), hemp (e), jute (f), sisal (g), and kenaf (h). Natural fiber fabrics: jute (i), ramie–cotton (j), and jute–cotton (k). (From Majeed, K. et al., *Materials and Design*, 46, 391–410, 2013 [9].)

TABLE 1.1 Worldwide Production of Most	FABLE 1.1   Worldwide Production of Most Used Commercial Natural Fibers					
Fiber Type	World Production (10 <sup>3</sup> ton)					
Sugarcane bagasse	75,000					
Bamboo	30,000					
Jute	2,300					
Kenaf	970					
Flax	830					
Grass	700					
Sisal	378					
Hemp	214					
Coir	100					
Ramie	100					
Abaca	70					

fibers, natural fibers benefit from lower density, less tool wear during machining, no health hazards, biodegradability, availability of natural and renewable sources, and lower cost per unit volume basis [13–14]. Natural fibers also provide a higher degree of design flexibility, because they will bend rather than break during processing. However, their specific stiffness and strength do not match those of synthetic fibers, and they suffer from high moisture absorption and poor wettability to some resins. Natural fibers generally work well as reinforcements of inorganic polymers, synthetic

polymers, and natural polymers because of their high strength and stiffness as well as low density [15]. Typical strength and stiffness values for flax fibers are actually close to those of *E*-glass fibers [16], which, in turn, gives higher specific properties on account of the smaller density. However, being materials of a natural origin, the scatter in mechanical properties is higher than for synthetic fibers, because of variations in the fiber structure emerging from changing climate conditions during growth (where the fibers are sourced), area of growth, age of the plant, processing methods, and fiber modifications [7,17]. The lack of standardized procedures for testing natural fibers also helps in the scattering of properties. Table 1.2 shows the main factors related to the stage of production that affect the fiber properties [3].

Other drawbacks include the difficulty to create a strong bond between the fibers and matrix, and the moisture absorption, with consequences on the composite strength. Many other factors influence the behavior of fibers, such as their length, physical properties (e.g., dimensions, defects, structure, and cell wall thickness), cellulose content, and spiral angle of the cell layers. Some variations in the chemical composition also exist between plants of the same species; among the plant constituents (stalk and root); and between world region, age, environmental conditions, and soil characteristics. Table 1.3 compares the most relevant mechanical properties of some nonwood lignocellulosic fibers and synthetic fibers (for comparison) [3,18]. Because of the high degree of variability of natural fibers and testing methods, the mechanical properties have a large scatter. Another feature is their hollow nature, which not only offers the potential for reduced weight but is also a challenge for waterproofing [19]. For comparison purposes, the most typical values for each quantity

riber rioperties	
Stage	Factors Affecting Fiber Properties
Plant growth	Plant species
	Crop cultivation
	Crop location
	Fiber location in plants
	Climate
Harvesting	Fiber ripeness, which affects:
	Cell wall thickness
	Fiber coarseness
	Fiber-structure adhesion
Fiber extraction	Decortication process
	Type of retting method
Supply	Transportation conditions
	Storage conditions
	Age of fibers

#### TABLE 1.2 Factors Related to the Production of Natural Fibers That Affect Fiber Properties

Source: Dittenber, D.B., and GangaRao, H.V.S., Composites: Part A, 43, 1419–1429, 2012 [3].

70.04	~~~ /													
	Density	Length	Diameter	Tensile Strength	Tensile Modulus	Specific Modulus	Elongation	Cellulose	Hemicellulose	Lignin	Pectin	Waxes	Micro-fibrillar Angle	Moisture Content
Fiber	(g/cm <sup>3</sup> )	(mm)	(m1)	(MPa)	(GPa)	(approx.)	(%)	(wt.%)	(wt.%)	(wt.%)	(wt.%)	(wt.%)	(degrees)	(wt.%)
E-glass	2.5-2.59	I	<17	2000-3500	70–76	29	1.8-4.8		I					I
Abaca	1.5	Ι	Ι	400–980	6.2 - 20	6	1.0 - 10	56-63	20-25	7–13	1	3		5-10
Alfa	0.89	Ι	Ι	35	22	25	5.8	45.4	38.5	14.9	Ι	2		I
Bagasse	1.25	10-300	10–34	222–290	17-27.1	18	1.1	32-55.2	16.8	19–25.3	I	I		I
Bamboo	0.6 - 1.1	1.5-4	25-40	140-800	11–32	25	2.5-3.7	26-65	30	5-31	I	I		I
Banana	1.35	300-900	12–30	500	12	6	1.5-9	63-67.6	10–19	5	I			8.7–12
Coir	1.15-1.46	20-150	10-460	95-230	2.8–6	4	15-51.4	32-43.8	0.15-20	40-45	3-4	I	30-49	8.0
Cotton	1.5-1.6	1060	10-45	287-800	5.5-12.6	9	3-10	82.7–90	5.7	4	0-1	0.6		7.85-8.5
Curaua	1.4	35	7-10	87-1150	11.8–96	39	1.3-4.9	70.7-73.6	9.6	7.5-11.1				
Flax	1.4 - 1.5	5-900	12-600	343-2000	27.6-103	45	1.2-3.3	62-72	18.6–20.6	2-5	2.3	1.5-1.7	5-10	8-12
Hemp	1.4 - 1.5	5-55	25-500	270–900	23.5-90	40	1 - 3.5	68-74.4	15-22.4	3.7 - 10	0.9	0.8	2-6.2	6.2-12
Henequen	1.2	I	Ι	430–570	10.1 - 16.3	11	3.7-5.9	60-77.6	4–28	8-13.1		0.5		I
Isora	1.2 - 1.3	I	Ι	500-600		I	5-6	74		23		1.09		I
Jute	1.3 - 1.49	1.5-120	20-200	320-800	8-78	30	1-1.8	59-71.5	13.6-20.4	11.8-13	0.2 - 0.4	0.5	8.0	12.5-13.7
Kenaf	1.4	I	Ι	223-930	14.5-53	24	1.5-2.7	31-72	20.3-21.5	8-19	3-5		I	
Nettle	Ι	Ι	Ι	650	38	Ι	1.7	86	10	Ι	Ι	4	Ι	11-17
Oil Palm	0.7 - 1.55	Ι	150 - 500	80–248	0.5 - 3.2	2	17-25	60-65	I	11-29	I	I	4246	Ι
Piassava	1.4		I	134-143	1.07-4.59	2	7.8-21.9	28.6	25.8	45			I	
PALF	0.8 - 1.6	900-1500	20-80	180-1627	1.44-82.5	35	1.6 - 14.5	70-83		5-12.7			14.0	11.8
Ramie	1.0 - 1.55	900-1200	20-80	400 - 1000	24.5-128	60	1.2 - 4.0	68.6-85	13-16.7	0.5 - 0.7	1.9	0.3	7.5	7.5–17
Sisal	1.33-1.5	006	8-200	363-700	9.0–38	17	2.0-7.0	60–78	10.0-14.2	8.0 - 14	10.0	2.0	10-22	10–22
L.	Distantion I		II on Door	0 011	L L	0171 CF Y		Doubour 0. Doubour	1 T				oto Door Doto	Tool Tool
Source.	DINGINGI' I	J.D., allu U	ialigaNa0, n	.v.s., compo	sues. Furly	4, 40, 1419	-1429, 2017	2, Dalucio,	E.J., Introducti	on to com	nostre mai	eriuis ues	ign. Duca Naiu	11. 1aylol
	& Francis,	2011 [3, 18]												

Typical Physical and Mechanical Properties of Natural and Synthetic Fibers TABLE 1.3

8

can be approximated to the average of the presented range. The specific modulus values were obtained by the average stiffness and density, and the most attractive fibers from this point of view are curaua, flax, hemp, jute, pineapple leaf fiber (PALF), and ramie. Values in the same order of magnitude are found between wood and nonwood fibers. The most commonly used synthetic matrix materials used with natural fibers are PP, polyester, polyurethane, and epoxy. Most of the components made of natural fiber composites are fabricated by press-molding, even though a large range of processes are currently feasible [20]. Figure 1.3a compares the specific modulus of some natural fibers, and also *E*-glass fibers, showing in some of the cases a possibly higher performance of natural fibers, more specifically for ramie, PALF, kenaf, jute, hemp, flax, curaua, and bamboo. On the other hand, a much larger scatter can also be found for natural fibers, because of the bigger variations in stiffness and density. Figure 1.3b shows the evaluation of the cost per weight of some natural fibers and *E*-glass. In this scenario, all natural fibers behave better or at least identically to *E*-glass [3].

Mineral-based natural composites (i.e., asbestos) are naturally occurring mineral fibers (silicate-based minerals) or modified fibers that are processed from minerals. Asbestos is minerals that originate from nature in the form of fiber bundles. Mineral fibers are basically divided into six fibrous materials: amosite, crocidolite, tremolite, actinolite, anthophyllite, and chrysolite asbestos. The amosite asbestos, also known as brown or gray asbestos because of the presence of magnesium and iron, can be used as building materials, fire retardants, or thermal insulation products. Crocidolite or blue asbestos is not typically used commercially. Tremolite is formed by metamorphism of dolomite and quartz sediments. When heated, it is converted to diopsite and becomes toxic. Actinolite can be found in metamorphic rocks, and it is formed by the metamorphism of rocks with magnesium and dolomite shales. Chryso LITE or white asbestos are extremely soft silicate minerals of phyllosilicates. The fibers are extremely strong and long hollow cylinders. In 2006, asbestos mining reached 2.3 million tons [6]. At that time, Russia held the biggest extraction



**FIGURE 1.3** Comparison of the specific modulus of the most common natural fibers and *E*-glass fibers (a) and of the cost per weight of some natural fibers and *E*-glass (b). (From Dittenber, D.B., and GangaRao, H.V.S., *Composites: Part A*, 43, 1419–1429, 2012 [3].)

share of 40.2%, followed by China with 19.9%, Kazakhstan with 13.0%, Canada with 10.3%, and Brazil with 9.9%. Typical applications of asbestos fibers require properties such as inflammability, thermal, electrical, and sound insulation, adsorption capacity, wear and friction properties, and chemical inertness. Mineral fibers are usually combined with cement or woven to produce fabrics or mats.

Animal fibers are composed of proteins, for example, silk, wool, human hair, and feathers. Wool distinguishes itself from the other fibers by being crimped, elastic, and growing in staples [21]. In general, fibers taken from animals include sheep wool and goat, alpaca, or horse hair. Silk fibers come from natural proteins, and they can be woven into textile fabrics. The most widely known form is extracted from larvae cocoons of the mulberry silkworm. Silk fibers have a triangular prism-like structure, allowing silk fabrics to refract light in different angles and thus to produce different colors. Human hair is a filamentous biomaterial growing from follicles in the human dermis. It is primarily composed of a protein called keratin (approximately 95%). Feathers are highly complex integumentary structures that are produced in vertebrates, originate from follicles in the epidermis or outer skin layer that produce keratin proteins [22], and constitute the characteristic plumage of birds.

#### **1.3 BIOPOLYMERS**

The recent advances in biopolymers are triggered by the international interest to develop materials that are eco-friendly and do not depend on petroleum, because these resources are depleting and new solutions must be found. For instance, government institutions in countries such as the United States are establishing goals for production to account for a minimum amount of biomaterials. This is a challenge, because the property improvement of biopolymers is costly, and these can currently cost approximately 10 times more than common resins (PLA and starch-based resins are the cheapest ones). Until this cost problem is addressed, a possibility is the combination of natural fibers with a petroleum-based resin to make a composite that is not fully eco-friendly, but is partially disposable through incineration, and can eventually give good life cycle assessment (LCA) indicators. A partial solution to this problem would be milling the semi-biocomposites into small particles and their respective use as powder reinforcements in polymer mortars [23]. Currently, fabrication of natural fiber composites with natural polymers is feasible with biopolymers such as rubber, starch, soy protein, and PLA. Test results showed that soy protein generally behaves the best and rubber behaves the worst, due to issues of interfacial strength between the matrix and natural fibers. Starch polymers are easy to handle during the fabrication process, but they are sensitive to moisture. However, Mohanty et al. [24] showed that proper additives can partially eliminate this limitation and give the composite a good resistance to humidity and they also act as compatibilizers with jute fibers. Ochi [25] studied a biocomposite of hemp-reinforced starch, reporting an improvement of the tensile and flexure strength of the composites with increasing fiber content (until wt.70%). Values of 365 and 223 MPa were obtained for the tensile and flexure strengths, respectively. A comparison between hemp/starch and flax/starch composites was carried out by Nättinen et al. [26], showing that, for a fiber content of 10%, the mechanical behavior was similar (strength of 7.9 MPa, modulus of 0.68 GPa, and impact strength of 6.8 KJ/m<sup>2</sup> for the hemp composite, compared with 7.6, 0.60, and 12.8 for the flax composite). Some experiences with rubber seed oil–based polyurethane are also available in the literature, along with other types of oil (e.g., tung oil, peanut oil, walnut oil, or linseed oil [3]). Despite these options, there are two types of bio resins, soy-based and PLA, which offer the most cost and performance potential to replace petroleum-based polymers.

Biopolymers based on soy resins are one of the most researched nowadays, in the form of either soy protein concentrates or soy protein isolates, obtained by purification of defatted soy flour [3]. These polymers are characterized by reduced strength and sensitivity to degradation by humidity and, on account of this, they can be mixed with other polymers and thus produce soy-based matrices [27–28]. Actually, this is a very good option, although the individual characteristics and cost of soy matrices (and biopolymers in general) do not allow them to be a replacement for other solutions. Within this scope, some successful attempts obtained very interesting improvements in resistance to moisture and mechanical properties in soy matrices and their respective composites [29]. In the work by Mohanty et al. [30], natural composites made of soy bioplastic and short hemp fiber as reinforcement were tested, and the tensile modulus and strength of a 30% fiber reinforcement improved up to nine times the matrix strength. PLA is the other kind of bio resin that is already used in several applications, and it has a large industrial market. Processing of this material includes a few steps: raw material originating from dextrose or other renewable land materials, fermentation to convert into lactic acid, and polymerization. This material is completely biodegradable by a process of hydrolysis, forming lactic acid and, eventually, carbon monoxide. Thus, the use of PLA can reduce pollution, if replacing components made of harmful materials. A few years ago, PLA was very expensive, although some advances made it more affordable, in such a way that currently the bottleneck in its use is the supply capacity. Despite this fact, availability is expected to increase due to worldwide awareness to this material and creation of processing facilities. Porras and Maranon [31] experimentally characterized a full biocomposite made of bamboo fabrics as reinforcement and PLA as resin. An examination of the composite by scanning electron microscopy showed a strong bond between the fibers and resin, and mechanical testing revealed excellent energy absorption, which made these composites viable for use in some structural applications. Baghaei et al. [32] produced PLA reinforced with hemp composites, with hemp content between 10 and 45 wt.%. The natural fiber coupons were fabricated by compression molding and characterized regarding the mechanical performance, porosities, and thermal characteristics. The mechanical tests revealed tensile and flexural strengths that were approximately 2 and 3.3 times those of the neat PLA (considering fiber content of 45 wt.%). The impact characteristics improved approximately two times those of the PLA, but the tests showed that very small fiber contents actually reduced the impact properties.

#### 1.4 **BIOCOMPOSITES**

Biocomposites can be made of natural fibers with synthetic resins, natural resins with synthetic fibers, or both natural components. These materials have been used for decades, with application in aircrafts since the 1940s [9]. Nowadays, the use of these

materials extends to the construction industry, vehicle parts, household applications, and others. Natural fiber composites have a number of interesting characteristics, such as lower environmental impact,  $CO_2$  neutrality, and lower  $CO_2$  emissions than synthetic composites when composted or incinerated. In addition, they weigh less and are cheaper. Studies regarding their use as load-bearing components are also encouraging [33]. One of the differences among synthetic composites is the large property variation, because of the following reasons: dissimilar testing protocols, moisture conditions, physical properties, cell dimensions, chemical composition, microfibrillar angle, structure, defects, scatter in the mechanical properties of the fibers and matrix, and fiber-matrix interaction. The tendency for moisture absorption of natural fibers is also a major issue, as it highly influences the mechanical properties of the composites. There is a clear relationship between the moisture content of the natural fiber and the noncrystalline regions and voids. This issue was studied in detail in the work by Rowell [34]. The equilibrium moisture content of the fibers for a specific air humidity (i.e., the real moisture content of the fibers after exposure to a given amount of humidity) also has a major effect on the composite properties. For example, at the same air relative humidity of 65%, abaca fibers have a moisture content of around 15%, compared with 7% for flax. The transcrystallinity at the interface of natural fibers also affects their composite strength. Some surface treatments (stearation) can induce this effect. These issues were addressed by Zafeiropoulos et al. [35] for flax/isotactic PP with as is, dew-retted, duralin-treated, and stearic acid-treated fibers, showing a more than 100% improvement of the interfacial shear strength for the treated fibers.

In general, modification of the fiber surface can improve adhesion to the matrix. On the other hand, a weak interface reduces the efficiency of the stress transfer between the fibers and matrix, leading to premature damage in the composites and lower strength. The treatment methods are basically divided into physical and chemical methods. The former changes the structural and surface properties of the fibers and promotes the mechanical bonding of the matrix, although it does not change the chemical composition. Stretching, calendaring, and thermotreatment are examples of physical methods that are applicable to natural fibers. The corona treatment is an example of a physical process for surface activation that changes the surface energy of the fibers [36]. Another possibility is the plasma treatment, which induces different surface modifications depending on the gas, by modification of the surface energy and creation of surface cross-links [37]. Chemical treatments act by improving the adhesion with a third material between the fibers and matrix. This material promotes the compatibility between the fibers with hydrophilic behavior and the hydrophobic matrix. One chemical method is the silane treatment, which gives hydrophilic properties to the interface by using silanes that are used as primers to promote adhesion [38]. The largely used alkaline treatment or mercerization disrupts the hydrogen bonding in the fiber structure, thus increasing the fiber anchorage [39]. The acetylation treatment makes the surface of natural fibers more hydrophobic by coating the OH groups of fibers [40]. Another possibility by which the strength of natural fiber composites could be improved is the maleated coupling. The maleic anhydride not only acts on the surface but also improves the interfacial bonding [41]. Finally, the enzyme treatment is environmentally friendly and cost-effective, and it acts by promoting reactions on the fiber surface that improve adhesion [42]. Table 1.4 gives, as

## TABLE 1.4Mechanical Properties of Hemp-Reinforced Natural Fiber Composites andDifferent Resins

	Tensile s	trength (MPa)	Young's		
Matrix	Resin	Composite	Resin	Composite	Source
PLA	47.5–51	75–85 (30% hemp fibers)	3.5–5	8–11 (30% hemp fibers)	[43]
PP	22.8-35.46	28.1–45.33 (40% hemp fibers)	1.07-1.1	3.5–3.72 (40% hemp fibers)	[44]
Polystyrene	34.1±0.68	40.4±0.65 (22.5% hemp fibers)	—	_	[45]
Epoxy	25	60±5 (30% hemp fibers)	0.7	3.6±0.4 (30% hemp fibers)	[46]
Polyester	12.5±2.5	60±5 (35% hemp fibers)	1.1±0.2	1.75±0.5 (35% hemp fibers)	[47]
Unsaturated polyester	25±5	65±2.5 (30% hemp fibers)	1.5±1	8.75±1.25 (30% hemp fibers)	[48]

an example, the mechanical properties of natural fiber composites with hemp fibers and different matrices [43–48]. The main conclusion to be drawn here is that, notwithstanding the matrix material, the addition of the natural fibers highly improves the strength and stiffness of the resulting material. It is also visible that, due to the chemical reactions between the hydroxyl groups on the fiber surface and the thermoplastic resin, composites with thermoplastic resins excel those with thermoset resins. Table 1.5 compares the mechanical properties of different natural fiber composites as a function of the fiber loading [49–52]. Overall, the introduction of the reinforcement in the polymer significantly improves the Young's modulus as the wt.% content of the fibers is increased. The tensile strength of the composites increases as well, except for the results of the palm leaf fibers/PP composite. In general, the strength improvements are more modest. The impact strength increased for the ramie fibers/ PP composite, whereas there is no available data for the other composite systems.

A merit comparison between glass and natural fiber composites (on average) is shown in Figure 1.4. Price can be similar between both composites, but glass composites excel in mechanical performance while having a significant recyclability penalty. Thus, if natural fibers are to replace glass fibers for a given application, this will have to occur in a way in which the mechanical properties are safeguarded. Natural fiber composites do not match glass composites in terms of mechanical properties, as opposed to what occurs with specific properties, especially the stiffness. Therefore, natural fibers are more suitable for providing stiffness in applications that are neither under moisture nor under any adverse environmental conditions. Addressing the issues of moisture and performance of natural fiber composites is possible, as previously mentioned, but this will require new approaches. Actually, in the same manner that metal parts or structures cannot be replaced by synthetic fibers without any design modifications, because of

#### TABLE 1.5 Mechanical Properties of Natural Fiber Composites with Different Resin and Matrix Combinations

	Fiber Content (wt.%)	Young's Modulus (MPa)	Tensile Strength (MPa)	Impact Strength (kJ/m²)	Source
Ramie fibers/PP	0	1300	35	2.8	[49]
	10	1400	42	3.0	
	20	1600	51	4.2	
	30	2250	66	4.7	
Palm truck fibers/high-density PE	0	475	17.5	_	[50]
	20	750	17	_	
	30	975	18	_	
	40	1500	20	_	
Palm leaf fibers/PP	0	800	27.5	_	[51]
	7	700	23.5	_	
	15	650	21	_	
	28	675	17	_	
Pineapple leaf fibers/polycarbonate	0	1100	67.5	_	[52]
	5	1150	67	_	
	10	1450	66	_	
	20	2000	71	—	



FIGURE 1.4 Merit comparison of glass and natural fiber composites (on average).

the intrinsic differences in properties and fabrication processes, the replacement of glass by natural fiber composites also requires new designs and solutions to obtain the best performance. The immediate applications of natural fiber composites are restricted to limited performance parts, where these materials can really excel because of their bio characteristics and, eventually, cost advantage (or at least nondisadvantage). Examples are selected components for the automotive industry with low strength requirements, such as panels and trims, which also improve the bio credentials of vehicles. Apart from this, any component whose performance demands are within the reach of natural fiber composites can potentially be fabricated in these materials. Examples are wood parts, since the consumer demands for water and moisture absorption for wood components are usually low. Replacement of unreinforced plastics is also a chance for development, and the use of low-cost plant fillers is ongoing in the electronics industry. The replacement of the fillers by fibers can give significant performance improvements. At the moment, there are still many challenges to be overcome for natural fiber composites to be able to be applied in all current glass applications (to be discussed in Section 1.8). Nonetheless, the eventual success of such replacement surely relies on the ongoing and future research and the development of new designs that favor the mechanical properties of these materials.

#### 1.5 BENEFITS AND APPLICATIONS OF NATURAL FIBER COMPOSITES

In past decades, natural fibers and natural fiber composites received attention from researchers in several industries, such as in civil construction, automotives, and biomedicine [53], mostly based on three factors: reduction of costs, weight reduction, and sustainability. The mechanical behaviour of lignocellulosic fibers (non-wood or plant) and their composites, either with biological or synthetic materials as matrix, have been studied extensively by the scientific community in parallel to industrial use in vehicles and construction. Actually, natural fibers of flax, hemp, sisal, or jute can replace glass or other kinds of synthetic fibers in epoxy, polyester, PVC, PE, or PP matrices, with the following benefits:

- Lower costs because of reduced cost of raw materials, smaller cycle times, lower weights, and reduction in the fuel consumption (vehicle parts)
- Identical mechanical properties of glass-reinforced parts, with fabrication advantages such as smaller tool wear, good sound insulation, and geometrical stability
- Eco-friendliness, renewability of the raw materials, recyclability, no toxicity, and CO<sub>2</sub> neutrality.

Many literature examples exist on the use of natural fiber composites in automotive applications, mainly for interior vehicle parts [1,54–55], with either thermoplastic or thermoset matrices. The selected materials for these applications should meet requirements of minimum strength and strain to failure, impact and flexural properties, sound insulation, fire resistance, processing characteristics (dwell time and temperature), odor, dimensional stability, and energy absorption under crash conditions. Bledzki and Gassan [16] reported an application of jute, coffee bag wastes and PP bags in trim parts of Brazilian trucks after recycling. Saxena et al. [54] concluded that using natural fiber composites in vehicle applications as trim parts, panels, shelves, and brake shoes can give an advantage of 10% in weight, fabrication process energy savings of 80%, and an overall reduction of 10% in the cost of parts. Moreover, around 6000 natural fiber composite parts could be introduced in vehicles with this potential advantage. In locomotives, components such as the gear casing, doors and side panels, interior furnishing and seating, luggage racks, berths, chair backings, modular toilets, and roof panels in natural fiber composites can also bring benefits to weight, cost, corrosion resistance, and weight-reduction-driven fuel consumption savings. For civil engineering applications, natural fiber composites from bast fibers are, in general, the best, whereas flax gives the best balance between strength and stiffness to cost and weight. Jute-reinforced composites are very common, but their strength and stiffness does not match flax. Because of the specific stiffness advantages compared with glass composites, natural fiber composites are an excellent solution for reinforcement of existing infrastructures. In general, in the development of natural fiber composites, which are biodegradable, the replacement of synthetic materials such as glass-reinforced composites without compromising their distinctive characteristics is currently a big challenge and will continue to be so.

Natural animal fiber composites are scarcely used in industrial or other applications. Animal fibers, such as wool or spider silk, are made of proteins and find useful applications in bioengineering and medicine. Wool is the most used natural fiber, although it suffers from low fracture resistance, which is its biggest limitation. A major application of wool fibers is the fabrication of rock wool fabrics or panels that are used in the construction industry on account of their good fire resistance and sound absorption. Silk fibers are characterized by their stability even when exposed to varying environmental conditions; they have a low weight, and their composites are very tough and impact resistant. Some applications of these fibers were reported in automotive, aerospace, and sport equipment industries [6]. Feathers meet application in cement-bonded feather boards, which are resistant to decay and termite attacks due to the keratin. These feather boards can be employed in paneling, ceilings, and insulation, although not as structural components. Animal feather composites can compete with conventional materials with regard to a few specific applications.

There is also a history of application of mineral fibers or asbestos in corrugated panels (e.g., roofing compounds), gaskets, pipeline wrapping, sheets, rods, shaped moldings, and thermal and/or electrical insulation. Fabrics of mineral fibers also find application in parts that involve friction, such as brake or clutch pads, because they are durable to friction and are heat and oil resistant. Mineral fibers can be fabricated with biosynthetic matrices to produce a large variety of products. Chrysotile with rubber matrix finds application in packings, gaskets, and heavy-duty insulation parts as compressed boards. More specific applications include reinforcement agents in coatings and adhesives. Mineral fibers have a significant limitation, related to health hazards, including lung, eye, and skin diseases, which causes numerous deaths under working conditions. Because of this and environmental concerns, these fibers are being less used.

#### 1.6 LIFE CYCLE ASSESSMENT OF NATURAL FIBER COMPOSITES

Natural fiber composites have become feasible alternatives to glass composites since the 1990s, and some of them are highly attractive for their use in vehicle and leisure parts. Traditionally, thermoset matrices are generally used, but thermoplastics such as PP have recently attracted attention because of processing and recyclability issues [56]. As previously referred to, natural fiber composites are considered to have a number of environmental advantages. Since the environmental benefit of these materials is a driver for the increased use of these materials, a comprehensive evaluation of the environmental impact of natural fiber composites for all product stages, between raw material extraction and end-of-life disposal, should be carried out. According to the definition by Duflou et al. [57], the LCA analysis balances the environmental costs and benefits of different materials for a given application, while considering the different phases of the product. More specifically, the LCA methodology gives an assessment of the sustainability of materials that quantifies the effect on the environment of the raw materials and their extraction or production, energy consumption for fabrication, impact during life, waste generation and recycling, and incineration or disposal after their lifespan (Figure 1.5). This comparison should be made on an equivalent functional basis, since natural fiber composites are usually lighter, even compared with synthetic composites.

A potential difference between synthetic and natural fiber composites that stands out immediately is the energy requirements to fabricate the fibers. Glass fibers are produced at around 1550°C, because of the high melting temperature of glass, which makes this a major issue. Regarding the matrix, energy consumption can be in the form of mineral oil extraction, separation, refinement, and polymerization. When considering bio materials as a matrix, for example, PLA, PHA, or modified starch, different results can be expected. Several LCA studies are available in the literature, namely comparisons with synthetic composites, for which natural fiber composites typically aim to substitute. In the study by Mohanty et al. [24], the authors stated that the required energy to fabricate natural fibers, by weight, is between 20% and 25% that of synthetic fibers. Different investigators used LCA analyses to conclude that, in the whole fabrication process, natural fiber composites only spend approximately 60% of the energy used by synthetic composites [58]. The presented value considers the effect of material extraction or harvest, further processing, transportation, and composite fabrication. Other studies showed that the energy needed to fabricate a natural fiber fabric is only 30%–40% of that required to fabricate a glass mat [59]. Wötzel et al. [60] presented a comparative LCA study of a panel for an Audi A3, considering the original acrylonitrile butadiene styrene (ABS) copolymer part and a hemp fiber (66 wt.%)/epoxy composite. The study is somehow incomplete, since it does not account for some important aspects, such as the component use and endof-life disposal, but it models the inputs, energy use, and pollution until the part fabrication. The authors concluded that the natural composite uses 45% less energy



**FIGURE 1.5** Typical phases of a composite part, with impact on energy and emissions.

and emissions are lower. Nonetheless, emissions of some polluting substances such as nitrates and phosphates are higher because of their fertilizer application in hemp crops, although this is not very significant. Schmidt and Beyer [61] focused on an insulation component of a Ford vehicle, originally made of ethylene propylene diene copolymer (EPDM), PP, and glass fibers. This component was weighted against a tentative new design by replacing hemp fibers (30 wt.%) with glass. This was a more in-depth study, as it covered all previously mentioned stages of the product. The natural fiber component showed significant advantages regarding the cumulative energy demand (CED) (savings of 88.9 MJ), CO<sub>2</sub> pollution (8.18 kg), and generic emissions. Corbiere-Nicollier et al. [62] evaluated transport pallets made of either the original glass-reinforced PP or china reed fiber-reinforced PP. For an equivalent performance, the natural fiber component required 53 wt.% of fibers, as compared with 42 wt.% for the original counterpart. The entire life cycle was assessed, ending with incineration in both cases. Overall, the natural composite showed significant advantages regarding the environmental impact, except nitrate emissions.

Duflou et al. [57] suggested three indicators to study LCA: (1) the CED, which is a global environmental factor and a major driver; (2) greenhouse-gas (GHG) emissions, because of the climate change and global warming implications, measured in CO<sub>2</sub> equivalents or CO<sub>2</sub>e; and (3) aggregate environmental impact score, usually expressed in milli-ecopoints (mPT). By dividing the LCA analysis in the production, use, and end-of-life phases, Duflou et al. [57] proposed a comprehensive comparison between different natural fiber and glass-reinforced composites. Table 1.6 evaluates the indicators CED, GHG, and mPT for production of different matrices, fibers, and composite fabrication [63–66]. The matrix advantage is mainly with regard to the GHG and ecopoints, whereas CED reductions are much smaller, except for linseed oil monomer (ELO). Major reductions in CED and GHG can be found for natural fibers with respect to glass fibers, which are their main competitor, whereas mPT data are inconclusive. The values for the different fabrication processes are mainly indicative, as these quite vary between material choices. The LCA analysis for the use phase has some specificities that cannot be neglected for the sake of a realistic analysis (e.g., different lifespan of materials and weight). In vehicles, weight reduction has a double impact, because it reduces both fuel consumption and emissions, and because of this the major share of fiber-reinforced materials (approx. 44%) goes to transportation systems [67]. Shifting vehicle parts from glass to natural fiber composites brings weight benefits between 22% and 27% [68]. Table 1.7 provides a comparison of CED and GHG during the use life for vehicles and other parts in traditional materials and their equal functionality equivalents in synthetic and natural fiber composites [62,69-72]. Carbon fiber composites provide a massive saving related to steel, aluminum, and even glass composites, because of the weight savings for the same function. Replacement of glass with natural fibers is also recommended. A limitation of this analysis is that it considers an equal lifespan between synthetic and natural fiber composites, since analyses of the use life of these materials are not available, and lifespan is highly dependent on the moisture level in the composite. Table 1.8 is related to the CED and GHG impact of different end-of-life disposal strategies (SMC refers to Sheet Molding Compound glass composites, and GMT

#### TABLE 1.6 Comparison of CED, GHG, and mPT for Different Materials and Fabrication Processes

Material	CED (MJ/kg)	GHG (kg of CO <sub>2</sub> e/kg)	Ecopoints (mPT/kg)
Matrix			
Epoxy	76–137	4.7-8.1	734
Unsaturated polyester (PES)	62.8-78	2.3	644
PP	73.4	2.0	276
Modified starch (Mater-BI®)	54.8	1.3	275
PLA (Ingeo 2009 <sup>TM</sup> )	67.8	1.3	312
PHA (generic)	59-107	0.7-4.4	—
Linseed oil monomer (ELO)	19	1.2	_
Reinforcement			
Carbon fiber (generic)	286-704	22.4-31	833
Carbon nanofiber (CNF)	654-1807	70–92	—
Glass fiber (generic)	45	2.6	264
Flax fiber	9.6-12.4	0.4	350
Hemp fiber	6.8-13.2	1.6	—
Jute fiber	3.8-8.0	1.3-1.9	—
Sugarcane bagasse	11.7	—	_
Composite Fabrication Proce	ess		
Sheet molding compound	3.5-3.8	_	13
Resin transfer molding	12.8	_	46
Pultrusion	3.1	_	11
Autoclave	21.9	_	_
Injection molding	21.1-29.9	0.5-1.2	126

Source: Suzuki, T., and Takahashi, J., Prediction of energy intensity of carbon fiber reinforced plastics for mass-produced passenger cars. Proceedings of JISSE-9, Tokyo, Japan, 29 November–11 December, 2005; Patel, M., Energy, 28, 721–740, 2007; Boustead, I., Eco-profiles of the European Plastics Industry. Association of Polymer Manufacturers in Europe, Brussels, Belgium, 2005; Ecolizer 2.0. 2003–2011. Openbare Vlaamse Afvalstoffenmaatschappij, Mechelen, Belgium [63–66].

refers to Glass-Mat Reinforced thermoplastics) [70,72–75]. The recycling technique depends on the material, and it can include mechanical or thermal methods for composites and remelting and recasting for metals. Accumulation in landfills is also an option for composites, actually the most common a few years ago, but it is not ecoefficient because it does not allow recovering the embodied energy of the materials. Moreover, composites still need treatments to reduce the environmental impact of the wastes. In general, fiber composites are incinerated, which allows the embodied energy to be recovered. Glass composites can equally be incinerated, but glass fibers

#### TABLE 1.7 Comparison of CED and GHG during Their Use Life for Vehicles and Other Parts

			CED	
Vehicle Part	Original Material	Composite Replacement	Change (GJ/Part)	GHG Change (kg of CO <sub>2</sub> e/Part)
Propeller shaft	Steel	Carbon and glass fiber/epoxy	-3.7	-227
	Aluminum		-2.5	-158
Car closure panel	Steel	Carbon fiber/epoxy	-26.9	-2096
	Aluminum		-6.8	-531
	Glass fiber/ poly(ethylene terephthalate - PET)		-13.1	-1023
Car door	Steel	Glass fiber/PP	-2.0	-150
	Aluminum		+0.8	+67
Car interior	Talc/PP	Bagasse/PP	-19.3	-206
Transport pallet	Glass fiber/PP	China reed/PP	-0.6 to -2.3	_

Source: Corbiere-Nicollier, T. et al., Resources, Conservation and Recycling, 33, 267–287, 2001; Song, Y.S. et al. Composites: Part A, 40, 1257–1265, 2009; Puri, P. et al., International Journal of Life Cycle Assessment, 14, 420–428, 2009; Luz, S.M. et al., Resources, Conservation and Recycling, 54, 1135–1144, 2010; Schexnayder, S.M. et al., Environmental Evaluation of New Generation Vehicles and Vehicle Components. Report ORNL/TM-2001–266, Oak Ridge National Laboratory, Oak Ridge, 2001 [62,69–72].

are incombustible, which leads to an energy consumption of approximately 1.7 MJ per glass fiber weight [76]. Natural fiber composites have the logical advantage of being combustible and, thus, helping the process. Recycling can essentially be carried out in four ways: (1) mechanical recycling, (2) chemical treatment, (3) pyrolysis, and (4) fluidized-bed processing [57]. Natural fiber composites, in particular, can be recycled by using many techniques, without a significant loss of mechanical properties. In the work by Bourmand and Baley [77], a sisal/PP composite showed reductions of only 10.1% and 17.2% in tensile modulus and tensile strength, respectively, after seven cycles, in opposition to a glass/PP composite that showed 40.1% and 52.5% property losses, respectively. Biodegradation is another scenario that is used to dispose biocomposites [78]. Table 1.9 compares natural fiber composites against the original counterparts in vehicle applications using the CED change during the production, use, and end-of-life phases [60,71]. Detailed information about the components and use scenario is given in the source references. For the three parts, the end-of-life strategy is incineration (with energy recovery). Natural fibers consistently provide less energy in incineration on account of the lower required mass for the same application. On the other hand, natural fiber composites behave a lot better in the production and use phases, resulting in a lower accumulation of CED during the entire life.

In conclusion, there is a so large number of variables that are necessary to include in the study and so many different materials and applications that a conclusive generic study that these materials are actually more eco performing in all applications actually does not exist. Nevertheless, in comparison to general materials, natural composites spend approximately 20 MJ less of energy for 1 kg of material and

#### TABLE 1.8

		Landfill	I	Recycling	Recovery	
	CED (MJ/kg)	GHG (kg of CO <sub>2</sub> e/kg)	CED (MJ/kg)	GHG (kg of CO <sub>2</sub> e/kg)	CED (MJ/kg)	GHG (kg of CO <sub>2</sub> e/kg)
SMC		_	7	0.4	-7.5	0.9
GMT	0.09	0-0.02	11	0.9	-25.2	1.9
Carbon fiber composite	0.11	0.02	10–15	—	-31.7 to -34	3.2–3.4
Natural fiber composite	—	_	—	_	−12 to −34	2.3–2.9
Steel	—	—	11.7– 19.2	0.5–1.2	—	
Aluminum	_	_	2.4-5.0	0.3-0.6	_	_

#### CED and GHG Impact of Different End-of-Life Disposal Strategies

Source: Puri, P. et al., International Journal of Life Cycle Assessment, 14, 420–428, 2009; Schexnayder, S.M., Environmental Evaluation of New Generation Vehicles and Vehicle Components. Report ORNL/TM-2001–266, Oak Ridge National Laboratory, Oak Ridge, 2001; Leterrier, Y., Comprehensive Composite Materials, 1073–1102, Oxford: Pergamon, 2000; Hedlund-Aström, A., Model for end of life treatment of polymer composite materials. Royal Institute of Technology, Stockholm, Sweden, 2005; Duflou, J.R. et al., CIRP Annals: Manufacturing Technology, 58, 9–12, 2005 [70,72–75].

#### TABLE 1.9

#### CED Comparison during the Production, Use, and End-of-Life Phases between Original and Natural Fiber Replacement Components

			CED C	CED Change (MJ/Part)			
Part	Original	Natural Fiber Composite Replacement	Production	Use	End-of-Life	Source	
Car interior	Talc/PP	Bagasse/PP	-222	-19313	+62.3	[71]	
Side panel of small vehicles	ABS	Hemp/epoxy	-59	-71	+27	[60]	
Side panel of large vehicles	ABS	Hemp/epoxy	-59	-118	+27	[60]	

prevent the release of 1 kg or more of  $CO_2$  for 1 kg of material into the atmosphere [79]. Thus, despite not being fully conclusive, the discussed and other available studies highly reinforce the need to develop these materials.

#### 1.7 POTENTIAL OF NATURAL FIBER COMPOSITES AND DRIVERS FOR CHANGE

Recent advances in natural fiber composite technology enabled the development of materials that exhibited attractive performance and sustainability. To date, these materials have been applied mostly in vehicle products and some construction applications, with the previously mentioned advantages. If these new materials are to be generalized to other sectors of industry, as, for example, household products or goods, there are basic inherent properties that they must accomplish: performance for the desired function, usability, reliability, and durability. The discussion is divided into three main areas of actuation: mechanical properties, environment, and cost-effectiveness.

#### 1.7.1 MECHANICAL PROPERTIES

Currently, natural fiber-related technology is being improved to provide better mechanical characteristics of the bio-based components. With this large effort, it will be possible for biocomposites to exploit other fields of application that are not currently in use. But for this to happen, the knowledge of the materials, fabrication processes, and design methods must reach a much higher degree of confidence. These issues, together with proper standardization for these materials, can give them a distinctive edge over conventional materials. At the moment, large efforts are being made to make biocomposites a solution for load-bearing parts in construction. In fact, some authors tested the use of cellular plates and beams as structural parts made from hemp, jute, and flax fibers in polyester resin [80] in the construction industry (house building). The components were experimentally tested, and the results showed that the cellular arrangement of the natural fibers can improve the composite mechanical properties just enough to compete with other engineering materials (e.g., glass fiber composites or common construction materials) and make them viable to load-bearing applications. This line of research is to be followed in the future to make civil construction a strong application of natural fiber composites. Applications in other sectors of industry rely on additional improvements. However, based on the current state of the art, some limitations of these materials still need to be addressed for them to be considered competitive against synthetic composites. It was previously mentioned that natural composites are a cost-effective solution compared with other materials, but it is also true that if a 100% biological and recyclable solution is needed, costs increase and this also needs further research efforts. Moreover, ecological superiority over synthetic composites is not yet fully true because of the fabrication techniques that consume large amounts of energy. Another feature to be improved is the resistance to moisture and temperature, and here a long path exists, knowing that there are limits to the materials themselves. For example, currently it is possible to make a part fully biodegradable with the proper choice of a bio matrix, although biodegradation would be high. Significant improvements in some key aspects of bio materials such as large nonlinearity/relaxation, long-term performance, and small impact resistance can occur by improving the processing of the fibers and composite fabrication. It can be concluded that new frontiers will emerge for these materials when the following characteristics are met with a significant degree of comparison with the other materials: durability, dimensional stability, environment resistance, and fire resistance [7].

One of the material-related fields that has endured major enhancements recently is nanotechnology. Common natural fibers contain a small amount of nanocrystalline cellulose. The artificial fabrication of natural fibers with this structure could produce fibers with 10% of the strength of carbon nantubes, but with a cost approximately 1000× less [7]. Research on this field mainly uses wood pulp to produce nanocellulose, but other non-lignocellulosic products can be used with this purpose: hemp [81], wheat [82], or flax [83]. Some authors [33] obtained cellulose nanofibers with a mixed chemical/mechanical technique and combined them with a starch polymer. Preparation of the nanofibers enabled cleaning the fiber surface of hemicelluloses, lignin, and pectin, and also the defibrillation of nanofibers from the initial fiber bundles. It is also possible to fabricate microfibrillated cellulose from wheat and soy by cryocrushing, disintegration, and fibrillation, producing fibers with a diameter between 30 and 40 nm [84]. Many other works used similar techniques to produce these nanomaterials from soy, root crops, wood, seaweed, cotton, hemp, cereals, and sea squirts, among others. Composites made with these nanofibers experience a major improvement in their tensile strength and stiffness. Nanotechnology can also be used differently to improve natural fiber composites by application of coatings, diminish the effects of biodegradation, or increase the fire resistance of the materials. With the recent efforts under way, it is a matter of time until nanoconcepts give natural fiber composites the performance, durability, value, service life, and utility that makes them more competitive, while maintaining their ecologic features.

#### 1.7.2 Environment

Natural fiber composites fit in the concepts of sustainable economy, since synthetic materials are replaced by bio-based and renewable ones. These materials also have the potential to be more cost-effective for identical structural characteristics, and there is the opportunity to produce or grow the fiber plants in controlled facilities or farms. Compared with synthetic resins and fibers (or even conventional materials) that these materials can potentially replace, the carbon footprint will be tremendously reduced. Synthetic fibers and resins have posed difficulties with regard to their disposal for decades, accounting for approximately 20% of the total landfill space, depending on the country. This is a strong motivation for the replacement of synthetic composites, since landfill capacity is scarce and overcrowded. In terms of saving the environment, it is more urgent to replace the matrix than the fibers by natural equivalents, since petroleum-based resins take hundreds of years to degrade [85]. Recycling is an opportunity, although recycled petroleum-based resins lose some characteristics by incorporation of external substances, which affects the adhesion between fibers and matrix. On the other hand, PLA can be reconverted practically without affecting