



Natural fibre composites

Materials, processes
and properties

Edited by Alma Hodzic and Robert Shanks

Natural fibre composites

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Part I

Natural fibre reinforcements

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Wood fibres as reinforcements in natural fibre composites: structure, properties, processing and applications

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Abstract: Wood fibres are the most abundantly used cellulose fibre. They have been extensively used in the modern composite industry due to their specific characteristics. This chapter systematically describes the structure, properties, processing and applications of wood fibres as reinforcements in natural fibre composites: first, the nature and behaviour of wood fibres and the developed technologies for the modification of wood fibres to enhance physical and mechanical properties (e.g. surface functionality and tensile strength) are investigated; the matrices and processing techniques for the development of wood fibre composites are then discussed; and finally, the properties and applications of wood fibre composites in industrial sectors are presented.

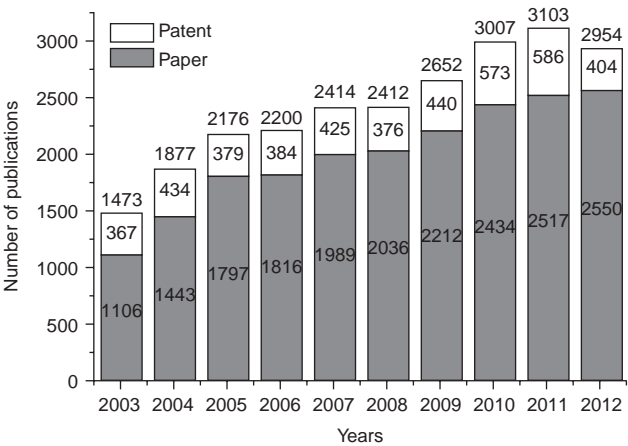
Key words: wood fibres, structure, physical and mechanical properties, modification, process technique, wood fibre composites.

1.1 Introduction

Natural fibres are rich in cellulose, abundantly available and easy to handle and process. However, the application of natural fibres in composites came to a near-halt from the 1940s due to the low price and the steadily rising performance of technical and standard plastics.^{1,2} In the 1990s, critical discussion about the preservation of natural resources and recycling had led to a renewed interest in natural renewable materials. With the recent increase in environmental awareness, exploiting natural fibres has raised great interest and become of importance. Among these natural fibres, wood fibres are the most abundantly used cellulose fibre having extensive use in the fibreboard, pulp and paper, and many other industrial sectors. Over 55% of paper and paperboard production³ and 80% of natural fibre reinforced composites⁴ are made from wood fibres. The use of wood composite materials dates from centuries ago.⁵ In ancient China, wood, bamboo and straw were used to reinforce



1.1 Yuchang Lou in Fujian, China: a wood–clay composite building that is over 600 years old.



1.2 Number of publications on wood fibres and products.

clay for making walls, and many natural fibre-reinforced clay buildings are still in use, e.g. the ‘Tulou’, which dates from 1200 years ago, one of the oldest wood–clay buildings, can be observed in the south of China (Fig. 1.1).

To date, numerous publications have reported the applications of natural fibres, e.g. pulp,^{6,7} ethanol⁸ and composite.⁹ Among these reports, more

than 85% focus on wood fibres. In addition, the growing importance of wood fibres can be evidenced from the increasing number of publications in the past 10 years (Fig. 1.2). It should be noted that these publications derive from the Google scholar database with the key word of wood fibre, and it is found that more than 75% of the publications are from journals or conferences, and about 70% of these reports focus on wood fibre composites.

The development of nanotechnology (NT) and biotechnology in the past 10 years has pushed the research and development of wood fibres a step further, and enlarged the role of wood fibres in many industrial sectors, such as pulp and paper, and material industries.

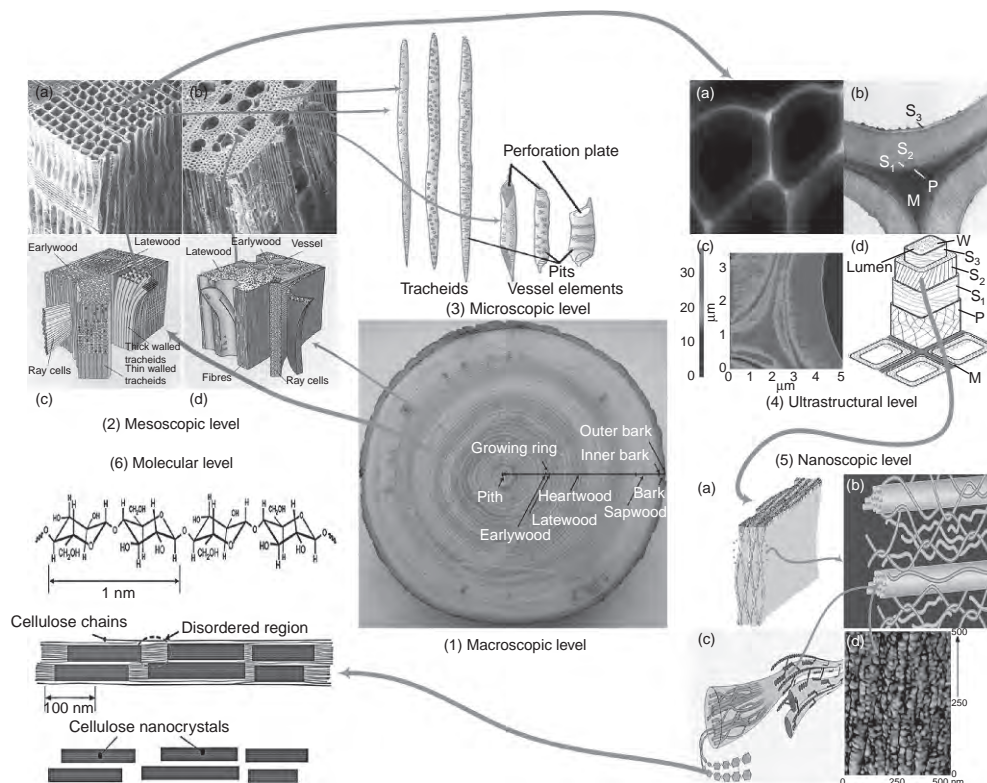
1.2 Wood fibres: nature and behaviour

1.2.1 Structure of wood fibres

Wood fibres consist of both live and dead cells in the wood, depending on the location and the age of tree from which they are extracted. The hierarchical structure of wood fibres gives this fibrous material excellent performance properties, e.g. high strength to weight ratio. Wood fibres can be obtained from timber by chemical, mechanical, biological processes, and many combined processes.

At the macroscopic level (normally 0.1–1 m), wood fibres mainly exist within the layer of xylem in the wood¹⁰ (Fig. 1.3(1)). The dark strip in the centre of the stem is the pith, which represents the tissues formed during the first year of growth. The inner part of the xylem layer consists of dark coloured heartwood. The lighter coloured outer part is sapwood, which conducts water from the roots to the foliage of the tree. Both inner and outer parts are organized with many concentric growth rings (annual increments), each of which is distinguished by earlywood, composed of large thin-walled cells produced during the spring when water is usually abundant, and the denser latewood, composed of small cells with thick walls (Fig. 1.3(2c)¹¹). In addition, the inner bark layer comprises the tissues outside the vascular cambium, including secondary phloem, which transports the nutrients from photosynthesis in the leaves to the rest of the tree, cork cambium (cork-producing cells), and cork cells. The outer bark, composed of dead tissue, protects the inner region from injury, disease and desiccation. At the mesoscopic level (normally 1–10 mm), wood fibres form a continuum of cellular material.¹²

At the microscopic level (normally 0.01–6 mm), two kinds of wood cells with different hierarchical structures, namely tracheids (in softwoods and hardwoods) and vessels (only in hardwoods), can be easily distinguished¹³ (Fig. 1.3(3)), and the dimensions of both wood fibres are shown in Table 1.1.^{14–18}



1.3 Wood fibre from macroscopic to molecular levels: (1) macroscopic level; (2) mesoscopic level: 3D schematic of (a) softwood and (b) hardwood, scanning electron microscope (SEM) of (c) softwood and hardwood; (3) microscopic level; (4) ultrastructural level: (a) Raman image, (b) transmission electron microscope (TEM), (c) atomic force microscope (AFM) and (d) model of wood cell; (5) nanoscopic level: (a) schematic drawing of the cellulose aggregate structure in wood cell, (b) a schematic of cellulose fibrils laminated with hemicellulose and lignin, (c) AFM of a transverse section of the secondary wall of wood cell and (d) structure of cellulose in a wood cell wall; (6) molecular level.

Table 1.1 Dimensions of typical softwood and hardwood fibres

Types of fibres	Length (mm)	Width (μm)	Aspect ratio
Softwood	2–6	20–40	50–200
Hardwood	1–2	10–50	28–86

Tracheids constitute over 90% of the volume of most softwood¹⁹ and 50% of the volume of hardwood.¹⁵ Their average length is usually between 2 and 6 mm,^{14,16,18} and their width is between 20 and 40 μm ,^{14,17} with a length to width ratio (aspect ratio) often in excess of 50–200. In hardwood, the length of tracheids, which is only 1–2 mm,^{14,15} is considerably shorter than that of softwood tracheids, and the width is between 10 and 50 μm ^{14,15}, with a narrow aspect ratio of 28:86.¹⁴ In addition to tracheids, hardwoods have wider cells, namely vessel elements, which vary considerably in size and shape.²⁰ They are a series of broad and articulated cells (around 100 μm), which are long (many centimetres) and their function is to channel sap in almost straight lines. In some wood species, they may account for up to 50%–60% of the volumetric composition, but usually less than 10% by weight.¹⁴ The wide vessel elements of the early wood are found to be 13%–47% shorter than those of the late wood.²¹

At the ultrastructural level (normally 1–25 μm), the wood fibres are built up of four layers (Fig. 1.3(4)).^{22–25} These are middle lamella (M), primary wall (P), secondary wall (S), including the outer layer of the secondary wall (S_1), the middle layer of the secondary wall (S_2), the inner layer of the secondary wall (S_3), and the warty layer (W).^{26,27} The middle lamella is located between the cells. This layer is highly rich in lignin; the concentration of lignin in this layer is about 70%–80%,²⁸ which is about twice that in secondary wall. The high concentration of lignin can cement the cells together very well, but in the processing of wood fibres, separation of the lignin remaining on the fibre surface can result in a decrease of inter-fibre bonding. The primary cell wall is a thin layer (0.1–0.2 μm) which surrounds the protoplast during cell division and subsequent enlargement.¹⁴ It contains a randomly and loosely organized network of cellulose microfibrils. Due to the occurrence of pectin and protein, the properties of the primary wall layer differ from those of the secondary; in this layer strong interactions exist among the lignin, protein and pectin, as well as among the cellulose and hemicellulose. This obvious feature has a major influence on the separation of fibres. The secondary cell wall (1.2–5.4 μm) contains much more ordered microfibrils than the primary cell wall. It comprises a series of layers, namely S_1 , S_2 and S_3 . The warty layer (W) is comparable in thickness (0.1–0.3 μm) to the primary wall, and consists of four to six lamellae which spiral in opposite directions around the longitudinal axis

of the tracheid.¹⁴ The outer secondary cell wall (S_1) has a crossed fibrillar structure. Although the S_1 layer has a large microfibril angle (MFA), about $50\text{--}70^\circ$,²⁹ this layer is considered to play an important role in determining the transverse mechanical properties and surface properties of fibres^{30–32} as well as pulp fibre properties.³³ The main bulk of the secondary wall is contained in the middle secondary cell wall (S_2 , $1\text{--}5\text{ }\mu\text{m}$). The microfibrils in this layer spiral steeply about the axial direction at an angle of around $5\text{--}30^\circ$,^{29,34} and have a pronounced influence on the properties of fibres. The S_2 layer is the thickest cell wall layer and controls the strength of the entire fibre. The inner secondary wall (S_3 , $0.1\text{ }\mu\text{m}$), sometimes also known as the tertiary wall,³⁵ is at the lumen boundary and forms a barrier between the lumen and the rest of the cell wall. Compared with the other layers in the secondary wall, the S_3 layer contains the highest concentration of lignin, about 53%.³⁶ In this thin layer the microfibrils form a flat helix. The microfibrils in the S_3 layer are oriented almost perpendicularly to the microfibrils in the S_2 layer with MFA between 50° and 90° .²⁹ The innermost portion of the cell wall consists of the so-called warty layer, probably formed from protoplasmic debris. All softwoods have this segment in their cell wall; however, not all hardwoods do.²⁹

At the nanoscopic level (Fig. 1.3(5)),^{37–39} wood fibres have an important influence on the final performance of timbers. These influences include chemical reactions and physical effects. The wood fibres are built up by cellulose microfibrils ($10\text{--}25\text{ nm}$ ^{26,40}), hemicelluloses and lignins due to the formation of lignin–carbohydrate complex (LCC) by covalent bonds.⁴¹ Most of the microfibrils are not parallel to the cell axis and can form a particular angle, which is known as the MFA. The MFA was found to be a critical factor in determining the physical (e.g. shrinkage^{42,43}) and mechanical properties (e.g. stiffness,⁴⁴ and tensile strength⁴⁵) of wood fibres.

From the molecular point of view (Fig. 1.3(6))⁴⁶, the main chemical components of wood fibres are cellulose, hemicellulose and lignin. As shown in Table 1.2,^{47,48} the dominant component in wood fibres is cellulose. The

Table 1.2 Chemical compositions of hardwood and softwood fibres

Types of wood fibres	Cellulose (%)	Hemicellulose (%)	Lignin (%)	Extractives (%)
Original wood fibres				
Softwood	40–45	25–30	26–34	0–5
Hardwood	45–50	21–35	22–30	0–10
TMP wood fibres	37.07 ± 0.6	29.2 ± 0.1	13.8 ± 0.7	0.8 ± 0.6
Unbleached wood fibres				
Softwood	69.0 ± 2.5	22.0 ± 0.7	8.8 ± 1.8	0.2 ± 0.1
Hardwood	78 ± 0.5	19.3 ± 0.1	2.4 ± 0.4	0.3 ± 0.2
Bleached wood fibres				
Softwood	79.2 ± 0.2	20.0 ± 0.1	0.8 ± 0.1	0 ± 0
Hardwood	78 ± 0.2	20.3 ± 0.1	1.3 ± 0.1	0.5 ± 0.1

TMP, thermomechanical pulps.

cellulose of wood fibres is similar to other natural fibres, in that it consists of a linear chain of several hundred to over 10 000 $\beta(1\rightarrow4)$ linked D-glucose units⁴⁹ laid down in microfibrils in which there is extensive hydrogen bonding between cellulose chains, producing a strong crystalline structure in a crystalline region.^{50,51} Combined with the amorphous region, the cellulose microfibrils aggregate into larger microfibrils.⁵¹ The hydrogen bonds in the cellulose not only have a strong influence on the physical properties of the cellulose (e.g. solubility, hydroxyl reactivity) but also play an important role in its mechanical properties.⁵⁰

1.2.2 Physical and mechanical properties of wood fibres

The surface property is one of the key properties of wood fibres; it can affect the interfacial adhesion of resin on the surface of fibres and the mechanical properties of fibre-based composite. This property is influenced by fibre morphology, chemical composition,⁵² extractive chemicals and processing conditions.⁵³ Table 1.3 shows the surface properties of wood fibres in comparison with other natural fibres. Due to the high polar character of the surface, the fibres are less compatible with non-polar resin. Therefore, the combination of the inherent polar and hydrophilic features of wood fibres and the non-polar characteristics of resins gives rise to difficulties in compounding these materials, resulting in inefficient stress transfer of its composites under load. The use of different kinds of physical (i.e. corona discharge) and chemical surface treatment methods (i.e. coupling agents such as silanes) leads to changes in the surface structure of the fibres as well as to changes of surface properties.

The mechanical properties of wood fibres are of great importance for their use in the paper^{67,68} and composite industries.⁶⁹ The mechanical properties of materials can be characterized from two methods, namely, macroscopic tests (e.g. tensile test) and indentation tests. The macroscopic tests focus on measuring the mechanical performance of the whole sample, while the indentation tests focus on measuring a local area of the sample.⁷⁰ In

Table 1.3 Surface properties of natural fibres

Fibres	Surface area (m ² /g)	γ^d (mJ/m ²)	$(\zeta_0 - \zeta_{\infty})/\zeta_0$ (mV)	ζ_{plateau} (mV)	References
Flax	0.31 ~ 0.79	23.85	0.88 ~ 0.95	-1.1 ~ -0.21	54–57
Hemp	0.75	31.6	0.91	-0.1	55, 57, 58
Sisal	1.63	32.90–48.35	0.76 ~ 0.88	-1.7 ~ -0.4	55–57, 59
Coir	0.48	45.05	0.22	-4.6 ~ -3.8	56, 59, 60
Softwood	0.97	31	–	-18.6 ± 5.0	61–63
Hardwood	1.34	32–47	–	-7.3	64–66

γ^d : dispersive surface energy; ζ_0 : ζ -potential initial value; ζ_{∞} : ζ -potential final value; ζ_{plateau} : ζ -potential plateau value.

the macroscopic tests, the parameters of mechanical properties generally include such items as tensile strength and modulus, elongation, compressive strength and modulus, impact strength, and flexible strength and modulus. Researchers traditionally use the elongation, tensile strength and Young's modulus^{1,71} to evaluate the mechanical performance of wood fibres.

As aforementioned, the mechanical performance of wood fibres is influenced by their structure; in addition, the mechanical performance is influenced by the growing parameters, e.g. area of growth, climate, and the age of the plant.^{72,73} Wood fibres generally display higher mechanical performance compared with other natural fibres (Table 1.4). However, due to wood source, growth conditions, and chemical and mechanical treatments, the strength of wood fibres varies considerably, which is one of the main drawbacks for all natural products. The range between minimum and maximum characteristic values of wood fibres is noticeably wider than that of synthetic fibres although the wood fibres display a good Weibull modulus (Table 1.5) which describes the variability of the failure strength.

The first report about the mechanical properties of wood fibres did not appear until the end of the 1950s.⁹¹ The development of spectroscopic (especially X-ray diffraction (XRD) in 1912 and Raman spectroscopy in 1923) and microscopic (especially atomic force microscopy (AFM) in 1986) technologies enabled the characterization of mechanical properties on micro- or nano-scales for heterogeneous polymer or composites. These developed technologies have enlarged understanding of the mechanical properties of wood fibre (wood fibre itself is a polymeric composite) and the structure–property relationship,⁹² and optimized the utilization of wood fibres as reinforcements in composites.²⁴ Nanoindentation and AFM techniques have been employed to investigate the micro- or nano-mechanical properties of

Table 1.4 Mechanical properties of natural fibres

Fibres	Density (g/cm ³)	Elongation (%)	Tensile strength (MPa)	Young's modulus (GPa)	References
Flax	1.5	1.2–3.2	345–2000	15–80	74–76
Hemp	1.48	1.6	550–900	26–80	72, 77, 78
Sisal	1.5	3.0–7.0	468–700	9.4–22	79
Coir	1.2	17–47	175	4.0–6.0	1,71
Softwood	1.5	–	600–1020	18–40	80, 81
Hardwood	1.2	–	–	37.9	82
E-glass	2.5	2.5	2000–3500	70	1
S-glass	2.5	2.8	4570	86	1
Aramid	1.4	3.3–3.7	3000–3150	63.0–67.0	1

Table 1.5 Weibull modulus of natural fibres

Natural fibre	Weibull modulus	Gauge length (mm)	References
Flax	2.6	10	83
Hemp	2.86	10	84
Sisal	4.6	10	85
Coir	3.1	8	86
Wood	3–5	–	87
E-glass	6.61	10	88
S-glass	20.5	–	89
Aramid	10.4	5	90

Table 1.6 Effect of wood species on the mechanical properties of wood fibres

Type of fibres	MFA (°)	Elastic modulus (GPa)	References
Softwood			
Spruce	–	13.49 (CV 43.00%, earlywood)	93
Scots pine	5	21.00 (CV 16.00%, latewood)	93
(pulp)	–	12.2 ± 1.6	94
Hardwood			
Oak	3 ± 3	18.27 ± 1.74 (earlywood:	
Eucalyptus	–	latewood = 1:1)	94
(pulp)		9.10 ± 1.60	

wood fibres since the first report about the cell wall mechanics of softwood by using nanoindentation in 1997.⁹³

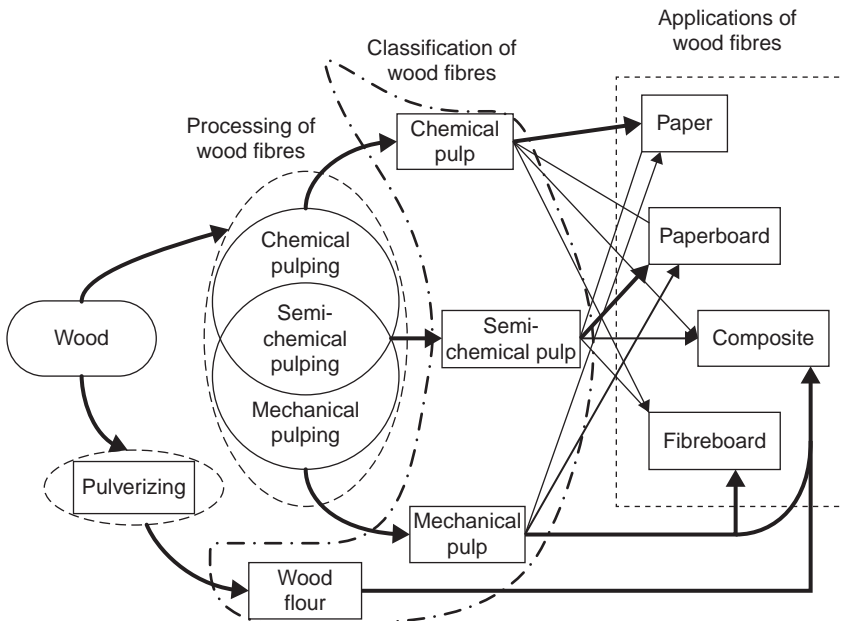
By using these new techniques, researchers have revealed various novel properties of wood fibres. These findings support the selection of wood fibres as composites, and the understanding of the interaction mechanism between wood fibres and the matrices. For example: (i) the identification of different properties among wood species and across growing stages: at cell wall level, it seems that the elastic modulus of hardwood fibres is similar to that of softwood fibres; however, earlywood has a lower elastic modulus than latewood (Table 1.6); (ii) the anisotropic properties of wood fibres: Jäger *et al.*^{96,97} employed the Vlassak model to evaluate the relationship between indentation modulus, indentation direction and elastic material constants of spruce cell wall material $M^{\text{pred}}(E_t, E_l, G_{lt}, \nu_{lt}, \nu_{ll}, \delta_t)$, and then using an error minimization procedure to analyse the values of the elastic material constants – the values for the longitudinal elastic modulus, transverse modulus and shear modulus are reported as 26.3, 4.5 and 4.8 GPa, respectively; (iii) the interfacial compatibility between S_2 and S_3 layers: by using a nanoindentation-AFM technique the interfacial compatibility in the cell

wall of spruce was further investigated⁹⁸ and it has been found that the S_3 layer has a less polar character than the S_2 layer; hence polyurethane (PUR) was found to have a better adhesion to the S_3 layer and poorer adhesion to the S_2 layer compared with urea formaldehyde (UF). It is proposed that differences in the polarity of the adhesives used and in the surface chemistry of the two cell wall surfaces examined account for the observed trends.

In addition, nanoindentation and AFM have been widely used to reveal much more detail about wood fibres, e.g. cell wall lignification,⁹⁹ melamine modification,¹⁰⁰ stiffness and hardness of wood fibres,¹⁰¹ and conformability of wet wood fibres.¹⁰²

1.2.3 Processing of wood fibres

The separation of wood fibres includes two methods (Fig. 1.4): a pulping process and a pulverizing process. Pulverizing is the process by which the wood is reduced into small particles (180–425 μm). It is the main step for the production of wood flour, which is mainly used as filler in plastics. For dry mechanical processing, the final products typically have low aspect ratios (only 1–5). These low aspect ratios allow wood flour to be more easily metered and fed than individual wood fibres, which tend to bridge. However, the low aspect ratio limits the reinforcing ability. Pulping is the process by



1.4 Processing and applications of wood fibre.

which the macroscopic structure of raw wood is reduced to a fibrous mass. It is achieved by rupturing bonds within the wood structure. It can be accomplished chemically, mechanically or by some combination of these treatments. These treatments are (i) chemical,^{103–105} (ii) mechanical^{106,107} and (iii) semi-chemical, which combines (i) and (ii) to separate wood fibres.^{108–110}

The main commercial chemical treatment technique is the sulphate or kraft process; an acid sulfite process is also used. The chemical process involves the use of chemicals to degrade and dissolve lignin from the wood cell walls, releasing high cellulose content fibres. Chemical pulping processes yield pulps with higher strength compared with mechanical processes. However, these processes are low yield (40–55%)^{111,112} (Table 1.7) and are very capital-intensive.¹¹¹ Products from the chemical treatment process (chemical pulp) are always used for paper (e.g. tissue), paperboard, etc.

Stone groundwood (SGW), pressure groundwood (PGW), refiner mechanical pulps (RMP) and thermomechanical pulps (TMP) are the main products of wet mechanical treatments. Wet mechanical treatment involves the use of mechanical force to separate the wood fibres. Mechanical defibration of wood and chips results in only small material losses and the gross composition of the resulting pulps differ only slightly from that of the original. However, the fibre structure is somewhat damaged. Mechanical treatment under wet conditions can obtain higher yield (Table 1.7), but these processes are electrical energy-intensive and produce paper with lower strength, higher pitch content, and higher colour reversion rate compared with chemical processes. Mechanically produced pulp has a higher proportion of broken cell fragments (called ‘fines’) among the fibres. The mechanical pulps can be used for paper (printing paper), paperboard, composite and fibreboard.

The semi-chemical techniques normally involve pretreatment of wood chips with a chemical method. There are several types of semi-chemical pulps in production, e.g. chemimechanical pulps (CMP), chemithermo-mechanical pulps (CTMP) and neutral sulfite semi-chemical (NSSC) pulps. NSSC is the most common product, made primarily from hardwood species and noted for its exceptional stiffness and high rigidity. The yield of semi-chemical pulping is 58.7–95%¹²⁹ (Table 1.7). Its primary use is for the production of paperboard as well as printing papers, greaseproof papers and bond papers. The semi-chemical pulps are still used for composite, but very much less for fibreboard.

1.2.4 Applications of wood fibres

As a result of a growing awareness of the interconnectivity of global environmental factors, the principles of sustainability, industrial ecology and ecoefficiency, and also green chemistry and engineering, are being integrated into the

Table 1.7 Pulp yield and relative strength achieved using various pulping methods

Classification	Process	Yield (%)	Strength			References
			Tensile index (Nm/g)	Tear index (mN·m ² /g)	Burst index (kPa·m ² /g)	
Chemical treatment	Kraft pulping	40–50	92.0–98.5	8.6	6.8–7.3	113, 114
	Sulfite pulping	45–55	85–132.7	7.4–12.2	4.43–6.42	115, 116
Mechanical treatment	Soda pulping	45–55	69.9–83.6	3.2–9.2	4.2–7.34	117, 118
	Stone groundwood (SGW) pulping	90–98.5	28.2	2.2	0.86	119, 120
	Pressure groundwood (PGW) pulping	95.5	18.4	2.8	0.90	119, 120
	RMP	90–97.5	28.1	2.8	0.66	119–121
Semi-chemical treatment	TMP	91–95	27.6	3.2	1.10	120, 122, 123
	Chemimechanical pulping (CMP)	80–90	49–63	5.45–5.5	3.11	115, 120, 124
	Chemithermomechanical pulping (CTMP)	80–95	51.8	6.4	2.4	120, 124, 127
	NSSC	58.7–80	30.90–35.57	3.73–4.08	1.38–1.60	126–128

Table 1.8 Production of commercially important fibre sources during 2002–2011

Year	Production of wood fibres (t)	Production of cotton lint (t)	Production of flax (t)	Production of hemp (t)	Production of jute (t)	Production of sisal (t)	Fibre share of wood (%)
2002	167 257 547	18 885 577	781 762	78 110	2 863 037	284 355	87.96
2003	170 046 697	19 467 665	786 350	69 824	2 798 567	305 867	87.89
2004	174 785 477	24 530 832	1 013 105	72 793	2 562 784	325 679	85.98
2005	173 985 135	24 476 010	1 008 656	78 664	2 766 596	330 895	85.86
2006	175 826 129	24 449 953	661 958	113 270	2 870 460	366 497	86.07
2007	180 993 713	25 066 925	541 087	77 079	2 823 671	370 519	86.24
2008	177 469 786	22 487 098	527 851	71 336	2 691 315	370 786	87.16
2009	160 557 039	20 898 428	382 538	79 721	3 045 089	393 953	86.62
2010	170 871 722	23 717 040	315 583	81 048	2 828 533	363 558	86.22
2011	173 309 240	26 102 935	315 084	81 964	2 861 996	411 864	85.34

development of the next generation of materials, products and processes.¹³⁰ In 2003, the UK government established highly ambitious long-term goals relating to climate change, with the objective of moving towards a ‘low carbon economy’ and a target to cut carbon dioxide (CO₂) emissions by 60% by mid-twenty-first century. The White Paper states that this should be achieved without detriment to the UK’s competitiveness or security. Then, to effectively reduce CO₂ emissions while keeping economic growth, different countries have begun to search for new development paths, among which low-carbon development has become widely advocated.¹³¹ These actions have accelerated research in natural fibres for application in many industrial sectors.

Wood fibres are the most important source among the natural fibres, e.g. as shown in Table 1.8 (these data are arranged based on reference 132), showing a share of wood fibres of over 85%. Wood fibres are primarily used for the paper and paperboard industry (about 80.5%), representing over 55% of total paper and paperboard production.³ Some 17.03% of wood fibres are used in composites, with wood-fibre-based composites making up over 80% of natural fibre reinforced composites.⁴

1.3 Modification of wood fibres for composites

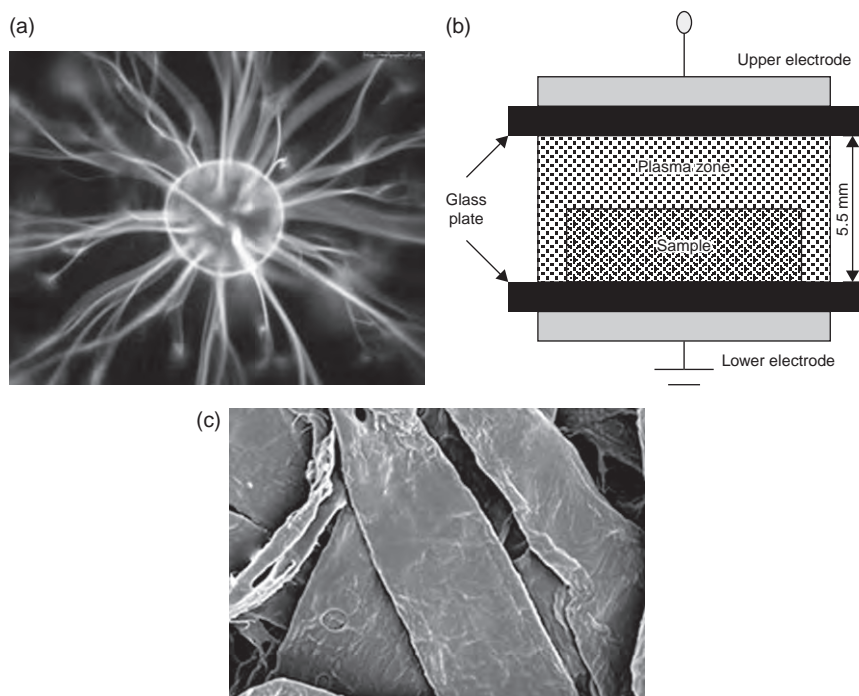
The use of wood fibre to make low cost and eco-friendly composite materials is a subject of great importance. However, certain drawbacks of natural fibres (e.g. higher polar and hydrophilic) cause natural fibres to be poorly compatible with polymers, which results in the loss of mechanical properties upon atmospheric moisture adsorption.⁶⁵ Compared with glass fibres, natural fibres show lower mechanical properties. In order to improve the mechanical properties and the interfacial property of natural fibres, various modifications of the natural fibres have been investigated. These modifications are of three types: physical, chemical and NT.

1.3.1 Physical modification

Physical modification has always been carried out by using instruments to change the structural and surface properties of the fibres, with the aim of increasing the strength of fibres and the interfacial compatibility between wood fibre and matrices. Traditional methods involve thermotreatment,^{133,134} calendering^{135,136} and stretching of these, thermotreatment is the most useful to modify natural fibres. When the fibres are subjected to heat treatment above the glass transition temperature of lignin, it is postulated that the lignin will be softened and migrate to the fibre surface. Kraft lignin has a glass transition temperature of 142°C.¹³⁴ Lignin begins to degrade at around 214°C; hence, heating the fibres to 200°C would be expected to cause some softening.¹³⁸ Thermal treatments can increase the crystallinity, dimensional stability, hydrophobicity of lignocellulosic fibres.

Thermal treatment is an efficient modification for forest products. The final properties of the products may significantly depend on the modification of hemicelluloses. This treatment can improve the moisture resistance of wood-based panels.^{139,140} Hydrothermal treatment to modify wood flour can increase the storage modulus of poly(lactic acid) (PLA)–wood flour composites by up to 55.65% without any other chemical reagents.¹⁴¹ Saturated steam under pressure at various temperatures above 100°C results in a decrease in the thickness swelling of the panels while mechanical properties, flexural properties, internal bond strength and screw withdrawal resistance, decrease.¹⁴² Medium density fibreboard (MDF) panels made from thermally-treated wood fibres at 180°C for 30 min appear to be a practical choice for achieving a low thickness swelling of MDF products.

Surface modification by discharge treatment,^{143,144} such as low-temperature plasma, sputtering and corona discharge, is of great interest in relation to the improvement in functional properties of natural fibres. This technique was found to be effective for the improvement of the compatibility between hydrophilic fibres and a hydrophobic matrix. Scientists in some industrialized countries, such as France, Japan and the United States, have carried out surface treatment of different fibres with various plasma techniques since the 1960s. To date, scientists in most countries have studied this topic to develop their own industrial projects. Plasma technology has been widely used as an effective method for surface modification of natural fibres such as flax,^{145,146} sisal,¹⁴⁷ and keratin.¹⁴⁸ Plasma treatment (Fig. 1.5)^{149–151} mainly causes chemical implantation, etching, polymerization, free radical formation and crystallization, whereas the sputter etching brings about mainly physical changes, such as surface roughness, and this leads to increase in adhesion.¹⁴⁵



1.5 Schematic of plasma treatment: (a) plasma lamp; (b) plasma system and (c) wood fibre after discharge treatment.

The processing of wood fibres can result in various chemical compositions in the wood fibres, as shown in Table 1.2. The discharge treatment¹⁵² (diffuse coplanar surface barrier discharge (DCSBD) plasma) of wood fibres can result in polar carbonyl groups ($\text{C}=\text{O}$) and a considerable increase of free surface energy to reduce the water uptake of wood. The discharge treatment (corona discharge) of wood fibres obtained by mechanical processing was reported to produce 2.4 carboxyl and 10.9 carbonyl functions per hundred C9 units of lignin.¹⁵³ The cold Ar plasma treatment can result in the generation of higher phenoxy radical concentration in CTMP,¹⁵⁴ the concentration of which was four times that of TMP. By using heteronuclear single quantum coherence (2D-HSQC) spectroscopy and nuclear magnetic resonance spectroscopy of carbon (^{13}C -NMR), it was found that the generation of phenoxy radicals can lead to cross-linkages of lignin monomeric units and formation of new inter-monomeric C–C and C–O bonds. In both fibres, the chemical structure of lignin was heavily modified by plasma treatment and the CTMP forms much more radicals than chemical pulp.¹⁵⁵

In addition, a variety of surface modifications can be achieved depending on the type of discharge. Carlsson *et al.*^{156,157} studied the effects of hydrogen

and oxygen plasma treatments on wood fibres and found that the hydrogen plasma treatment reduced the hydroxyl groups and the water absorption of the wood fibres. By contrast, the oxygen plasma treatment displayed an improvement of water wettability.

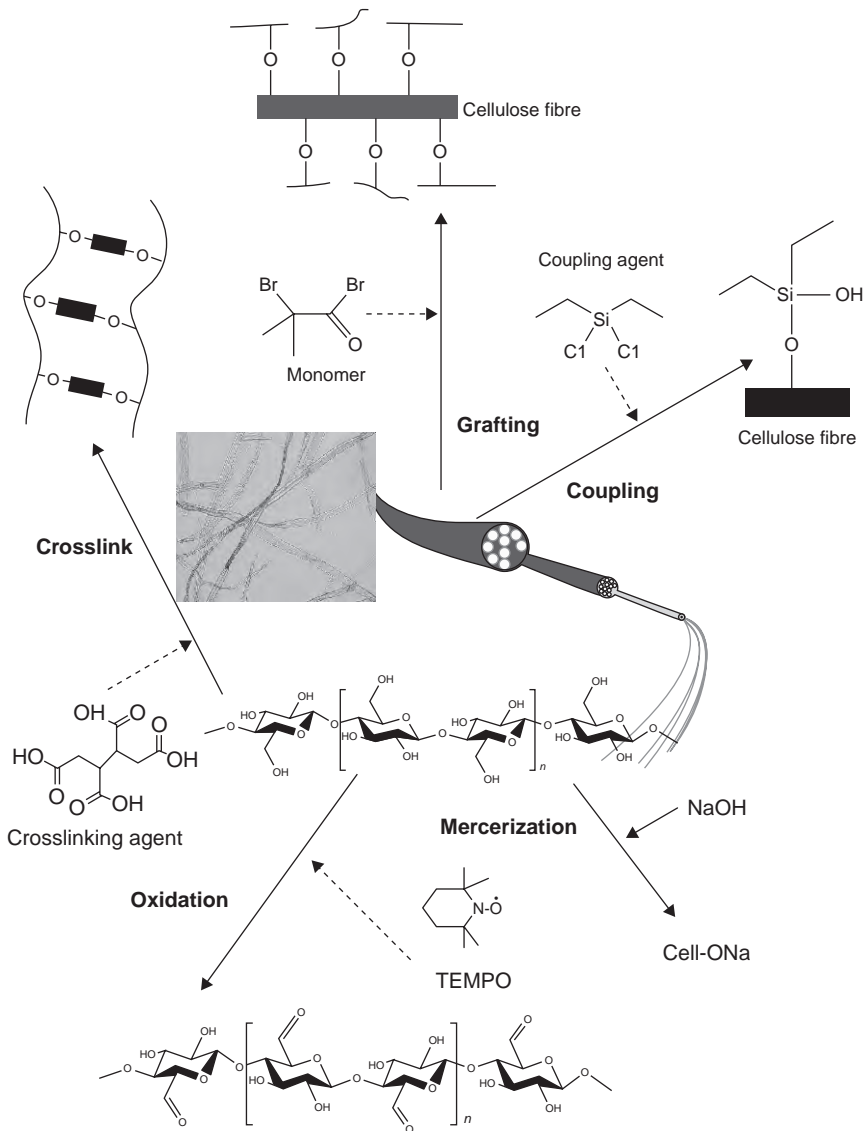
1.3.2 Chemical modification

Chemical modifications utilize chemical agents to modify the surface of fibres or the whole fibre. They can modify the structure of wood fibres or introduce new hydrophobic functional groups into the surface of wood fibres to reduce the hydrophobicity of fibres. The modification can be classified into five methods: mercerization, oxidation, crosslink, grafting and coupling agent treatment (Fig. 1.6).

Mercerization

Mercerization is an old method of cellulose fibre modification, which is an alkaline treatment method for cellulose fibres. The process was devised in 1844 by John Mercer of Great Harwood, Lancashire, England, who treated cotton fibre with sodium hydroxide.¹⁵⁸ This treatment caused the fibres to swell; about 25% of hydrogen bonds are broken during the swelling process in the post-treatment (drying).¹⁵⁹ These bonds will re-bond and the consequent effects of the re-bond have been reported in the literature: including (i) decreasing the spiral angle of the microfibrils and increasing the molecular direction;¹ (ii) producing fibre fibrillation, i.e. axial splitting of the elementary fibres (or microfibrils that constitute the elementary fibre).^{160–162} This process leads to a decrease in fibre diameter, increasing the aspect ratio and the effective surface area available for wetting by a matrix in a composite; there is also an increase in fibre density as a consequence of the collapse of its cellular structure; and (iii) changing the fine structure of the native cellulose I to cellulose II.^{163–166} These changes may result in an improvement in fibre strength and hence stronger composite materials.^{161,167,168}

It was reported that after immersion in alkali for 48 h, the globular pultrusion presented in the untreated fibre disappeared, leading to the formation of a larger number of voids. Systematic investigations¹⁵⁹ have already revealed three important phenomena of cellulose swelling in aqueous alkali, i.e. (i) the passing of the swelling value through a maximum, depending on lye concentration; (ii) a qualitatively similar, but quantitatively different, behaviour of all the alkali hydroxides in aqueous solution from LiOH to CsOH on interaction with cellulose in an aqueous medium; and (iii) a phase transition within the region of crystalline order above a lye (alkaline) concentration of 12–15% due to a so-called intracrystalline swelling caused by inclusion of NaOH and H₂O into the crystallites.



1.6 Main chemical treatments and modification mechanism of natural fibre.

The mercerization process increases the number of hydroxyl groups on the wood fibres surface, which in turn favours water absorption,¹⁶⁹ therefore, wood fibres with mercerization should not be suitable for hydrophobic matrices. It was reported that the alkaline treated wood fibres incorporated in polypropylene (PP) can induce the hexagonal phase of iPP and the mechanical

performance of the wood-PP composite was increased.¹⁷⁰ Alberto *et al.*¹⁷¹ modified wood fibres with (i) cold water, (ii) hot water and (iii) hot water with sodium hydroxide (1% concentration), and found that the third treatment could increase the compatibility factor significantly for the fibres from *Jambire* and *Wimbe* with a compatibility factor of 84.77% and 83.77% respectively. The main mechanism of the reinforcement by alkaline treatment may be due to the degradation of hemicellulose and amorphous content, as the alkaline treatment products are more effective than the polar extractive treatment.¹⁷²

Oxidation

Oxidation modification can be achieved under mild conditions. In this case carboxyl groups, aldehyde groups and ketone groups can be introduced into the cellulose chains by the selective oxidation of primary or secondary hydroxyl groups in the chains. In 1938, Yackel *et al.*¹⁷³ firstly employed NO₂ as oxidant to oxidate cellulose selectively. After that, various primary^{174–181} and secondary^{185–186} oxidative systems have been reported. Recently, due to the excellent selective oxidation, TEMPO-NaBr-NaClO and TEMPO-NaClO-NaClO₂ oxidative systems^{181,187–197} have received much attention around the world. Potthast *et al.*¹⁹⁸ investigated the new functional groups on the surface of hemp fibres which were introduced by the TEMPO oxidation system. Results showed that the content of hydroxyl groups was influenced by the concentration of oxidant and the treatment time. Matsui *et al.*^{199,200} investigated the influence of ozone oxidation pretreatment on the graft copolymerization of methyl methacrylate on the surface of hemp fibres and found that, with the increase of oxidation time, hydroperoxide (HPO) increased from 0 mol/cell. molecule to 160 mol/ cell. molecule, and the CI of the fibres decreased from 69.7% to 68.3%, but the degree of grafting increased significantly from 14% to 129%.

Crosslinking

Multifunctional compounds which have more than two functional groups have always been used as crosslink agents to crosslink the inter-chains of cellulose by reacting with the hydroxyl groups. The crosslink modification of cellulosic fibres has always been carried out by etherification²⁰¹ and esterification.²⁰² The crosslinking of cellulose has been found important for commercial application in textile finishing of cellulose-based fabrics with end-use properties, e.g. wrinkle resistance, permanent press and easy care properties. Wood fibres are the main contributor to the hygro-expansion (which is one of the drawbacks of wood fibres) of wood fibre-based composites. The crosslinking modification can reduce the transverse coefficient of hygro-expansion of the wood fibres²⁰³ and result in the improvement of environment and dimension stability of wood fibre-based composite.^{77,204}

Grafting

Chemical modification through graft copolymerization is an effective method that improves the compatibility of wood and other natural fibres with hydrophobic matrices. The technique involves the grafting of various monomers onto the surface of cellulosic fibres.^{205,206} The reaction is usually initiated by free radicals of cellulose molecules. The cellulose is exposed to high-energy ionizing radiation. After treatment with the selected ions (transition metal ions), oxidative reagents (as initiating agents), initiate free radicals on cellulose.²⁰⁷ The radical sites initiate the grafting of alkyl acrylates (such as methyl, ethyl, butyl and propyl), vinyl monomer (such as methyl methacrylate and acrylonitrile) to the cellulosic surface. Maleic anhydride (MA) grafting treatment has been reported to function efficiently for natural fibre-based composite. Among the grafting treatments, MA grafting is the main method^{208–226} for the modification of wood fibres. The type and concentration of MA can influence the mechanical performance of the composite. It has been reported that MD411D displayed a better performance at 2% concentrations.²¹⁰ Compared with the other natural fibres (e.g. cotton), wood fibre-based composites display better mechanical performance under low fibre loading (<10%).²¹⁶ The length of fibres has a positive effect on the tensile modulus and modulus of elasticity (MOE).²²⁰ In addition, the other additives (e.g. compatibilizer^{210,212}) also affect the mechanical performance of composites. Other grafting methods, e.g. methyl methacrylate (MMA) grafting,^{227–230} styrene grafting,^{231–233} cetyl alcohol²³⁴ grafting and polymer grafting^{235,236} have also been reported.

Coupling

In wood fibre composite industries, the coupling modification is the most important method. Coupling agents can be defined as the substances that are used in small quantities to treat a surface so that bonding occurs between filler and matrix. Coupling agents can be subdivided into two broad categories: bonding agents and surfactants (also known as surface active agents). At present, over 40 coupling agents have been used in the production and research of natural fibre composites.²³⁷ The most popular treatments include the use of silanes^{238–251} and isocyanates.^{236,248,252,253}

1.3.3 Nanotechnology (NT) modification

Nanotechnology (NT) is defined by the US National Nanotechnology Initiative as the understanding, manipulation and control of matter at dimensions around 1–100 nm. Currently, most major governments around the world are investing heavily in NT²⁵⁴ and many see it as the fuel for the next Industrial Revolution. With the large amount of fundamental research

under government funding today, NT has applications across nearly all economic sectors and allows the development of new critical enabling science with broad commercial potential, such as nano-structured materials, nanoscale-based manufacturing processes and nano-electronics. It has been demonstrated in recent years that NT can be used to modify natural fibres to introduce new functions onto the surface of fibres and enhance the performance of the final fibre-based products. This modification has been used in the textile^{255,256} and paper industries²⁵⁷ successfully. It is believed that the application of NT to the modification of wood fibres offers high economic potential for the development of fibre-based industry.

Various approaches have been developed to immobilize nanoparticles on the surface of wood and other natural fibres, among which, layer-by-layer (LbL) deposition^{258–261} and the sol-gel process^{262–265} are the main approaches that have commonly been employed by researchers. Different kinds of nanoparticles (e.g. AgNPs,^{258,266–269} TiO₂,^{269,270} SiO₂,^{271–273} ZnO^{274,275}) have been developed to impart multifunctional properties (e.g. anti-bacteria, UV resistant, anti-wrinkle finishing, water repellent) to fibres.

A novel way of combining biological technology with NT was first reported by Juntaro *et al.*²⁷⁶ in 2007. This green technique firstly employed bacteria *Gluconacetobacter xylinus* strain BPR 2001 to treat natural fibres (hemp and sisal), then fabricated the bacterial cellulose on the surface of natural fibres. These modified natural fibres were then incorporated into the renewable polymers cellulose acetate butyrate (CAB) and poly-L-lactic acid (PLLA). It was found that the modified sisal PLLA composites showed an increase in parallel strength of 44% and an increase in off-axis composite strength of 68%.

1.4 Matrices (binders) of wood fibre composites

Wood-based composites can be classified as follows, based on the form of their structural components:

- (i) Fibrous composites (composed of fibres in a matrix), such as fibreboard, MDF and wood plastic composite (WPC);
- (ii) Laminar composites (composed of layers of materials), such as plywood, woven and non-woven textile composite;
- (iii) Particulate composites (composed of particles in a matrix), such as particle boards (PB).

In wood fibre composites, wood fibres are the dispersed phase. The matrix may be inorganic compound, natural polymer or synthetic resin. The composites' shape, surface appearance, environmental tolerance and overall durability are dominated by the matrix, while the fibrous reinforcement

carries most of the structural load, thus providing macroscopic stiffness and strength.²⁷⁷

1.4.1 Inorganic compounds

The inorganic compounds used for wood fibre composites are cement, clay and lime. In ancient Egypt and China, clay had first been used to make walls. In the early to mid-1940s, natural fibre–cement composite was first investigated in Australia.²⁷⁸

Wood fibre–inorganic compound composites are one of the most successful applications of wood fibres in the composite industry. They have been widely used as corrugated or flat roofing materials, cladding panels and water containers, in a large number of building and agriculture applications. One of the drawbacks associated with wood fibres in inorganic application is their dimensional instability when the composites are subjected to changing relative humidity (RH) atmosphere. This instability is promoted by: (i) the water sensitivity of cellulose fibres, (ii) the effects of carbonation, high alkali content of the cement matrix and the generation of incompatible stresses. However, the addition of wood fibre can bring three benefits: (i) improving the toughness, tensile strength and the cracking deformation of the composite; (ii) increasing the solids retention and (iii) capturing CO₂ and locking it up in buildings.

1.4.2 Natural polymers

Natural polymer matrices include natural polymers and synthetic natural polymers. The natural polymers typically include starch and rubber. The synthetic natural polymers include PLA and poly(hydroxy butyrate) (PHB), etc. which are made from natural products (e.g. sugar, vegetable oil) by various chemical polymerization methods.

The use of starch to produce biodegradable plastics began in the 1970s. Starch-based products have prevailed all over the market, from packaging to the toy industry.²⁷⁹

Starch has been considered as one of the most promising materials for biodegradable plastics because of its natural abundance and low cost. However, low water resistance and poor mechanical properties are the main drawbacks of starch-based plastics. Using wood fibre to reinforce thermoplastic starch did not gain attention until the end of the 1990s. In 2001, researchers^{280,281} reported that (i) the addition of wood fibre could increase the modulus and the tensile strength of the composite by up to 156% and 120%, respectively, and reduce the water uptake of the composite,²⁸¹ and (ii) the addition of wood

flour could influence the rheological behaviour of the composite and result in a slight decrease of mechanical performance.

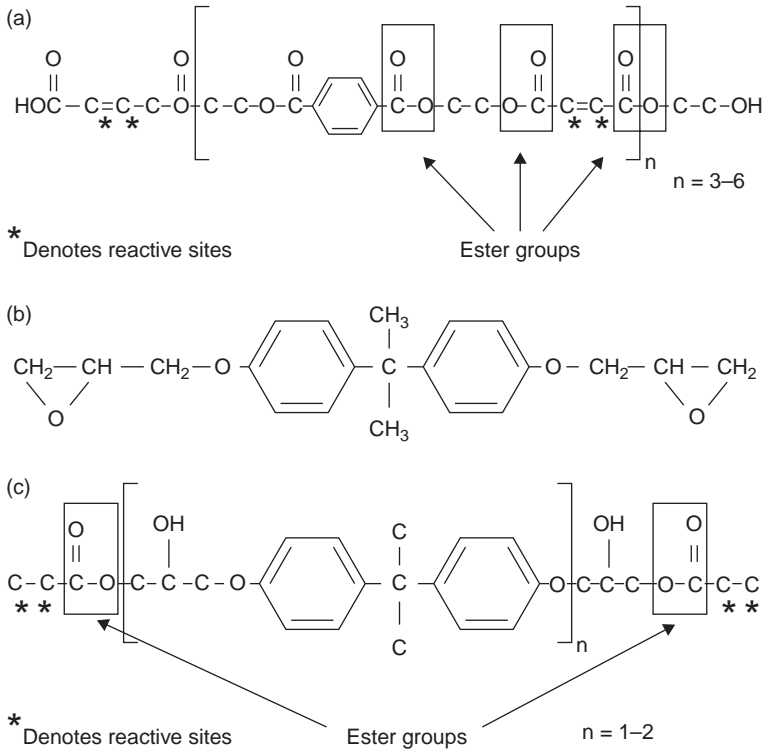
Wood flour has been used as filler for rubber matrix. The addition of wood flour could increase the modulus, hardness and tensile strength of rubber. Wood fibre–rubber composites can be processed by extrusion, injection moulding, calendering and milling. They can be used for automotive tyres, sheeting, metal braids, belts, diaphragms and gaskets, roofing and ship fenders.

PLA is a versatile biopolymer made from renewable agricultural raw materials, and is fully biodegradable. It can be processed similarly to polyolefin. The final PLA-based products display good stiffness and strength. PLA–wood fibre composites can be processed by extrusion, injection moulding, film and sheet casting, and spinning, providing access to a wide range of materials. The main drawbacks of PLA-based products are low toughness and thermal stability. Due to their inferior interface,²⁸² the addition of wood fibres without any treatment has negative effects on the mechanical performance. In addition, the low aspect ratio of wood fibre will reduce the strength of composites.²⁸² Grafting²⁸³ and coupling^{284,285} treatments of wood fibres, as aforementioned, have been reported to improve the strength of the PLA–wood fibre composites significantly. In addition, the nanoscale wood fibres without treatment have been found to have a positive reinforcement effect on the PLA matrix.²⁸⁶

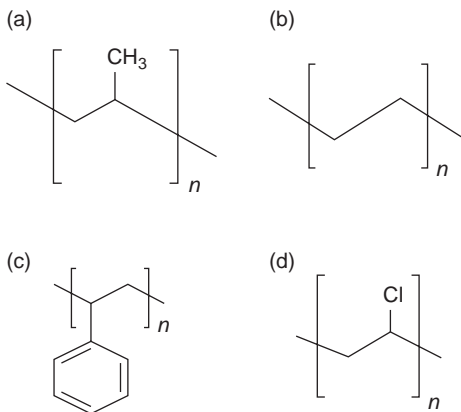
1.4.3 Synthetic polymers

The synthetic polymers that are used as the matrix for wood fibres include thermoplastics and thermosets. Extensive research has reported the incorporation of wood fibre with various synthetic polymers. Wood–synthetic polymer composites, namely, WPCs, can be dated back to as early as 1900. Over the past 110 years, the matrices used for WPC have shifted from thermosets to thermoplastic. Currently, the raw materials for WPCs consist of a broad range of materials, from those manufactured from waste to those consisting of pulped wood and engineering resins. Formulating for the desired outcome is key for wood fibre–based composites outperforming conventional filled or reinforced products. The main applications of WPCs are decking, consumer goods, car interior parts, house-ware, construction, etc. WPCs have many inherent benefits, including consistent material quality and attractive visual appearance.

PP,^{250,287–299} polyethylene (PE),²⁸⁹ polystyrene (PS)^{300–303} and poly(vinyl chloride) (PVC)^{304–307} are the most commonly used thermoplastic matrices. The use of thermoplastic composites is diverse, ranging from automotive applications where they serve as door panels and gearshift knobs, to



1.7 Chemical structure of thermoset matrices: (a) polyester; (b) epoxy; (c) vinyl ester.



1.8 Chemical structure of thermoplastic matrices: (a) polypropylene; (b) polyethylene; (c) polystyrene; (d) poly(vinyl chloride).

Table 1.9 Advantages and disadvantages of synthetic matrices

Resin	Advantages	Disadvantages
Polyester	Tough and rigid; processed by thermoplastic operations; recycled into useful products as basis of resins in such applications as sailboats	Subject to attack by acids and bases; low thermal resistance, poor solvent resistance
Epoxy	High mechanical and thermal properties; high water resistance; long working times available; temperature resistance; low cure shrinkage	More expensive than vinylesters, critical mixing, corrosive handling
Vinyl ester	Very high chemical and environmental resistance; higher mechanical properties than polyester	Postcure generally required for high properties; high styrene content, higher cost than polyester; high cure shrinkage
Polypropylene	Low cost, low coefficient of friction; excellent flexural, impact strength; excellent moisture fatigue and chemical resistance	High thermal expansion; UV degradation; poor weathering resistance; flammable
Polyethylene	Lightweight; resistance to stain and organic solvents; low moisture absorption; resistance to electrolytic attack; good fatigue and wear resistance; excellent stress and crack resistance	High thermal expansion; poor weathering resistance; subject to stress cracking; difficult to bond; flammable; poor temperature capability; low strength/stiffness
Polystyrene	Low cost; non-hygroscopic; good optical clarity; easily processed; good thermal stability; good property retention; good creep resistance; easily decorated; easily bonded; good toughness	Thick sooty smoke Poor weatherability Highly flammable Highly notch sensitive Poor resistance to petroleum solvents
poly(vinyl chloride)	Relatively inexpensive; very compatible with additive and plasticizers; good weatherability can be made to be extremely flexible; easy to decorate; excellent abrasion, cut through, and solder resistance; heat shrinkable; flame retardant; lightweight compared with steel alternatives; good insulator	Hydrochloric gas can be formed when burned; not good for food packaging because of; plasticizer leakage; non-biodegradable; unable to use in Europe (banned); can be highly reactive when mixed with acetal

building materials where they have been successfully used as decking and window framing. Polyester,^{308–310} epoxy⁸² and vinyl esters (VE)³¹¹ are used as the thermoset matrices. The main chemical structures, advantages and disadvantages of each of these matrices are given in Figs 1.7, 1.8 and Table 1.9 respectively. The thermoset matrix-based composites include furniture, housing panels, doors and windows, ceilings, flooring, household articles, containers, mechanical components and corrugated boards.

1.5 Process techniques of wood fibre composites

The processing of wood fibre composites has a crucial influence on the properties of the final products. Generally, the selection of process techniques is based on the matrices, the feed form of the raw ingredients, final product (e.g. form, application) and costs.

The processes of compression,^{45,312–316} extrusion^{289,293,294,296,306,317–319} and injection moulding^{291,313,315,320–322} have been used for the inorganic compound, natural high polymer, thermoplastic matrices. These methods are also the most popular processes within the wood–fibre composite industries. The sheet moulding compound (SMC)³²³ and resin transfer moulding (RTM)^{315,324–326} are suitable for thermoset matrices.

Moisture control, along with the specific mechanical energy (SME) generated during processing, plays an important role in establishing the final composite properties. Maintaining the desired pressure and temperature throughout the component over the required period of time is the basic requirement of all composite manufacturing processes.³²⁷ The feature of low thermal stability limits the process temperature and the matrices for natural fibre-based composites. Maintaining the critical aspect ratio of the wood fibres is essential in order to obtain the reinforcing characteristic of the fibre.

Compounding is the process of blending wood fibre with matrices. It is a critical step for the manufacture of wood fibre composites. The purpose of a compounding operation is to produce a pelletized feed stock that can be processed further, such as by injection moulding, extrusion or thermoforming. There are several types of compounding processes, including extrusion, kneading, and high-shear mixers. Manufacturing processes can be divided into open mould and closed mould. The open mould process comprises wet lay-up, bag moulding and autoclave moulding. The closed mould process comprises transfer moulding, compression moulding and injection moulding.

Another main stream of wood fibre composites is to use resins as binder for the composites, such as MDF, PB, and oriented strand boards (OSB) and hardboard (HB). The composites generally consist of over 90% wood fibre or particles, and less than 10% of resin binding systems. Random distribution

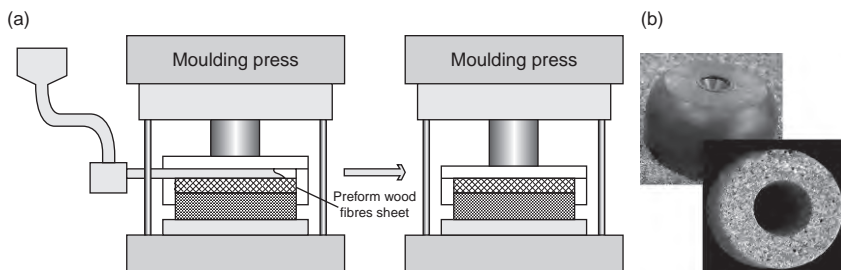
of resin over the surface of the fibre or particles governs the properties of final products. Hardboard, one of the fibreboards, can also be manufactured without any binder, using water as a medium.

1.5.1 Compression

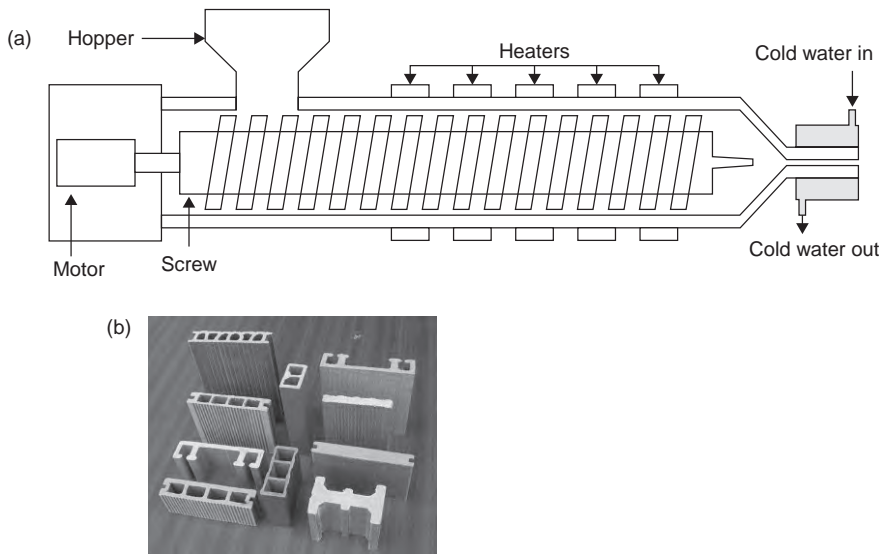
Compression (Fig. 1.9) is a method of moulding in which the moulded material, generally preheated, is first placed in an open, heated mould cavity.³²⁸ The compression moulding technique has proved suitable for the production of profiles with any thermoplastic and thermoset composites. The mechanical properties and dimensional stability of compression moulded composites are influenced by the design of the mould cavity, location of the charge and processing parameters, e.g. compound temperature, mould cooling or heating rate, closing speeds and the filler loading.³²⁹ The compression method is common, popular in the manufacture of WPC. It can be connected with other processes (e.g. extrusion,^{150,330,331} injection³¹⁴) to form a new process. It also can be used as one component of the manufacturing process (e.g. SMC, bulk moulding compound (BMC)).

1.5.2 Extrusion

Extrusion (Fig. 1.10) is by far the most important processing method for WPC. Non-synthetic polymers, e.g. cement,^{317,332} starch^{289,333} and PLA,³³⁴ can be used as the matrices in this process. It involves two types of extruders: single-screw and twin-screw. Due to the modular character³³⁵ and other advantages (e.g. excellent compounding capability, good functional versatility) over single-screw extruders, twin-screw extruders dominate today's market for the manufacture of wood fibre composites. A suitable combination of processing variables including screw speed, screw configuration, throughput



1.9 Schematic of compression moulding (a) and compression moulded wood fibre composite products (b).



1.10 Schematic of extrusion moulding (a) and extrusion moulded wood fibre composite products (b).

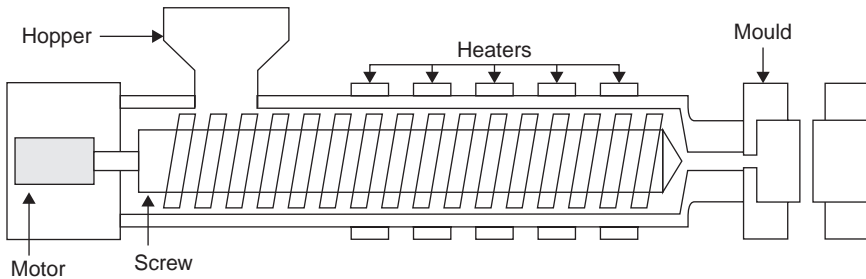
rate and barrel temperatures is necessary to limit thermal degradation and darkening of the filler.

Extrusion has overcome the shortcomings of Hatschek and other conventional processing systems of wood fibre cement, and has improved the mechanical performance. The extruded wood fibre cement products offer advantages in terms of the versatility of section profiles, end product performance characteristics, and production throughput. Wood fibres offer a desirable balance of performance and cost in this composite. A comparison between the extruded and cast fibreboard³³⁶ reveals that the extruded products are better in terms of strength, stiffness, toughness, fibre distribution, fibre orientation, and bond of fibre with matrix, even in the presence of a higher percentage of air voids.

1.5.3 Injection moulding

Injection moulding is a versatile process accounting for nearly one-third of polymeric materials processed, particularly where intricate shapes are needed in cyclic, high volume production. The benefits include excellent dimensional tolerance and short cycle times coupled with few post-processing operations (Fig. 1.11). One of the challenges posed by injection moulding wood fibre composites is to produce pellets of a consistent quality. Injection moulding requires a polymer with a low molecular weight to

(a)



(b)

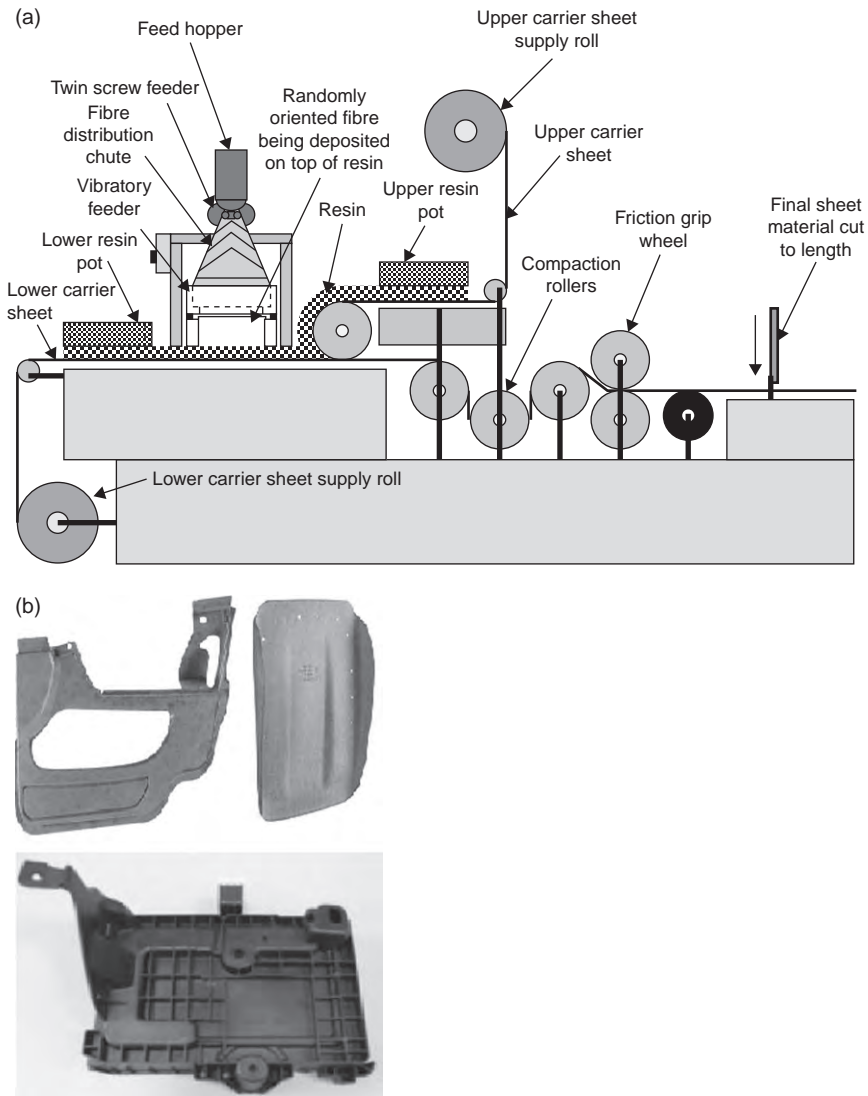


1.11 Schematic of injection moulding (a) and injection moulded wood fibre composite products (b).

maintain low viscosity. The process temperature³³⁷ and the fibre length³³⁸ have little influence on the mechanical properties of the composite. It has been reported that wood fibre reinforced PP composites manufactured by the injection moulding process showed better performance than those from the compression moulding process^{339–342}

1.5.4 Sheet moulding compound (SMC)

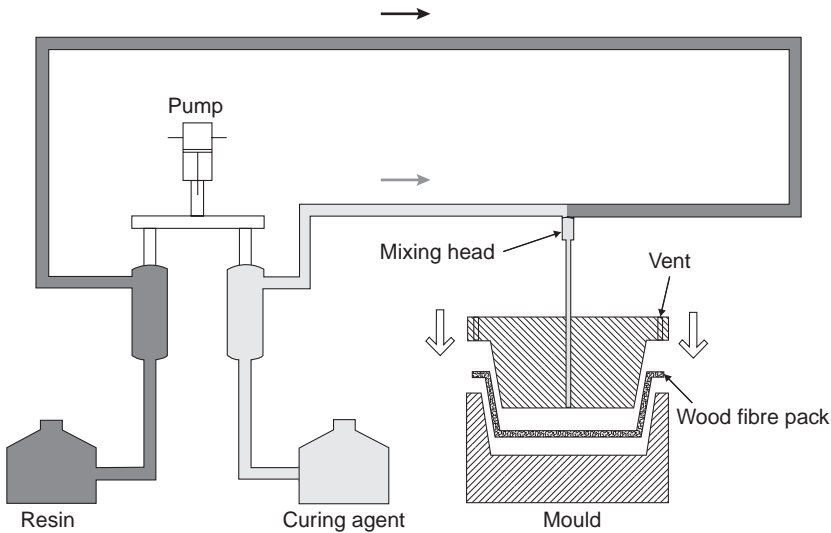
SMC is produced in a two-step production process^{343,344} (Fig. 1.12), which is normally used for short fibre reinforced thermoset composite. The first step is the preparation of the so-called prepreg. The prepreg must remain in a homogeneous state during flow to the extremities of the mould. The second step is the compression. The processing parameters affect the properties of the end product and are critical to ensure the uniformity of mat weight, short wood fibre content and final sheet viscosity.



1.12 Schematic of SMC (a) and SMC moulded wood fibre composite products (b).

1.5.5 Resin transfer moulding (RTM)

RTM is a low pressure and low-temperature method of transferring a liquid thermoset resin into a closed mould, which offers a dimensionally accurate and high quality of surface finish of composite. RTM is applicable to various forms of fibre reinforcement (Fig. 1.13). RTM of natural fibre composite



1.13 Schematic of RTM.

could provide additional technological, economical, ecological and environmental benefits; however, there are additional influencing factors, including fibre washing, edge flow and fibre concentrations.³⁴⁵ The utilization of wood fibre as a glass fibre replacement to make fibre reinforced composite under the RTM process has been reported to have a positive contribution to stiffness and strength of composites.³⁴⁶

1.6 Properties of wood fibre composites

1.6.1 Mechanical properties

Among the mechanical properties, the tensile, impact and flexural strength are the most important parameters for evaluating the performance of wood fibre composites. Various factors are found to have effects on the mechanical properties of wood fibre composite. Some of the important factors are listed in Table 1.10. The list of factors as given in Table 1.10 is not exhaustive but emphasizes the complexity of the subject. All the factors are almost equally important in influencing the properties of wood fibre composites.

1.6.2 Physical properties

One drawback of wood fibre composites, compared with other conventional inorganic fibres for composites, is the inherent susceptibility of cellulosic

Table 1.10 Factors affecting properties of natural fibre reinforced concretes

Factors	Constituents	Results	References
Fibre types	1. Softwood, hardwood	Softwood > hardwood	82, 347, 348
	2. Wood flour vs sisal	Sisal > wood flour	349
	3. TMP vs hemp	Hemp > TMP	350
Fibre geometry	Length (30, 300 μm)	300 μm > 30 μm	333
Processing of fibre	TMP, CTMP	TMP < CTMP	82
Fibre orientation	Isotropy, anisotropy	Isotropy > anisotropy	82
Fibre modification	1. Various coupling agents: (i) pyromellitic dianhydride (PMDA); (ii) benzophenone-3,3',4,4'-tetracarboxylicdianhydride (BPDA); (iii) 1,4-phenylene diisocyanate (PPDI); (iv) methylene-bis-diphenyl diisocyanate (MDI); (v) γ -mercaptopropyltriethoxysilane (MRPS); (vi) γ -methacrylopropyltriethoxysilane (MPS)	(i) In CAB matrix PMDA and BPDA are the best; (ii) in rubber matrix MPS is the best	248
	2. Coupling, surfactants, benzylation	Benzylation > surfactants > coupling	351
Processing	1. Compounding (extrusion vs mechanical blending)	Extrusion > mechanical blending	352
	2. Injection moulding vs compression moulding	Injection moulding > compression moulding	231, 339

fibres to moisture expansion. Due to the abundance of hydroxyl groups,³⁵³ mainly in the amorphous regions of hemicellulose in the cell wall of the wood fibres, the fibres will swell in the presence of moisture or water. As a consequence, mechanical properties, such as stiffness and strength, are negatively influenced.³⁵⁴ Lignocellulosic-fibre reinforced plastics also tend to swell considerably on water uptake, both in the plane of the composite panel and in the thickness direction, and therefore reduce the dimensional stability of the composite.^{355,356} This drawback limits the application of natural fibres, and results in extra energy consumption for the fibre drying. By using chemical treatment or physical treatment, this negative influence could be reduced significantly. Another limitation of using wood fibre in fibre composite applications is its low thermal resistance and thermal degradation initiating around 200°C.³⁵⁷ Therefore, most polymers used for wood fibre composites have relatively low melting temperatures. The degradation is determined by the wood species and composition.

Table 1.11 Comparison of the thermal insulativity of natural fibre-based materials and cement-based concrete

Material	K-value (W/mK)
Fibreglass	0.033–0.40
Sheep wool batts	0.038–0.043
Wood fibre batts	0.038–0.044
Cotton batts	0.038–0.045
Flax batts	0.042
Hemp batts	0.043
Wood fibre board	0.08
Hempcrete	0.12–0.13
Cement based concrete	0.3178

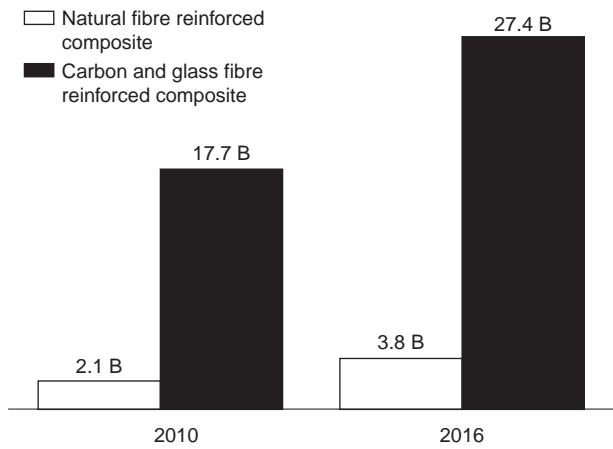
Table 1.12 Embodied energy and carbon of fibreglass, timber and wood fibre-based board

Material	Embodied energy (MJ/kg)	Embodied carbon (kgC/kg)
Fibreglass	28	0.417
Timber	8.5	0.125
Particle board	9.5	0.139
Medium fibreboard	11	0.161

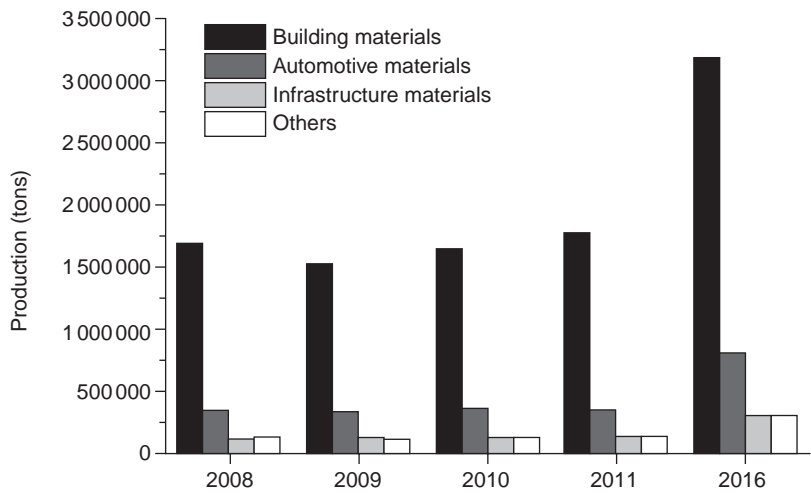
However, wood fibre also displays some positive effects on the composites due to its pronounced characteristics. The fibrous shape can dissipate wave energy and convert sound energy into heat.^{358,359} Wood fibre-based composites display a higher sound absorption coefficient over other fibre-based composites.³⁵⁸ The excellent characteristics of low thermal conductivity and climate friendliness of wood fibres are other advantages worthy of attention for developing functional composites for specific requirements (Tables 1.11 and 1.12).^{360–362}

1.7 Applications of wood fibre composites

The use of wood and other natural fibres in composites is predicted to grow in future due especially to their sustainability, renewability and environmental factors. In 2010, the global natural fibre composite market reached US\$2.1 B³⁶³ (Fig. 1.14). It is estimated that the market for natural fibre composite will reach US\$27.4 B in 2016.³⁶⁴ It should be noted that these data exclude the wood fibreboard market, which has reached US\$8.5 B.¹³² Wood fibre composites have been exploited extensively in the previous century. These oldest resources are still being widely used in a large number of applications, and this is showing a robust growth rate (Fig. 1.15). Among these products, the building and automotive sectors are the main users of these materials, sharing 70.8% and 12.5% of the natural fibre composite market



1.14 Comparison of composite market of natural and synthetic composites (US\$ billions).



1.15 Application of natural fibre composites.

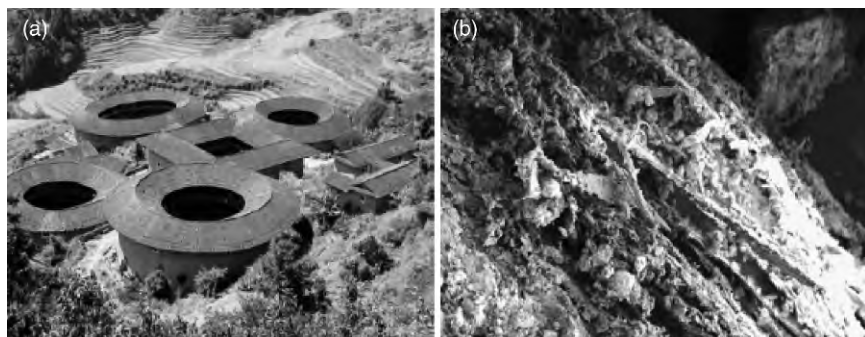
respectively. As one of the natural fibres, wood fibre composites overall possess 80% of the market share of natural fibre composites (Table 1.13).

1.7.1 Wood fibre composites in building applications

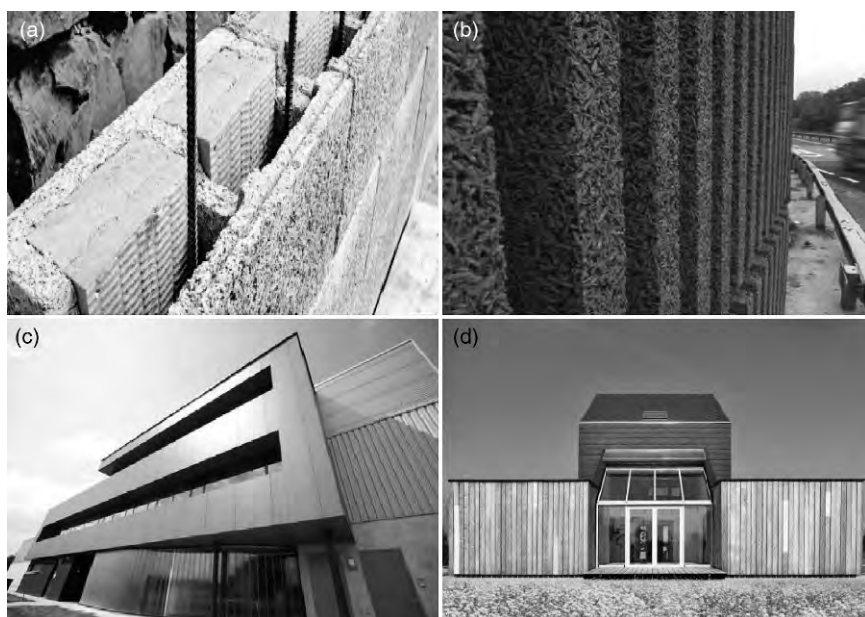
Using wood fibre composites in building has the longest history. One of the most successful examples is the Tulou building (Fig. 1.16a). Tulou buildings, built with wood fibre–clay composite, are a unique type of rural dwelling

Table 1.13 Wood fibre composites products

	Sector			
	Building	Automotive	Industrial/infrastructure	Other
End market application	Cladding; doorframes and components; ducting; fascia, soffits and barge boards; pre-finished floorboards; roofline products; shingles; stairs; window frames and components; balustrades; blinds; coving; dado rails; decorative profiles; interior panels; interior panels; kitchen cabinets; laminate flooring; sound proofing; shelving; skirting boards	Door and head liners; ducting; interior panels; rear shelves; spare tyre covers; truck floors	Handrails; industrial packaging; marine pilings/bulkheads; pallets/crates totes; piles/docks; railings; railway sleepers; rubbish bins; signage	Black piano keys, hot tubs



1.16 Wood fibre–clay composite in building: (a) Tulou; (b) scanning electron microscope (SEM) morphology of Tulou.³⁷⁰



1.17 Wood fibre–cement composites in building: (a) wall; (b) sound wall; (c) cladding; (d) house.

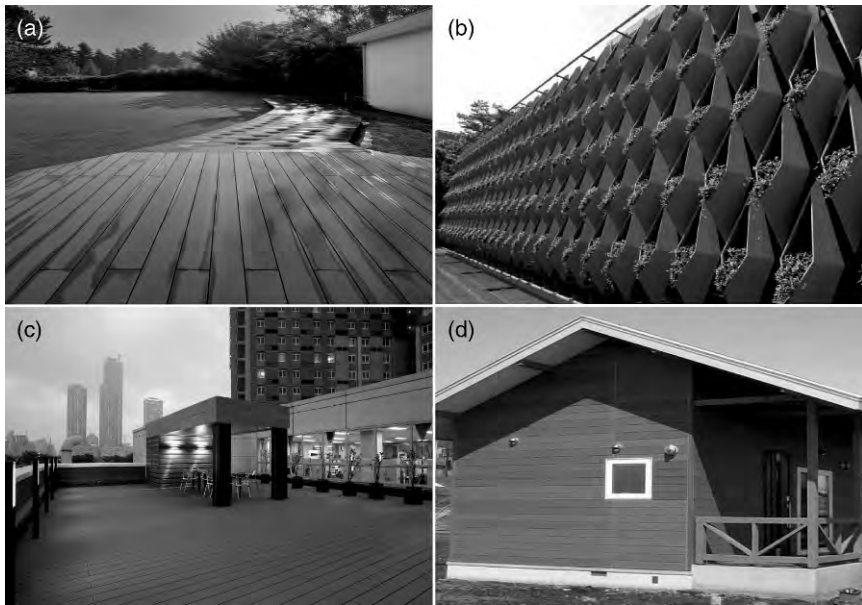
in south China. This wood–clay composite building has displayed excellent mechanical durability. After 1240 years, the compressive strength of walls in some Tulou still reaches up to 30.17 MPa,^{365,366} due to the reinforcement of wood fibres (Fig. 1.16b). It also displays excellent environmental performance, e.g. the annual household energy consumption for this kind of building is only

2.43×10^4 MJ/y, which is far lower than for other rural buildings in this region. Compared with modern building (cement building), this kind of building can make residents feel thermally comfortable.³⁶⁷

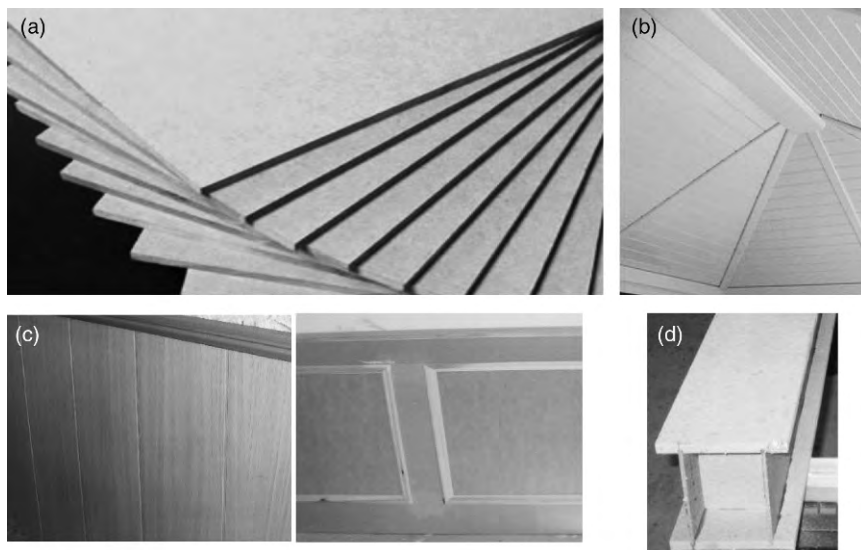
The second form of wood fibre composites is the wood fibre–cement composite, which has been used to manufacture a wide variety of products primarily for the construction industry, such as for insulating concrete forms, noise barriers, cladding and house-building (Fig. 1.17). This kind of composite is dimensionally stable when subjected to water soaking, as compared with common synthetic binder–wood fibre composites. In addition, this kind of composite presents excellent fire resistance.

The third form of wood fibre composite is wood fibre–plastic composite. This kind of composite has the perfect natural look and feel of wood. It is generally lightweight but offers similar levels of strength and structural support as solid wood. The development of wood fibre–plastic composites exceeds that of other plastic industries since its emergence in the late 1980s. Wood fibre composites are currently undergoing a significant market expansion as a replacement for solid wood, mainly in building applications such as steps, trellises, flooring and cladding (Fig. 1.18).

The fourth type of wood fibre composite includes MDF and HB. This group of wood fibreboards is the engineered wood products formed by the



1.18 Wood fibre–plastic composites in building: (a) steps; (b) trellises; (c) wood flooring; (d) cladding.³⁷²



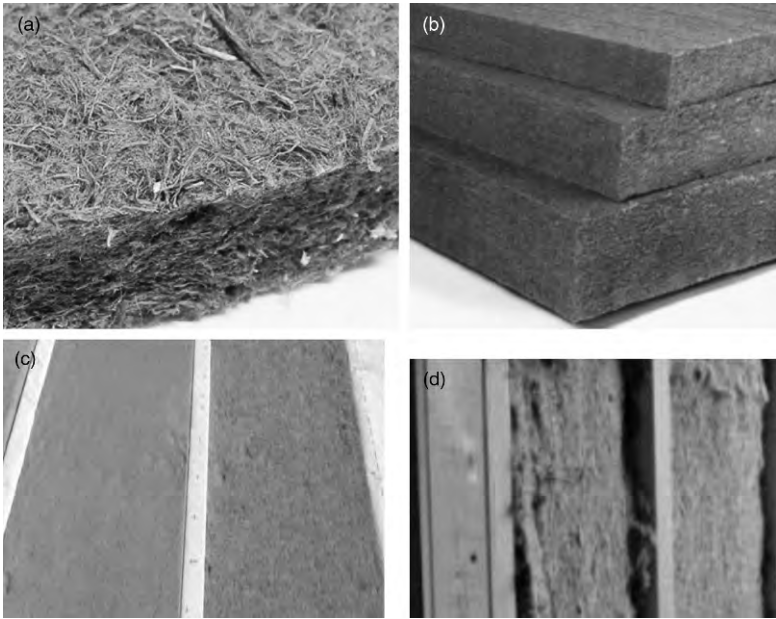
1.19 MDF in building construction: (a) MDF raw material; (b) roofing; (c) wall panels; (d) box beam.

separated fibres but can be used as building materials. It is normally stronger and denser than PB. MDF has been used for building and construction over the last several decades (Fig. 1.19). MDF is ideal as a wall covering, offering better resilience to wear and tear and temperature changes than PB, plastic wall and solid wood panels. MDF may also be used for door panels, such as creating door skins which may be laminated to the whole surface and edge of an MDF board.

The fifth type of wood fibre composite is the wood fibre insulation board. These products are generally without, or with a very small amount, of resin binder added during their production. Wood fibre fabrics as insulation materials are normally loosely formatted and non-woven (Fig. 1.20). They can also sometimes be loose-filled or wet-sprayed on site. These green products can be produced by using 80% recycled newspaper, cardboard and paperboard. The products may also be treated with chemicals to obtain fire resistance or other properties. It has excellent acoustic properties and similar thermal resistance to other insulation materials, such as fibreglass.

1.7.2 Wood fibre composites in automotive applications

The application of natural fibre composite can be traced back to the 1940s. In 1942, Henry Ford developed the first prototype composite car made from hemp fibre.³⁶⁹ In the 1950s, the body of the East German Trabant was built



1.20 Wood fibre board as insulation: (a) non-woven mat; (b) softboard; (c) roofing; (d) loose wall insulation.



1.21 Natural fibre composites in automotive applications: (a) outside of hemp car; (b) inside of hemp car; (c) outside of wood car; (d) inside of wood car.

from composites containing natural fibres. However, owing to the development of petrochemical-based polymer, this green technology was not economical and lost its place in the automotive industry. In the 1970s, the first oil crisis revived natural fibre composites in the automotive industry. WPC was born at that time in Italy. In the 1980s, automotive door panels made from all-bio composite natural fibre and bioresin was developed. In the 1990s, Daimler-Benz pioneered the use of natural fibre in commercial vehicles as part of the 'Belem' project based in the Amazon delta in South America.³⁷⁰ This application not only improved the quality of life for the individuals involved in this new application, but it also became a commercial success and ensured its continuation. At the end of 2000s, the natural fibre composite for the automotive industry had become a fashionable concept in advanced automotive design (Fig. 1.21).³⁷¹ The well-established applications of natural fibres in the automotive sector are summarized in Table 1.14.

1.7.3 Wood fibre composites in aerospace applications

Natural fibre composite is the precursor of aerospace materials. As early as the 1920s, Messrs Caldwell and Clay had employed natural fabric composite to make airscrews. In the 1930s De Bruyne developed a material known as 'Gordon-Aerolite', a composite of paper and unidirectional, unbleached flax yarn impregnated with phenolic resin and hot pressed. This material was used experimentally to produce a full-scale main wing spar for the 'Bristol Blenheim' light bomber, and for the fuselage of the 'Supermarine Spitfire' fighter aircraft.³⁷² After the World War II, the application of natural fibre composite in aerospace material came to a near-halt. However, at the end of the 1990s, aerospace manufacturers started to look for a lightweight and sustainable aircraft material, and as might be expected natural fibre-based composites attracted their consideration. In 2005, the resurgence of the global automotive market and the interest of leading players in the aerospace industries for sourcing natural fibre reinforced composite products and technology sparked a project on natural fibre reinforced composites, called the AMTS programme, in South Africa, to address the research problems and aiming to fulfil the technical need for structural and exterior components.³⁷³ In 2006, researchers at the State University of New York first made real progress by using NT to create wood-based cellulose that was strong enough to strengthen plastics for aerospace application. A wood-and-sisal reinforced honeycomb core has also been developed with high mechanical performance and other functional properties for aerospace application.

1.8 Future trends

Wood fibres are low cost, lightweight, very tough and environmentally friendly compared with other traditional reinforcing materials, such as

Table 1.14 Vehicle manufacturers and use of natural fibre composites

Automotive manufacturer	Size	Model applications
Audi	A2, A3, A4 (and Avant), A6, A8,	Roadster, coupe, seat backs, side and back door panels, boot lining, hat rack, spare tyre lining
BMW	3, 5, 7 Series	Door panels, headliner panel, boot lining, seat backs, noise insulation panels, moulded foot well linings
Citroen Daimler-Chrysler	C5 A, C, E and S-class models, EvoBus (exterior)	Interior door panelling Door panels, windshield, dashboard, business table, pillar cover panel
Ford	Mondeo CD 162, Focus	Door panels, B-pillar, boot liner
Lotus	Eco Elise (July 2008)	Body panels, spoiler, seats, interior carpets
Mercedes-Benz	Trucks	Internal engine cover, engine insulation, sun visor, interior insulation, bumper, wheelbox, roof cover
Peugeot	406	Seat backs, parcel shelf
Renault	Clio, Twingo	Rear parcel shelf
Rover	2000 and others	Insulation, rear storage shelf/panel
Toyota	Brevis, Harrier, Celsior, Raum	Door panels, seat backs, Spare tyre cover
Vauxhall	Corsa, Astra, Vectra, Zafira	Headliner panel, interior door panels, pillar cover panel, instrument panel
Volkswagen	Golf, Passat, Bora	Door panel, seat back, boot lid finish panel, boot liner
Volvo	C70, V70	Seat padding, natural foams, cargo floor tray

glass fibres. Scientific work and technical application in combining wood fibres with various matrices over the previous century has culminated in the growth of new markets and industries. The major market for wood fibre composite is in building construction, and this will continue and control the building material market. The increasing acceptance in the automobile

industry indicates that the wood fibre reinforced composites are becoming more mature materials, and attention has been drawn to the use of wood fibre composites in aerospace materials. The main drawback of wood fibres is their weak mechanical performance when compared with glass fibre or carbon fibre. This drawback limits the application of wood fibre composite in hi-tech engineering materials, but much research has been initiated to develop enhanced products.

The science and technology of the interface between wood fibre and matrix has always been an interesting and important research topic. Various modifications, such as alkali treatment, oxidation, crosslinking, grafting and coupling treatment, have been assessed and continue to be optimized.

The development of NT seems to be capable of bringing wood fibre composites to their full potential in the near future. Wood-derived nanocellulose could reach higher tensile strengths than carbon fibre. Since the first report on the superior performance of nanocellulose composite in 1995, the application of nanocellulose has been pushed into a new era and there has emerged extensive research in the application of nanocellulose. Most major governments around the world are investing heavily in nanocellulose. In 2002, the European Commission underpinned the 'NANOFOREST' project to develop a research and development roadmap, and to recognize the newly emerging development in NT and related areas suitable for practical application in the forest products sector. In 2004, the US initiated 'NT for the Forest Products Industry – Vision and Technology Roadmap'. In 2011, Innventia opened the world's first pilot plant for the production of nanocellulose, which has a capacity of 100 kg/day. This facility makes it possible to produce nanocellulose on a large scale, which is an important step towards the industrialization of the technology, promoting the application of nanocellulose in composites and boosting the exploitation of wood resources. In 2012, Forest Product Laboratory (FPL) USA opened a pilot plant for the production of wood-derived cellulosic nanomaterials. Although the large scale production of wood nanocellulose is still on its way, a boost from a new generation of wood fibre composites will come in the next decades. It is predicted that this new material would create jobs and contribute \$600B to the economy by 2020.

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