Wood Fibres for Papermaking



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Prologue

The pulp and paper industry has been confronted with economic and efficiency issues, determined by the accessibility and cost of raw materials, especially due to the fact that paper represents an important commodity. In this situation, the main preoccupations are oriented to the use of new, renewable resources and to approach their complex processing with the aim of recovering other components (hemicelluloses, lignin and extractives), along with manufacturing different kinds of paper products. Thus, there are new possibilities of extending the use of biomass compounds, chemically or biochemically modified, in the synthesis of other polymers or to obtain products with chemical and energetic values. Therefore, upon the application of new technologies it is possible to increase the value of renewable resources associated with improving the efficiency of the pulp and paper industry. Based on these considerations, Helene Chavaroche suggested that I initiate a project to gather information concerning new tendencies/technologies within the pulp and paper industry, capable of developing different compounds based on cellulosic products. This project ended last year with the book Pulp Production and Processing: From Papermaking to High-tech Products, published by Smithers Rapra Technology Ltd. The book contains the following chapters: 1) Biorefining and the pulp and paper industry; 2) Pulping fundamentals; 3) Chemical pulp bleaching; 4) Oxygen bleaching; 5) Chemistry and physics of cellulose and cellulosic substances; 6) Physico-chemical characterisation of cellulose from Broussonetia papyrifera bark and stem, and Eucommia ulmoides Oliver stem; 7) Cellulose fibres in the papermaking process; 8) Cellulose esters from traditional chemistry to modern approaches and applications; 9) Lyocell processes and products; 10) Functional cellulose

microspheres; 11) Processing cellulose fibres to the micron and nanoscales; 12) Optical properties of cellulose esters and applications to optical functional films; 13) Antibacterial fibres; and 14) Recent advances in the processing of biomass feedstocks for biohydrogen production.

Raw material plays an important role in the pulp and paper industry. Keeping in mind the new tendencies in the evolution of this industry it was considered of interest to discuss special aspects concerning wood fibres, as the traditional resources used since the 1950s (softwoods) have been diversified by adding hardwoods and annual plants. At the same time, many of them are more accessible in the South and South-east of the world. Initially, this contribution was proposed by Maria Cristina Area and was to contribute to the above-mentioned book. However, considering the aspects to be treated represented a special focus, Helene Chavaroche decided to edit this as a supplementary book - Wood Fibres for Papermaking. In the last two decades the majority of the pulp and paper industry has moved to the South and South-east of the world, due to the favourable conditions of high forest productivity cultivated with fast-growing species such as eucalyptus, willow and poplar hybrids, allowing a decreased price of pulp and paper products. These species are characterised by particular fibre properties that determine many of the studies into the manufacture of paper which exhibit good physico-mechanical characteristics. At the same time, there is interest regarding the conditions required to extend the use of these species to be exploited as raw materials in biorefining processes, with the possibility of recovering other compounds. This situation determined that in natural forests, large surfaces were to be afforested with the above-mentioned species, which are valuable resources for the pulp and paper industry. Study of the anatomical structure and chemical composition are crucial because these aspects influence the chemical processing of wood (chemical consumption, yield and so on), and the characteristics of fibrous materials; that is why the main structural differences between traditional and new raw materials are underlined, and a correlation with paper properties is discussed. Some influences are explained by existing differences between normal and abnormal

wood (reaction wood), and juvenile wood. Characteristics of the cell wall are analysed, taking into account the basic properties of the fibres from the point of view of their paper potential. This systematic analysis is very useful to obtain paper with predetermined characteristics, similar to those manufactured from traditional species but employing new species on a large scale.

Therefore, I appreciate this book represents a fundamental contribution for those working with these new materials (especially hardwoods) from the South and South-east of the world, where the majority of pulp and paper production is now located. Along with the book issued last year, this book can be recommended both to scientists and producers involved in teaching, research and industrial fields.

Valentin I. Popa

2014

Preface

Paper is generally obtained using natural fibres from renewable resources; it is biodegradable, recyclable, and an important vehicle of communication and culture. It is manufactured by depositing a dilute suspension of pulp on a fine wire that allows water to drain through it while retaining the fibre network. Once separated from the wire, the fibre layer is dried and pressed, forming a sheet with particular characteristics, according to the raw material and methods of preparation used. The overall behaviour of paper (chemical and mechanical properties, stability, degradation and so on) is strongly dependent upon the nature, origin and characteristics of components as well as upon their interactions. The characteristics of the fibre strongly influence the properties of the paper. For example, long fibres make a strong sheet, because their length allows a better interweave, whereas this does not happen with very short or rigid fibres. However, certain paper qualities require the presence of short fibres as they fill the interstitial spaces making a sheet more 'closed', which improves formation.

Firs and pines dominated the global picture of raw materials used by the paper industry until the 1950s. At that time, the interest in introducing new species, mostly hardwoods, led researchers to intensify efforts investigating the characteristics of the fibres, and their combinations, which could represent the relationship between fibres, pulp and paper.

The pulp and paper industry has shown, mainly in the last two decades, a strong North-South displacement. This is to a large extent due to the favourable climate, which promotes tree development. Similarly, the paper fibres have gone from being almost exclusively

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softwoods from natural forests of the cold regions of the northern hemisphere, such as spruce and fir, to fast-growing species of short fibres, such as eucalyptus, and willow and poplar hybrids from plantations.

These new species, which are beginning to dominate the paper panorama, not only differ from classic species in fibre length, but they present particular characteristics, such as large amounts of juvenile wood, different fibrillar angle and so on, because trees are increasingly used at a younger age.

This leads us to question whether the old paradigms concerning the relationships between fibre characteristics and pulp properties are still valid or should be reviewed and updated, in which case, the basic fibre parameters, their influence in pulping and refining, and their impact on paper quality should be redefined.

The purpose of this book is to survey publications, mainly of the last decade, to verify which morphological characteristics of the fibres authors currently consider relevant, in order to establish the state of the art for this topic. Relatively recent data were surveyed because of the continuous changes that occur in the species due to genetic improvement.



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Natural Forest and Forest Plantations

1.1 Introduction

Since the invention of the stone groundwood process for newspapers in Germany (1845), the soda-pulping process in England (1866), the bisulfite process in Sweden (1875), and the kraft process in Germany (1882), a new panorama opened in paper manufacture, allowing its production to grow to massive levels and making possible the popularity of wood as a raw material *par excellence*. Nowadays, 90% of printing paper is made from wood-derived fibres, mainly from bleached kraft pulp (composed mainly of cellulose and hemicelluloses) or from mechanical pulps (lignin-containing fibres). The chemical components of the fibres, the manufacturing process, and the additives used affect, to a variable extent, the permanence of paper but this is a small drawback compared with the massive variety of products achieved [1].

Boreal forests, which form the taiga, are found only in the northern hemisphere, making a wide coniferous forest belt, which extends across Canada, northern Europe, Russia and northern Japan. The temperate mixed hardwood-softwood forests are found almost exclusively in the northern hemisphere mid latitudes, covering the eastern parts of the USA, central Europe, Japan, eastern China, New Zealand, Tasmania, southern Africa and South America. The number of species in the latter forests is considerably higher than in boreal forests, ranging from masses dominated by conifers to others formed essentially by pure hardwoods. In the boreal and temperate zones, only the conifer species belonging to the *Pinaceae* family have industrial economical value. It includes the genera: *Pinus, Larix, Picea, Pseudotsuga, Thuga* and *Abies*. In addition, in the boreal and temperate zones, the angiosperm genera *Betulaceae* and *Salicaceae* are important, including the following genera: *Betula*, *Alnus*, *Quercus*, *Fagus*, *Ulmu*, *Acer*, *Salix* and *Populus*, the last two being the most widespread in the temperate zone [2]. The warm climate forests of both hemispheres contain a large number of hardwoods and few conifers.

For many years, the paper industry has sited mills in locations where the local wood supply is most suitable. It can improve its products further by becoming more aware of the influence of tree species and tree age on product performance, and selecting the most appropriate wood source [3].

Coniferous species (also named evergreen, softwoods and long fibre woods) were historically the most used due to the existence of pure natural forests. Moreover, fibre length (L) was always considered a very important factor for pulp quality. For example, the total production of pulp in 1960 was 60 million tonnes (Mt), comprising 78% softwoods, 16% hardwoods and 6% others [4]. However, although by 1940 the *Populus* species in the USA were generally considered weed trees, in the 1960s there was a growing interest generated by these species because of their natural abundance and low use, as stated by a literature survey of these species published in 1968 by the Forest Products Laboratory of the Forest Service of the US Department of Agriculture [5].

Some dicotyledonous angiosperm species (broadleaf, deciduous, hardwoods, short fibre woods), reeds and agricultural residues (bagasse, straw and so on) are increasingly included in paper raw materials. The delay in the utilisation of hardwoods is due to several factors. First of all, in natural forests, in contrast to the situation with conifers, most trees are arranged in mixtures of various species of different characteristics; this leads to difficulties in finding the correct pulping conditions and produces pulps of irregular quality. Dense species also create problems because of their difficult impregnation during the pulping process. Moreover, their irregular shapes make debarking difficult. Finally, their tissue is less uniform, and their fibres are shorter than those of conifers and anatomically simpler. However, these problems have been overcome in the past 40 years, thanks to the development of implantation techniques and genetic improvement,

resulting in an increase in the consumption of hardwood. Moreover, these species provide other interesting properties for certain paper grades that do not require high strength. These woods now represent over two-thirds of the total mass of the utilisable timber, and its global availability is not only limited to certain areas of the world.

1.2 Forest Plantations

The planting of trees as a means of forest regeneration or afforestation has been practiced for a long time. Heritability studies began to show their transcendence back in the 1960s, mainly in the case of coniferous trees [6], whereas in the 1970s, the immense potential of genetic studies to improve the species began to be recognised [7].

The use of natural forests is being increasingly substituted by plantations of indigenous or exotic species (mostly pines and eucalyptus). For this reason, there were no references regarding plantations of fast-growing wood species in the classical pulp and paper books [8–10], but they became increasingly apparent in later decades [11–13]. In the southern hemisphere, especially in South America, forest plantations have been, and remain, the main source of raw materials for the pulp and paper industry [14].

Planted forests account nowadays for 7% of the total forest area or 264 million hectares (ha). In 2005, 2.8% of the total global forest cover consisted of productive forest plantations, accounting for an area of approximately 110 million ha. According to the Food and Agriculture Organization of the United Nations (FAO), there was an increase of approximately 40% in the area of the world's forest plantations used for productive functions from 1990 to 2005 (Figure 1.1). Studies indicate that this growth is set to continue [15].

During 2005–2010, the area of planted forest increased by about 5 million ha per year, most established through afforestation, i.e., planting of nonforested areas in recent times, particularly in China [16]. Southeast Asian forests will produce about 563.8 million tonnes/year of woody biomass for the period 1990–2020; the annual production

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of woody biomass decreased by about 1.5% over the same period. Up until 2009, only 0.08% of the 2.4 million ha of deforested land was converted into forest plantations, and the majority of these lands are still available for plantation [17]. There are more than 1.3 million ha of timber plantation in Australia, mostly made up of conifers or pine exotic (nonnative) trees, including Radiata pine, Caribbean pine, Slash pine and Maritime pine, and the native Hoop pine in subtropical areas. Australia's native hardwoods, mainly eucalyptus, are increasingly grown in plantations (Tasmanian blue gum, shining gum, blackbutt and flooded gum). Plantations comprise less than 1% of the land area but they supply over 70% of the sawn timber produced. The Australian Government aims to triple the 1994 national area of plantations to more than 3 million ha by the year 2020 [18]. The forested area for industrial use has increased, particularly in South American countries. In Uruguay, for example, the plantations reached about 900,000 ha in 2010, while up to 1989 only 45,000 ha had been planted, and between 1993 and 2002, the average annual planting was 58,000 ha [19].



Figure 1.1 Trend in area of productive forest plantations between 1990 and 2005. Reproduced with permission from P. Rekacewicz,

C. Marin, A. Stienne, G. Frigieri, R. Pravettoni, L. Margueritte and M. Lecoquierre, UNEP/GRID-Arendal Collection, Vital Forest Graphics website. ©UNEP/GRID-Arendal Collection, Vital Forest Graphics [15] Approximately 38% of the wood fibre used for making wood pulp globally in 2004 was derived from plantations, whose principal purpose was to supply wood to fibre-consuming industries. The share of plantation fibre used for pulp is increasing annually (by way of comparison, the 1993 analysis showed it to be 29%). On the other hand, about 49% of the fibre used to make wood pulp in 2004 was sourced from managed natural forests [20].

In the USA, thousands of hectares of clonal plantations of poplar hybrids have been established by industrial companies during the past decade. Even though the initial primary purpose of these plantations was to provide fibres to the pulp and paper industry, the objectives have been expanded to include solid and composite wood products and, in some cases, biomass for energy production, carbon sequestration, or the remediation of environmental problems. Silviculture is intensive and rotations are short: 5-8 years for pulpwood and up to 15 years for solid wood products [21]. Between 1953 and 1997, the timber production of the southern USA doubled, its share of USA production increased from 41 to 58%, and its share of world production increased from 6.3 to 15.8%. The largest categories include softwood sawlogs (28%), softwood pulpwood (25%) and hardwood pulpwood (16%) [22]. Raw material utilised for the production of paper products consists of pulpwood and wood residuals from the manufacturing of other products. Naturally regenerated forests consist of natural pine, mixed with various hardwood species. Plantation forestry, which has been limited to pine species in the south, is an agricultural style of forest management that is displacing harvests from naturally regenerated stands. Pine plantations have expanded steadily, from practically none in 1950 to more than 12 million ha in the late 1990s. In 2007 they accounted for about 16% of all timberland. The utilisation of recycled fibre resulted in a relative drop in the demand for virgin wood fibre [23].

Radiata pine (*Pinus radiata*), originally known as *Pinus insignis*, is a native conifer of California, USA, and was first introduced into Australia around 1857 for ornamental plantings. Large-scale plantings using improved seeds started in the early 1970s. Radiata pine has been bred for three generations since the 1950s, with a

realised genetic gain of up to 33% for volume from the first generation and more than 10% gain predicted from the second generation. In 2004, 70% of the annual consumption of Maritime pine (Pinus *pinaster* Ait) for pulp production in Europe came from thinning. Although for three decades the main focus of breeding was growth, soil adaptation and the creation of new varieties, the quality of wood and fibre have recently become important criteria in the selection of trees. In terms of pulp and paper production, trees in which the wood has a low lignin and extractives content, and high polysaccharide (and especially high cellulose) content have been defined as elite trees. Moreover, the wood of elite trees should have a high density, long and coarse fibres with a high proportion of earlywood fibres, and should give rise to pulps with good strength [24]. In 2006, among the 1 million ha of softwood, Pinus radiata was the predominant species in Australia (73% of softwood plantations) growing in all states and territories except the Northern Territory. Improvements of growth rate in Radiata pine over the first two generations of breeding, in combination with improvements in silvicultural practices, have contributed to a decrease in rotation age from around 40-45 years to 27-30 years [25].

According to 1995 data, the planted forest area in Brazil was approximately 4 million ha, of which 60% was hardwood, mainly eucalyptus, and 40% was softwood (about 32 million m³ of softwood and 20 million m³ of hardwood). The planted forests in Chile covered about 1.5 million ha of which 80% was softwood (*Pinus radiata*). The estimated growth of these forests was 13 million m³ of softwood and 4 million m³ of hardwood. Although wood costs in Brazil are low, Chile probably has the lowest cost of softwood in the world. The planted forest area in Argentina was about 0.8 million ha with the same proportion of softwood and hardwood. The estimated growth was 8 million m³ of softwood and 8 million m³ of hardwood. Wood costs are at the same level as found in Brazil [12]. The planted forest area in Brazil increased to almost 7 million ha in 2010 and to 2.4 millon ha in Chile (70% and 60%, respectively in 5 years) [26].

The implementation of plantations has involved management systems and species determined by the environmental constraints. There are two basic kinds of hardwood plantations: those of *Eucalyptus sp*. in warm regions, and of *Eucalyptus* and *Salicaceae (Salix, Populus)* hybrids in temperate regions.

As some species of eucalyptus have a rapid growth in warm, humid climates, they are used as a solution to provide a hardwood cellulosic feedstock in tropical and subtropical regions. The genus *Eucalyptus* is native of the Australian continent and covers about 700 species. The most important species in the world market are *Eucalyptus globulus*, E. camaldulensis, E. saligna, and E. grandis, and Eucalyptus nitens in Chile in recent years. They are widely used because of their high productivity, wood quality, and great adaptability to different soil and climatic conditions [27]. Eucalyptus plantations in the world totalled 20,071,701 ha in 2009, the planted area of this species in that year (million ha) was: 8.4 in Asia; 7.5 in America; 2.4 in Africa; 1.3 in Europe and 1.0 in Oceania. The distribution of eucalyptus plantations by country was: 21% in Brazil; 19% in India; 13% in China; 5% in Australia; 3% in Uruguay; Chile; Portugal; Spain; Vietnam and Sudan; 2% in Thailand, South Africa and Peru; and 2% in Argentina [28]. In the Mediterranean region of Europe, plantations of eucalyptus (E. globulus and E. camaldulensis) and poplar (Populus × euramericana) have replaced the natural forests for industrial uses. In tropical and subtropical areas of South America the eucalyptus constitutes 65% of the plantations in Brazil, 90% in Peru and 80% in Uruguay. Eucalyptus globulus; E. grandis; E. saligna; E. urophylla; E. deglupta and hybrids of the last two species are especially abundant [29].

Eucalyptus pulp was first introduced as a market pulp during the early 1960s. The use of eucalyptus for pulping and papermaking in Brazil started in the 1930s in a sulfite mill (Gordinho Braune/Jundiaí), but the great success came in the mid-1950s with the new pulp and paper kraft mill of the Suzano Company in São Paulo state, Brazil [30].

The growth in volume of bleached hardwood kraft market pulp (BHKP) has been even more impressive, especially for Latin American. A fair part of this growth is explained by the rapidly growing use of BHKP in tissue paper production around the world.

Back in the 1950s and early 1960s, few people in our industry believed in the future of hardwood pulp in general, and particularly in hardwood market pulp. During the 1960s, understanding of the properties of hardwood pulps grew, and as a consequence hardwood pulps started to be used in various grades of paper and paperboard, initially in a small proportion but gradually in growing volumes. In the late 1970s, two large market pulp production units were built in Brazil: in Aracruz and Cenibra. In the 1980s and 1990s, new mills were built and new producers of eucalyptus market pulp such as Jari, Suzano and Votorantim in Brazil, and Arauco, CMPC and Santa Fe in Chile were introduced [31]. According to FAO statistics (**Table 1.1**), most producers of wood pulp for paper and paperboard in the northern hemisphere reduced their production in the last decade, whereas Brazil increased production by almost 100% [32].

Table 1.1 Changes in the 6 largest producers of wood pulp for paperand paperboard 1999–2010 (billion tonnes)										
	USA	Canada	Finland	Sweden	Japan	Brazil				
1999	57,053	25,371	11,579	10,694	10,904	7,121				
2010	49,300	18,536	10,508	11,878	9,387	14,064				
Percentage of										
change	-13.6%	-26.9%	-9.2%	11.1%	-13.9%	97.5%				
Adapted from Pulp, Paper and Paperboard Capacity Survey										
2007–2012, Food and Agriculture Organization of the United Nations,										
Rome, Italy, 2011 [31]										

Brazilian eucalyptuses show a seven-year growth cycle; this is the shortest of all trees worldwide, and translates into very high forest productivity (44 m³/ha/year), whereas in Scandinavia the rotation age of birch is 30–40 years. In the case of conifers, the rotation age of pines in Brazil is 15 years (productivity of 38 m³/ha/year), whereas in Sweden and Finland the rotation age of firs is 70–80 years, and in Canada it is 55–90 years [26]. In the years to come, BHKP will gradually replace the dominant bleached softwood kraft pulp used today, due mainly to the higher growth rate, lower cost and better delignifying properties of the former [33].

Figures 1.2 and 1.3 show the difference between trees of similar age grown in Argentina and Canada



Figure 1.2 Pine (plantation, 10 years old, Argentina) and spruce (approximately the same age, natural forest, Canada)



Figure 1.3 From left to right: 10-year-old willow (*Salix sp.*, approximately 34° latitude S, Delta, Argentina); 10-year-old birch (*Betula sp.*, approximately 47° latitude N, Quebec, Canada); and 7-year-old *Eucalyptus grandis* (approximately 29° latitude S, Corrientes, Argentina)

Besides *Eucalyptus*, the most cultivated broadleaf trees in Argentina are the *Salicaceae* (*Salix* and *Populus*). Genetic improvement has enabled, among other objectives, increased productivity, better

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product quality, crop adaptation to marginal areas and cost reduction. *Populus deltoides* and *Populus nigra* should be mentioned because of their ease of hybridisation, their adaptability to temperate and subtropical regions, and their ability to propagate vegetatively [34]. *Populus alba* hybrids are also usual in Argentina, whereas the most common clones and hybrids of willows are derived from *Salix babylonica* and *Salix alba* [35]. The wood from most plantations are destined for the manufacture of chemimechanical pulp for newsprint [36–41]. Examples of plantations of *Eucalyptus sp.* and *Salicaceae* in Argentina are shown in Figure 1.4.



Figure 1.4 Plantations in Argentina. (a) Eucalyptus grandis;
(b) Populus sp., and (c) Salix sp. Reproduced with permission from INTA Concordia, Entre Ríos, INTA 25 de Mayo, Buenos Aires and Delta of the Paraná River, respectively. ©INTA Concordia, Entre Ríos, INTA 25 de Mayo, Buenos Aires and Delta of the Paraná River [42]

The preferred conifers for plantations are generally pines. Plantations in South America, South Africa and New Zealand include *Pinus radiata*, *P. longifolia*, *P. betula*, *P. ponderosa* and *P. pinaster*. Pines cover 49% of the plantation areas in Argentina (especially *Pinus elliottii* and *Pinus taeda*), 78% in Chile (*Pinus radiata*) and 80% of the tropical forest plantations of Venezuela (*Pinus caribaea*).

Silvicultural management practices such as the selection of tree species and genotype, spacing, thinning type and its intensity, site preparation, fertilisation, irrigation and weed control, also generate new variables that significantly affect the growth and the quality of the wood, as shown by several studies carried out on Eucalyptus saligna [43]; E. grandis [44–46]; E. nitens [47]; E. grandis × camaldulensis [48]; E. globulus, E. grandis, E. tereticornis, E. urophylla and hybrid [49]; E. globulus [50]; clones of Populus trichocarpa Torr. & $Gray \times P$. deltoids and a native clone of Populus trichocarpa [21]; Populus \times euramericana, Populus alba and P. nigra [51]; Populus trichocarpa × deltoides [52]; Populus deltoides [40, 53]; and Populus nigra var. betulifolia [54]. Among conifers, silvicultural studies over the last decade have included: Pinus taeda [55-59]; Pinus resinosa [60]; Pinus sylvestris [61–63]; Pinus radiata [25, 64–66]; Picea glauca [67]; Picea mariana [68], Picea abies [62, 69-75]; Cryptomeria *japonica* [76] and various species [77].

1.3 Nonwood Plant Fibres

Despite wood fibres currently being the quintessential raw materials for the worldwide production of pulp and paper, the production of paper from wood is relatively new. At first, the paper was made exclusively of nonwoody plant fibres; however, growing concern for the environment has led to a rising interest in the use of nonwood fibres and crop residues as a source of raw material for this industry. Although there has been a dramatic increase in the use of recycled fibre, different sources have predicted an increase in the demand for virgin fibre [78]. It is then expected that the amount of nonwood fibres used in the pulp and paper industry will steadily increase in countries with a deficit of wood. The same trend can also be seen in countries rich in forest resources, where environmental and social pressures restrict the areas of forest available for timber production. It can therefore be anticipated that as the availability of pulpwood declines, nonwood fibres will partially replace wood. For this reason, despite the fact that this book is primarily on forest resources, it was decided to incorporate a brief overview of the use of nonwood resources in the pulp and paper industry.

The main types of nonwood plant materials used in the paper industry can be classified into three main groups:

- Natural growing plants (difficult to harvest: inefficient manually and costly with machines). This group includes bamboo, cane, reed, rush and so on.
- Agricultural waste (the most important and most widely used group). These fibres produce a variety of paper and paperboard. The problem is availability (limited to a time of year), so they need proper storage to avoid deterioration. This group includes straws of cereals and oilseeds (wheat, flax and rice) and industrial wastes (sugarcane bagasse).
- Industrial crops (plants especially grown for the stem/seed fibre content). The cost of growing, harvesting and handling limit their use to very special and good quality papers. This group includes bast fibres (flax, hemp, ramie, jute and kenaf), and fibres from fruits (cotton) and leafs (sisal).

Some of the main nonwood papermaking raw materials are cited below [79-82]:

• Sugarcane bagasse (*Saccharum officinarum*): Lignocellulosic waste from the sugar industry, its growth time is short, with a 1 year rotation. The major obstacle in pulping bagasse is the high pith content of the stalks (about 30% by weight of the stalk), so it requires depithing. The technology for production of most paper types out of bagasse is mature and it is increasingly exploited for this use. It can be used as a chemical and mechanical pulp.

- Bamboo (*Dendrocalamus strictus*): Mainly concentrated in Asia, and to a lesser extent in South America, it grows best in tropical and subtropical countries, but it can be found in almost every continent. Bamboo is usable in linerboard and in almost any other grade, and its fibre can be increased in paper furnishes without risk if bamboo pulp is available.
- Reed: The reed family includes reed (*Miscanthus*), amur silver grass (*Miscanthus Sacchaniflorsus*), Chinese silver grass (*Miscanthus Sikensis Anderes*), giant reed (*Arundo Donax Linn*) and others. Pulp is used in high-quality papers such as woodfree papers including copypaper with 70–80% reed pulp in the furnish. Even if reed is somewhat better than wheat straw pulp, its quality is still limited.
- Cereal straws: Wheat straw (*Triticum aestivum*), corn stalks (*Zea mays*), cotton stalks (*Goossypium*) and rice straw (*Oryza sativa*). These monocots have a short growth time with a one year rotation. The fibres are wastes of cereal harvesting. There are well-developed pulp technologies available and problems with black liquor treatment are being overcome. With high ash and low cellulose contents, cereal straws produce lower yields than bagasse with the same pulping conditions. It can potentially be used for most paper types.
- Kenaf (*Hibiscus cannabinus*): High yield dicot (21.3 Mt/ha in 3 years). Dual source of fibres: 57% long bast fibres, 41% short core fibres, therefore bark and core separation are required. Its potential as a raw material for papermaking has been known for a long time, with some industrial experience since the 1980s. Excellent papers are made using the existing technology. In blends of various proportions, it is used to make printing and writing paper, newsprint, multisack, linerboard, tissue paper, bleached paperboard, cigarette paper and other lightweight specialty papers.
- Jute (*Corchorus capsularis*): Bast fibres are characterised by their high cellulose content. Commercial jute consists of fibre bundles

of 1.8-3 m in length; individual fibres are 2-5 mm in length. In blends of various proportions, it is used to make printing and writing paper, and tag, wrapping and bag paper.

- Esparto grass (*Stipa tenacissima*): Fibres of esparto are fine and short, and produce a bulky, smooth, well-formed paper that is unsurpassed as the main ingredient for fine printing and lithographic paper.
- Ramie (*Boehmeria nivea*): Ramie fibres consist of pure cellulose, and are remarkable due to their length and width. They are the strongest and most durable of the vegetable fibres. Waste and short fibres are used for the manufacture of paper, and the average L and width is 120 mm and 0.05 mm, respectively.
- Hemp (*Cannabis sativa*): Hemp grows rapidly (8–10 ft in 4 months); its cultivation presents a high yield per hectare, requires little or no pesticides and herbicides, and allows crop rotation. There are well-developed pulp technologies available but it requires decortication and retting, and long fibres wrap around the equipment. In blends of various proportions, it is used to make specialty papers such as cigarette paper, lightweight printing and writing paper, condenser paper, and security and currency paper.
- Flax tow (*Linum usitatissimum*): It is an annual plant, cultivated in temperate climates for both its fibres and linseed oil. When planted densely for fibre production, the plant grows to a height of about 1 m. Bast fibres, known as linen, are separated from the inner bark by retting. In blends of various proportions, it is used to make specialty papers such as book paper, lightweight printing and writing paper, condenser paper, currency and security paper, and cigarette paper.
- Abaca, Manila hemp (*Musa textilis*): It is obtained from the leaf sheath of a plant grown mainly in the Philippines. In blends of various proportions, it is used to make specialty papers such as

superfine, lightweight, bond, ledger, currency and security paper, teabags, filters, linerboards, wrapping and bag paper.

- Sisal (*Agave sisalana*): Sisal leaves have an average L of 3.0 mm and average fibre width of 0.02 mm. In blends of various proportions, it is used to make superfine, lightweight, bond and ledger paper, currency and security paper, teabags, filters, publication paper, linerboard, wrapping and bag paper.
- Cotton fibres and cotton linters: Cotton fibres come from the seedpod of cotton plants, whereas linters are those fibres that remain on the pod after the long fibres of cotton have been removed for textile use. In blends of various proportions, fibres and linters are used to make high-grade bond ledger book and writing paper.
- Linseed flex: By-product of the linseed industry, this residue needs decortication before processing. The energy requirement for its cooking or thermomechanical pulping refining is low. It is used in wood pulp blends to improve strength, even if its brightness is low.

Pulp and paper from bagasse and other annual plants are mostly located in developing countries that use these plant species as priority crops for food, but without resources and defined policies for their efficient industrialisation. India and China are the undisputed leaders in the production of nonwood pulps [83]. Nevertheless, nonwood species are increasingly being considered as potential raw materials in very diverse countries as Finland [84, 85]; USA [86]; Bangladesh [87] and Uganda [88].

The Chinese pulp and paper industry has always been nonwood based. For many years, more than 80% of the domestic virgin pulp produced was made with nonwood fibre. However, the usage of nonwood pulp has been declining during the last couple of decades, as increasing amounts of wood pulp and secondary fibre have been used in the furnish. The share of nonwood fibre in the overall raw material furnish began to drop from 73% in 1980 to 40% in 2000, 33.7% in 2002 and 24% in 2005. Nevertheless, nonwood fibres will remain an indispensable fibre source for the growth of the Chinese industry. About 75–80% of nonwood pulp production is based on agricultural residue, 15-19% is made with agri-products, and 5–6% from long fibre nonwood species. Increases in reed, bamboo and wheat straw pulping capacities are expected [89, 90]. The share of nonwood fibre in papermaking furnish is likely to increase in the future only in the tissue paper sector (from 15 to 25%) [81].

India is endowed with rich nonwood fibre resources, in respect of diversity as well as abundance. They vary from essentially forestbased sources like bamboo to agricultural residues, i.e., bagasse and cereal straw. India is the world's largest producer of bagasse and the second largest producer of bamboo, next only to China, in both diversity and distribution; 125 species in 23 genera have been recorded (in which *Dendrocalamus strictus* accounts for 53%). However, the situation is not as impressive with regard to their utilisation. Bamboo is even better than hardwood in pulping characteristics. The only constraint is bulk and the resultant higher cost of transport. In view of the emphasis given to raising bamboo plantations and the success achieved in mass propagation techniques, it is expected that the availability of bamboo will increase in the future [91].

Nonwood crops such as flax, hemp, kenaf and jute contain both long bast fibres and short core fibres. Bast fibres – abaca, sisal and sabai fibres – are longer than or as long as softwood fibres in general. They are more slender than softwood fibres and have a lower coarseness value. This, together with the fact that long nonwood fibres are also strong, gives them excellent reinforcement ability. Reed, straw, bagasse, and some bamboo and esparto fibres are short. Their length as well as cell wall thickness (coarseness) is close to that of hardwood fibres. It should be remembered that nonwood raw materials contain so-called nonfibrous cells (parenchyma cells, known as fines). A high fines content together with a high hemicelluloses content causes difficulties in water removal from the pulp [92].

Because of their high silica content, the seasonality of growing and so on, problems related to the use of nonwood fibres include collection and transportation, storage and handling, washing, bleaching, papermaking and chemical recovery [93, 94]. However, the use of nonwood fibres is optimal for the production of corrugated board, printing and writing papers, and tissue. Cereal straw and sisal pulps remain excellent for cigarette paper production because of their high ash content.

Although nonwood fibres are normally associated with developing countries, these fibres play an important role in the global pulp production. During the period 1970–1995, the global trend in the pulp and paper industry showed a rapid growth of investment in the use of nonwood fibres. According to FAO, the total installed capacity of pulp from nonwood species was 7.62 million metric tonnes in 1970 and 24.862 million tonnes in 2000. Nonwood fibres represented 5.3% of the global production of pulp in 1983, whereas it was 12–15% in 2010. Total nonwood pulp production was 11,292 million tonnes in 2012, of which 5,916 million tonnes was pulp from straw, 1,227 million tonnes from bagasse, 1,774 from bamboo and the rest from esparto and other reeds and grass, cotton linters, flax, hemp, rags and other textile wastes [94, 95].

Traditional pulping processes applied to nonwood are soda and soda-anthraquinone processes, which are currently the most used [96–100]; although in recent years the research about the use of different types of organosolv processes has been intensified [101–107]. An application of prime importance nowadays is the use of these resources in biorefineries (fractionation of the raw material into different chemicals to produce high-value products and energy) but this is beyond the scope of this book. Some examples regarding the chemical composition, fibre characteristics, pulping processes and uses of nonwood raw materials are available in the literature [108–110].

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2 Anatomy, Structure and Chemistry of Fibrous Materials

2.1 Anatomy and Structure of Trees

The main part of the tree used for pulping is the stem. It has three main physiological functions, defined by the different kinds of cells found in the tree:

- Strength (to hold up the crown).
- Fluid handling (transport of water with dissolved minerals in an upward direction from the root to the crown).
- Storage (collecting and transporting of food reserves).

The functions of the support and transport of liquids are mainly carried out by elongated cellular elements oriented in the longitudinal direction of the stem, whilst the storage function is performed by small cells longitudinally and radially oriented.

The tree log is composed histologically of three parts: xylem or wood, cambium and bark. The pith, formed by the initial cellular elements of plant growth, is located in the centre of the stem. In old trees, the wood structure is generally denser and darker in the inner part (heartwood or heart) than in the outer part (sapwood). In a transversal cut of a tree log, a cross structure can also be observed, corresponding to the rays. Trees that grow in temperate latitudes, where the growing seasons of spring and summer are followed by periods of rest in winter during which no growth occurs, form a defined layer of wood each year, outside the preceding one. In these trees, a layered structure is distinguishable, each layer being constituted by the wood formed during one year. These structures are

called: growth rings, annual rings or annual layers, and are indicative, in general, of tree age. This effect is more pronounced in conifers. Each growth ring contains, as mentioned above, cells corresponding to earlywood (EW) or springwood and latewood (LW) or summerwood.

Conifers are more primitive and less evolved than angiosperms. They have a single type of longitudinal cell (named tracheid) which performs two of the physiological missions of the stem: transport and support. Wood in conifers consists of more than 90% longitudinal tracheids, unlike hardwoods, in which the fibre content varies between species, but generally ranges between 40-80%. Some conifers have a different kind of small cell named transverse tracheids or ray tracheids.

In contrast to conifers, broadleaf species have a particular heterogeneous structure. As a result of greater specialisation, they have cells responsible for mechanical support (libriform fibres), other types of cells to transport liquids (members of vessels), and several intermediate forms of transition elements (fibre-tracheids, vasicentric tracheids) whose function is both, transport and support. The vessel cells are tubular elements that are interconnected to form long vessels. An example of the different cell elements present in eucalyptus trees is shown in Figure 2.1.



Figure 2.1 Different kinds of cells present in *Eucalyptus*. Reproduced with permission from C.E. Núñez, *Revista de Ciencia y Tecnología*, 2009, 11, 11, 26. ©2009, Facultad de Ciencias Exactas, Químicas y Naturales, Universidad Nacional de Misiones [1] In a living tree, the sapwood is responsible for the storage and synthesis of biochemicals. The primary storage forms of photosynthates are starch and lipids, often stored in the parenchyma cells of the sapwood. Living cells of the sapwood are also the agents of heartwood formation. Biochemicals must be actively synthesised and translocated by living cells. For this reason, living cells at the border between heartwood and sapwood are responsible for the formation and deposition of heartwood compounds. Heartwood functions in the long-term storage of biochemicals of many varieties, known as extractives, which are a normal part of the plant's system of protecting its wood. Extractives are formed by parenchyma cells at the heartwood-sapwood boundary and are then exuded through pits into adjacent cells. In this way, dead cells can become occluded or infiltrated with extractives (tyloses).

General and more in-depth information regarding plant anatomy, particularly the species of the northern hemisphere, can be found in [2–8], whereas extensive information on hardwoods, especially *Eucalyptus* and *Acacia* can be found on the website Eucalyptus Online Book & Newsletter [9].

2.2 Chemical Composition of Wood

Cellulose, hemicelluloses and lignin are the main components of wood. Lignin is an amorphous, aromatic, water insoluble, heterogeneous, three-dimensional and crosslinked polymer. It can be considered as having been formed by random copolymers derived from unsaturated alcohol derivatives of phenyl-propane. Lignin is not composed of exactly repeating units and can be more exactly described as a macromolecule than as a polymer; phenylpropanoid units are linked together by more than 10 different types of aryl ether and carbon-carbon linkages. In addition to methoxyl groups, lignin has other functional groups, including phenyl hydroxyl, benzyl alcohol and carbonyl. The function of this network is to provide a mechanically strong composite material with cellulose fibres. Hemicelluloses are a mixed group of both linear and branched heteropolymers mainly comprising of five monomeric sugars, namely D-glucose, D-mannose, D-galactose, D-xylose and L-arabinose. They are linked together mostly by β -1,4-glycosidic bonds, but β -1,3-; β -1,6-; α-1,2-; α-1,3-; and α-1,6-glycosidic bonds can also be found. The degree of polymerisation (DP) of hemicelluloses is 100 to 200. which is much lower than that of cellulose. The major hemicelluloses of hardwoods and gramineous plants are glucuronoxylan and glucomannan, as well as galactoglucomannans, arabinoxylans and glucuronoarabinoxylans; whereas the main types of softwood hemicelluloses are galactoglucomannans, arabinoglucuronoxylan and arabinogalactan. Cellulose is a polymer consisting of linear β -(1-4) D-glucopyranosyl units. The cellulose chains have a strong tendency to aggregate into highly ordered structural entities, due to the uniformity of their chemical constitution and spatial conformation. The DP of cellulose present in a living tree is unknown, but the size of the native molecule is often stated to be 5,000–10,000 glucopyranose units [10].

The interactions of the polymers govern the performance of the cell wall assembly (besides their individual properties). The bonding of hemicelluloses to cellulose fibril surfaces is not based on covalent bonds but mainly on hydrogen bonds. This results in a strong but flexible connection of both polymers, as hydrogen bonds can easily be opened and reformed. Lignin is not bound directly to cellulose, but it is covalently bound to hemicelluloses. Hence, the amphiphilic hemicelluloses play an essential role in the maintenance of the cell wall assembly. These polymers mediate between the hydrophilic and stiff cellulose, and the more hydrophobic and pliant lignin [11].

The content of cellulose, hemicelluloses and lignin in typical plantbased raw materials is 40–50, 15–35 and 20–40%, respectively. Depending on the species, hardwoods and softwoods also have varying amounts of extractives (1–8%, including fats, waxes, alkaloids, proteins, gums, resins, starches and so on) and ash (approximately 0.8%). Due to the regularity of chains, cellulose fibres are characterised by a relatively high degree of crystallinity. The physical and chemical behaviour of the supramolecular structure of cellulose microfibrils prove that it is characterised by alternations of ordered crystalline regions and amorphous regions along the chain axis. The DP of cellulose chains and the degree of crystallinity of cellulose microfibrils depend upon the nature and origin of the vegetable fibres. More details on the chemistry of wood can be found in classical books of the specialty [12–16].

The fibrous elements of trees (tracheids and fibres) consist of a cell wall surrounding a central cavity. Although there are differences between the fibrous elements of softwoods and hardwoods, in all cases the ultrastructure (detailed structure that can be observed by electron microscopy) of the cell wall is nearly identical. The wood (secondary xylem) is biosynthesised by a succession of five major steps, including cell division, cell expansion (elongation and radial enlargement), cell wall thickening (involving cellulose, hemicelluloses, cell wall proteins, and lignin biosynthesis and deposition), programmed cell death and heartwood formation [17]. Heartwood extractives are formed at the heartwood periphery, from stored carbohydrate or carbohydrate translocated from the cambial region. Heartwood by definition ceases to contain living cells, although in some situations parenchyma cells can retain cytological and physiological activities [18].

2.3 Cell Wall Structure

The cell wall is originally thin and delicate, but it is modified as the cell matures, increasing in thickness and changing its chemical nature and physical structure. It is composed of several layers that are fabricated at different periods during cell differentiation. Young cells are connected by thin primary cell walls, which can be contracted and stretched to allow subsequent cell elongation. After cell elongation ceases, the cell wall is generally thickened, either by the secretion of additional macromolecules into the primary wall (P) or, more usually, by the formation of a secondary cell wall composed of several layers. In mature tissues such as xylem, the cell dies, leaving only the cell wall [19].

The observation of mature, thick cell walls, evidence a layered structure with concentric arrangement. The classical and best known models of the lamellar structure of the fibre wall have been proposed

by Scallan [20], and by Kerr and Goring [21]. There is a lignin-rich area between contiguous cells called the middle lamella (ML), which ensures the adhesion of a cell with its neighbours. The P also contains a high amount of lignin and is very thin. The secondary wall has a three-layered structure, differentiated by the different orientation of the crystallite regions within the three layers: the thinnest and secondary wall outermost layer (S1), the secondary wall middle layer (S2) and the secondary wall innermost layer (S3) [22, 23].

The smallest cellulosic strand, with an average width of 3.5 nm in the mature cell wall, is termed an 'elementary fibril' [13]. These fibrils, in turn, are organised into strands known as 'microfibrils' (5–30 nm wide), which are visible using an electron microscope. The diameter of microfibrils depends on the source of cellulose and the positions of the microfibrils within the cell wall. Microfibrils are combined into greater fibrils and lamellae, and are embedded in a hemicelluloses-lignin matrix within the cell wall in the fibre wood [6]. Other authors consider that the microfibril is the most fundamental, well-defined morphological entity, although it consists of nonuniform subunits [24].

The cellulose microfibrils are oriented in different directions in the different fibre wall layers, and the angle between the fibre axis and the microfibrils (microfibril angle (MFA)) has a profound effect on the mechanical properties of wood and thus on products derived from it. There are many methods available for measuring the MFA including: X-ray diffraction, orientation of cross-field pit apertures, polarised light, iodine staining, ultrasonic checking, confocal microscopy using birefringence, polarisation confocal microscopy using difluorescence, and measuring the orientation of cavities produced by soft rot fungi in wood cell walls [25, 26].

MFA orientation has an important influence on the physical properties of a wood fibre. The primary cell wall is made up of several layers of microfibrils arranged randomly, and embedded in pectic substances, lignin and hemicelluloses. Within the secondary wall, the layers S1, S2 and S3 differ with respect to the orientation of microfibrils. The S1 layer represents 5-10% of the total thickness of the cell wall and the angle of its microfibrils, with regard to the cell axis, is $60-80^\circ$. The S2 is the thickest layer of the cell and the most important in terms of mechanical strength. It accounts for 75-85% of the total thickness of the cell wall (90% of the cell wall in LW tracheids and libriform fibres), and its MFA is $5-30^\circ$ with regard to the cell axis. The S3 layer of the secondary cell wall is relatively thin, with a microfibril angle of $60-90^\circ$ [17]. Although the nature of the S3 layer still requires further investigation, studies made on a 17 year-old *Pinus radiata* showed that the microfibrils in the S3 layer fall from angles of over 80° to 54 and 51° for radial and tangential cell walls at the top of the butt log [27].

Technologically, the term MFA is usually applied to the orientation of cellulose microfibrils in the S2 layer which composes the greatest proportion of the wall thickness, since it is the factor that most affects the physical properties of wood. MFA is considered one of the main determinants of wood quality and it is considered to be a significant factor influencing the mechanical properties of clearwood (wood produced without any knots or other defects), especially the modulus of elasticity and shrinkage anisotropy. The MFA in individual tracheids has been related to the tensile strength and elastic properties of pulp fibres; a small MFA results in greater tensile strength while a large MFA results in greater extensibility. MFA is also known to influence the fracturing properties of wood with small angles favouring transwall as opposed to intrawall fracturing on tangential longitudinal surfaces under transverse shear [28]. An extensive review of cell wall features with regard to the mechanical performance of fibres has been published recently [11].

One of the most important biochemical events accompanying the formation of the secondary plant cell wall is the deposition of lignin. Lignification is one of the final stages of xylem cell differentiation, where lignin is deposited within the carbohydrate matrix of the cell wall by infilling of interlamellar voids, and at the same time, formation of chemical bonds with noncellulosic carbohydrates. Lignification of the ML and P typically begins after the start of secondary wall formation, while lignification of the secondary wall usually begins

when secondary wall formation is complete. The region of the cell wall named 'compound ML' contains the highest lignin concentration (weight/weight) whereas the S2 region contains the lowest. However, because the secondary wall has a much greater volume than the relatively thin ML, most of the lignin in wood is located in the secondary wall. It is thus important to differentiate between lignin concentration (within a cell wall region) and lignin content (a measure of lignin in bulk samples which is thus dependent on variations in anatomy) [29]. A demonstrative scheme of the percentages of major components of the cell wall layers is presented in Figure 2.2.



Relative oven-dry mass %

20% lignin + 40% hemicelluloses + 40% cellulose 20% lignin + 30% hemicelluloses + 50% cellulose 50% lignin + 20% hemicelluloses + 30% cellulose 70% lignin + 20% hemicelluloses + 10% cellulose 100% lignin + pectin

Figure 2.2 Demonstrative scheme of the percentages of major components of the cell wall layers

The distribution of the components in the cell wall, called topochemistry, is of great importance since it affects the reactivity, degradation and properties of the timber. For example, lignin distribution has been intensively studied in spruce and birch [30–33], and more recently in hybrids, clones and genetically modified species of *Eucalyptus* and *Populus* [34–39]. In addition to lignin, the topochemical distribution of phenolic extractives in woody tissue at a cellular level can also be determined by a new technique known as scanning ultraviolet microspectrophotometry [40–43].

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3 Abnormal Wood

3.1 Reaction Wood

When the tree is subjected to abnormal stress (e.g., gravity, persistent winds or other causes) it is pushed from its original direction while it tries to regain its correct orientation. As a result of the tree's reaction to disturbance, it develops an abnormal tissue named 'reaction wood'. Reaction wood in conifers (compression wood) forms on the lower side of a leaning or bent stem/branch, whilst in hardwoods (tension wood) it forms on the upper side. The tissue on the opposite side of the stem is named 'opposite wood'. The abnormal properties of reaction wood are usually undesirable for commercial use, especially when it is severe [1]. The structure of reaction wood and the composition of its elements are different in softwoods and hardwoods. In an individual tree, the reaction wood can be of considerable importance, but the disturbing effects of varying amounts of reaction wood should disappear in the large daily wood supply of a modern pulp mill. Pulps from compression wood fibres are definitely of lower quality than those from normal fibres, whereas tension wood fibres are considered to be quite acceptable [2].

In typical compression wood (Figure 3.1), annual layers are abnormally broad, so that the continued formation on one side of the trunk produces abnormally wide annual rings, resulting in an eccentric growth. Extreme forms of compression wood have large proportions of latewood (LW) which generates an abnormally high density. The tracheids in the compression zone of conifers are shorter and their walls are more lignified, denser and darker than those of normal wood. The compression tracheids are round and generate a large number of intercellular spaces. As the secondary

wall middle layer (S2) is very thick and its microfibril angle (MFA) is approximately 45°, compression tracheids can shrink and swell abnormally in the direction of the axis of the tracheid, and so, compression wood can shrink and swell abnormally along the grain [3].



Figure 3.1 Compression wood in a conifer stem (Misiones, Argentina)

Kraft pulp from *Pinus sp.* compression wood has been found to have a slightly higher kappa number, coarser fibres and greater amounts of a residual lignin-carbohydrate complex, containing galactans, than pulps from normal wood [4]. The compression wood tracheids exhibit a significantly decreased mean length and width, and higher coarseness than those of normal wood. Furthermore, the cell wall of compression wood has a lower content of glucose, mannose, xylose and arabinose, whereas it contains higher levels of galactose and lignin than normal wood. In addition, the content of phenolic and aliphatic hydroxyl groups within the lignin is slightly higher in compression wood [5]. Studies on *Picea abies* have demonstrated that the fibril angle of compression wood was much higher than that of normal mature wood (MW) [6]. More recently, the MFA of normal MW, juvenile wood (JW), opposite wood and compression wood of one branch of *Picea abies* (L.) from two different origins were compared. In all types of tissue studied, the cellulose MFA varied between 5-47°, but only compression wood samples showed an MFA above 25°. For all other tissue types, the MFA varied between 5-15° [7].

In a comparison of the mechanical properties of normal and compression wood in Norway spruce (*Picea abies*) it has been proposed that the observed high lignification in compression wood increased the resistance of the cell walls to compression failure. An increased percentage of *p*-hydroxyphenylpropane units in compression wood lignin have also been detected, which might contribute to the comparably high compressive strength of compression wood [8].

Reaction wood formation occurs when the plant perceives a change in the gravity vector or in the light environment, and so it induces an adaptive programme of cell division, cell wall modification and cell morphology changes that lead to organ repositioning. The reaction wood in woody dicotyledons (hardwoods), called tension wood, is usually formed in the upper side of leaning stems and branches, induced by decreased levels of the hormone auxin. Its location shows a gravity response that is opposite to that of compression wood in conifers. In contrast to compression wood, it has a greater proportion of cellulose and less lignin than normal wood. It is a lighter colour and becomes clearer when the logs are exposed to all types of weather [9]. In comparison with regular fibres, tension wood fibres in many species show an additional characteristic structural feature called the gelatinous layer (G-layer), which is believed to be an operational part of the tension wood fibre. The G-layer in tension wood fibres is an additional layer on the lumen side which can replace the secondary wall innermost layer (S3) or even the S2-layer of the secondary

cell wall, and can even fill the whole lumen of the tension wood fibre. The cellulose fibrils are oriented parallel to the axial direction (MFA $\cong 0^{\circ}$) and have a higher crystallinity than the cellulose fibrils in the secondary wall [10].

Tension wood in *Eucalyptus camaldulensis* has a higher content of alpha-cellulose and extractives, and a lower content of hemicelluloses and lignin than normal wood. The cell wall structure of the tension wood fibres is that of a secondary wall outermost layer + G-layer type. In gelatinous fibres the MFA is small and the fibres are long. Abnormal wood also has a reduced frequency of vessels [11].

The density and MFA of tension wood and normal wood have been assessed in the sapwood and heartwood of 10 year-old *Eucalyptus globulus* Labill from three different locations. Sapwood samples with high percentages of tension wood exhibited high density. The MFA was found to be very low in normal wood within the sapwood, so the tension wood could not be identified using the MFA [12].

An interesting anomaly has been found in *Acacia sp.*, in which tension wood has appeared in straight logs grown vertically. Continuous dark bands of about 5 mm width have been found to extend radially from the inner heartwood, in a spiral pattern, separated by continuous normal wood bands of similar width. The tension wood bands continued in the sapwood from the heartwood boundary to the cambium but were not coloured. The dark colour of the tension wood was probably caused by extractives. The MFA and cellulose crystallite width was of similar intensity throughout the band of tension wood, indicating similar microstructural characteristics at different cambial ages [13].

Typical values of crystal diameter (3.1 nm) and a relatively large MFA (20°) have been found in the secondary wall layers of single tension wood fibres of poplar (*Populus maximowiczii*). In the G-layer, the mean deviation from perfect parallel alignment was as small as +2.70/-0.05° and the diameter of the cellulose crystals (microfibril cross-sectional area) was fourfold higher than that found in the rest of the secondary cell wall [14].

The G-layer is mainly composed of cellulose formed into macrofibrils and other minor components including pectins, hemicelluloses and lignin. The size and angle of the cellulose structures found in the G-layer, and its interaction with the minor components, may all contribute to explaining the G-layer's physical properties. Studies on aspen plants (*Populus tremula* × *tremuloides*) demonstrated that the concentration of xyloglucan was higher at the interface between the S2 and the G-layer, supporting the hypothesis that xyloglucan serves to anchor the G-layer to the inner S2-layer, and so to the rest of the secondary cell wall [15].

3.2 Juvenile Wood

Little importance was given to the distinction between different zones of the tree stem in the past, as most of the wood used industrially came from adult trees sourced from natural forests. At present, however, the supply of mature trees with large diameters from native forests is constantly decreasing. To satisfy the increased demand for forest products, much of the future timber supply will come from plantations. The use of short-cycle trees has become common practice, through the adoption of species that grow relatively fast, such as Pinus and Eucalyptus. This resource will tend to be harvested in short rotation cycles and will consequently contain higher proportions of IW than the currently harvested lumber [16–17]. Important artificial regeneration programmes, breeding activities and research on tree improvement have also been devoted to black spruce (Picea mariana) in recent years, so it is assumed that large-scale reforestation with fast-growing genotypes is likely to shorten spruce rotations in Canada in the near future [18].

JW is commonly defined as the wood portion that extends outward from the pith, where wood characteristics undergo rapid and progressive changes in successively older growth rings. Older wood, beyond the juvenile core, has been referred to as MW, adult wood and outer wood. The precise determination of the transition age between JW to MW is difficult because transition is gradual, not abrupt. During a 'transition' period from 5–20 years-of-age (depending on the species), wood characteristics improve until they become relatively constant (MW). JW has significantly lower strength and stiffness, more longitudinal shrinkage, and less radial and tangential shrinkage than MW.

IW, corresponding to the central region of the tree, is present in an approximately uniform pattern throughout the diameter, extending from the base to the top, and may form part of the sapwood and heartwood in the trunk, when the latter is already present in the tree [19]. The cambium is a continuous sheath around the stem: in large stems it produces JW near the tree top and MW at its base. In softwoods, IW later becomes the inner part of the heartwood. There is no clear change from core wood to outer or MW within one year as this change occurs over several years. Heartwood formation and growth stresses may change the properties of JW, and decrease the shrinkage in the lower part of the stem [20]. In very old trees, containing a large amount of heartwood, JW can clog with tyloses [21]. Heartwood formation in Slash pine might begin as early as eight years old, whereas it might be delayed until 30 years-of-age in Loblolly pine in the south of the USA. Therefore, the heartwood in Loblolly pine is of little significance during a rotation age of 25-45 years. Once initiated, heartwood formation gradually spreads outward in rings from the juvenile core and beyond. The more rapidly growing trees have been found to contain a wider band of sapwood than the slower growing trees [17].

A schematic drawing of the existence of JW and MW at two ages in conifer trees from plantations is shown in Figure 3.2. A comprehensive review of alternative conceptual patterns for different variables, which change along the tree in softwoods and hardwoods, can be found in reference [22].



Figure 3.2 Schematic drawing of the existence of JW and MW at two ages in conifer trees from plantations

Both in softwoods and hardwoods, JW cells are generally smaller than those of MW. They could be 3–4 times smaller in softwoods, and twice as small in hardwoods. In addition, the cell structure is also different with JW containing a greater proportion of thin-walled cells. These characteristics make the density and resistance in JW lower than in MW [19]. Compared with MW, in conifers JW exhibits: lower strength, higher longitudinal shrinkage, lower specific gravity, lower proportion of LW, higher percentage of compression wood, thinner cell walls, lower cellulose content, higher lignin content and greater fibril angle. Young trees are presently harvested mainly for pulp because they are unsuitable for use as high-grade structural timber [23].

Furthermore, in conifers, the MFA is large in JW and small in MW. The MFA is larger at the base of the tree for a given ring number from the pith and decreases with height, increasing slightly at the top of the tree. Similar patterns occur in hardwoods, but with much less variation and much smaller MFA in JW [24]. The large MFA in fibres of JW of conifers implies low stiffness and therefore, low economic value.

The transition age varies among phytogeographic regions. Studies on the age of demarcation based on the MFA, on plantations of 20–27 year-old *Pinus taeda* L., showed that trees in southern USA had a smaller proportion of JW (8.4–10.4 years) than in northern USA (10.5 to over 20 years) [25].

Studies regarding the impact of spacing on plantations of white spruce (*Picea glauca*) demonstrated that properties differ between JW and MW. The juvenile-mature transition period ranged from 8–20 years. Wood properties in the JW zone were more affected by spacing than those in MW, especially the growth rate. For all studied plantation spacing, the growth rate in JW was greater than that of MW [26].

Morphological analyses of a 35 year-old mature *Pinus taeda* demonstrated that fibre lengths (L) of the JW samples are significantly shorter than those of MW samples, whereas MW tends to have a smaller fibre width (W) than JW. The L of JW from the top of the tree is shorter than that from the bottom of the tree, showing similar tendency to the W. The MFA of the JW at the bottom of the tree is higher than that from the top of the tree. The crystallite size of normal JW from the top of the tree is slightly thicker but shorter compared with that of JW from the bottom of the tree. In contrast, the differences in fibre coarseness and chemical structure were not as significant between groups [27].

An example of the mean L evolution, from pith to bark, in a 20 year-old *Eucalyptus grandis* is shown in Figure 3.3. As rings are not always evident in hardwoods, the author defined the 'theoretical

annual rings' depending on differences in the tissue [28]. It is clearly seen from the figure that JW seems to extend up to about 6–9 years, and MW from 15 years and older.



Figure 3.3 Evolution of L with age in *Eucalyptus grandis* (Argentina). Reproduced with permission from C.E. Núñez, *Revista de Ciencia y Tecnología*, 2011, 13, 15, 25. ©2011,
Facultad de Ciencias Exactas, Químicas y Naturales, Universidad Nacional de Misiones [28]

The juvenile-mature correlation in MFA enables its use for clone selection, finding superior genotypes and improved wood stiffness at a very young stage. The measurement of MFA in even very young trees, at the 2nd ring from the pith at breast height, has been demonstrated to be very effective in this regard when applied to 20 year-old *Cryptomeria japonica* trees [29].

The segmented regression method has been increasingly employed lately to identify the transition point of JW to MW [30].

Some findings reported in the last 12 years regarding the transition between JW to MW, in different species of conifers from natural and planted forest, are shown in Table 3.1 and Table 3.2. The same information for broadleaf species is presented in Table 3.3.

Table 3.1 JW	V to MW tran	sition in different conifer	species from natural forest, organised by age	e and gender
Species	Age (years)	Origin	Measurements	Reference
Picea abies	>300	4 sites of natural forest in Poland	Old spruces had three types of wood in their stems: JW, maturing (transition) and MW	[31]
			Taller trees and those with larger dimensions have considerably more JW than the lower and thinner spruces	
			The ratio between the widths of the LW to earlywood zones was used as the basis for the isolation of JW, transition and MW zones	
Pinus nigra	154	Natural forest in the Iberian Peninsula	Strong increase of the average density along JW (93 kg/m ³), reaching its maximum density at the end (547 kg/m ³)	[32]
	176		In MW (cambial age >50 years) the density varies only with a slight average decline (19 kg/m ³) in 80 years	
Pinus contorta	<120	Four sites of natural forest in British	The age of MW transition based on L characteristics was 18 ± 4.7 years	[33]
		Columbia, Canada	Employing MFA as an indicator, the transition was at an age of 15 ± 6.9 years	

[34]			[35]	[36]	
JW density: 410 kg/m ³ MW density: 516 kg/m ³	JW density: 391 kg/m ³ MW density: 552 kg/m ³	JW density: 573 kg/m ³ MW density: 652 kg/m ³	JW extends from the 15–17 th rings MW extends from the 70–75 th rings	The mean wall thickness of spruce tracheids rapidly increase in the first 20 growth rings, and then stabilised at a value slightly below 4 µm	Like spruce, wall thickness of pine tracheids increase rapidly for the first 20 growth rings, but then seem to go through a peak around growth ring 40 The perimeter of pine tracheids increase rapidly for the first 20 growth rings, and then continued to increase at a constant but slower rate
Natural forest in the Czech Republic			Natural forest in Wienerwald, Austria	Natural forest in Norway	
115	98	80	06	80	84
Picea abies	Pinus sylvestris	Larix decidua	Picea abies	Picea abies	Pinus silvestris

Table 3.2 JV	V to MW trai	nsition in differen	nt conifer species from planted forest, organised by age at	nd gender
Species	Age (years)	Origin	Measurements	Reference
Picea abies	40	Plantation in north Sweden	Average length and diameter of fibres was less affected by enhanced growth rate in JW (at 4 m height) than in MW (at breast height)	[37]
			JW has a narrower range of intraring variation of fibre properties than MW, although the change is faster in JW near the pith than in the outer rings	
Picea mariana	50	Plantation in Victoriaville,	Proposes to base the selection criteria on two parameters: wood density and growth rate	[18]
		Quebec	The age of transition of JW to MW is lower in slow- growing trees than in fast-growing ones	
			The ring density and LW proportion in annual rings of JW is high near the pith (more than 30%), decreases	
			to a minimum in the transition zone (at the 7^{th} ring), and then increases slowly but steadily to MW	
Pinus taeda	35	A tree from Orange	JW fibres tend to have larger width, smaller length and higher MFA than MW	[27]
		County, NC, USA	L, W and MFA of JW are lower at the top than at the bottom of the tree	
			At the bottom of the tree, the fibre coarseness of JW is slightly smaller than that of MW	

inus	25 and 28	Plantations in	L started to differentiate between trees at annual rings [36]	[9]
lvestris		north Sweden	13–15, and after the 16^{th} ring there is a slight tendency towards stabilisation (transition from JW to MW)	
			Proposes to use L from the annual ring around 15–20	
			and mgnet, at a tree neight from 0.5–1.5 m, for genetic analyses of L in Scots pine progeny tests	
inus patula	16	Plantations	The proportion of LW showed a gradual increase up [38]	8]
		in Veracruz,	to 14 years and faster thereafter	
		Mexico	The wood density showed a decrease in the early years	
			and then a gradual increase, levelling off at 12-14	
			years	
			Tracheid length increased progressively and stabilised	
			after 8-10 years	
			A segmented regression analysis using the average	
			annual values of tracheid length showed a wide	
			Variation detween trees in the transition age of 0-10	
inus caribaea	5715	Plantations in	June 1990 June June June June June June June June	61
	20 and 25	Nigeria	variability in wood properties, shorter cell length and	_
		0	thinner wall thickness with greater flexibility	
			MW (aged 20-25 years) is more stable in its	
			properties with higher pulp yield, longer cell length	
			and thicker cell walls	

Table 3.3 JW t	o MW transi	tion in different broad	dleaf species from natural and planted forest, orga	nised
		by age	e and gender	
Species	Age (years)	Origin	Measurements	Reference
Acer saccarum and	140	Natural forest in	The transition from JW to MW wood appears	[21]
Acer rubrum		Canada:	at 40 years	
		 Ottawa 	There is little difference in pulp properties	
		 Montreal 	between JW and MW pulp	
			The JW of very old trees (>100 years) contains	
			high levels of blockages and creates significant	
			processing problems	
Fagus sylvatica	83	Natural forest in	Average vessel diameter, width and height of	[40]
		the Czech Republic	pith rays reach statistically lower values in JW	
			than in MW	
			The number of rays/mm ² is three times higher	
			in JW than in MW	
			JW density: 726.07 kg/m ³	
			MW density: 701.50 kg/m ³	
Betula pendula	40	Waldsieversdorf,	L increases and MFA decreases from pith to	[41]
		Germany	bark	
			JW (from pith to the 15 th ring) contains fibres	
			up to 1.2 mm in length with MFA from 18 to	
			12°	
			MW (15th ring to outermost rings) contains	
			fibres up to 1.43 mm in length with a maximum	
			MFA of 12°	
Eucalyptus	32	São Paulo, Brazil	L increases in the radial direction from about	[42]
citriodora			0.7 mm at the pith to 1.3 mm at 45–55 mm	
			towards the bark. The transition from JW to	
			MW occurs at that point	

Eucalyptus grandis	9, 16–18	Plantations in:	Trees from plantations in tropical and	[43]
		 Three sites in Brazil 	subtropical climate sites have an MFA above and helow 10°, respectively	
		One site in	The lower growth rates in the subtropical	
		Argentina	plantations increase the tension wood with	
			smaller MFA	
Eucalyptus nitens	15	New Zealand	MFA decreases with height, reaching a	[44]
			minimum at 30-50% of the stem height before	
			increasing again toward the crown	
Eucalyptus	15	Australia	MFA remained constant with height in the	[45]
globulus and			range 0 to 13° with only 5° of difference	
Eucalyptus nitens			between the inner and outer parts of the stem	
Eucalyptus	14	Progeny test in	The mean value of MFA was 12.5° (min: 7.7°	[46]
urophylla		Republic of Congo	and max: 19.7°), the maximum values were at	
			the pith and the minimum at the bark	
Acacia mangium	11	Plantation in	In both, xylem maturation generally depends	[47]
and		Malaysia	on diameter growth. MW formation starts after	
A. auricuriformis			a certain diameter is reached, suggesting that	
Paraserianthes	7 and 8	Plantation in	accelerating lateral growth from an early stage	
falcataria		Indonesia	will produce MW as soon as possible	
Eucalyptus	11	Australia	In Eucalyptus spp., xylem maturation is	[47]
globulus			controlled by cambium age. MW formation	
			starts once a certain cambium age is attained.	
Eucalyptus grandis	14		This suggests that it is important to arrest	
			lateral growth at an early growth stage and	
			then accelerate it after reaching a certain	
			cambium age	

Eucalyptus grandis × urophylla clones	8	Plantations in four sites in Brazil	Overall mean of MFA was 8.8°, but significant differences between sites and between clones were found	[48]
Polupus deltoides cv. '129-60'	17	Plantations in Buenos Aires, Argentina	 Density increases significantly with tree height, from 0.368 g/cm³ to 0.413 g/cm³ at 15.6 m Density also increases with age, especially in the first 11–12 years: 0–3 years: 0.353 to 0.364 g/cm³ 3–12 years: 0.390 to 0.400 g/cm³ 	[49]
Populus clones	11 and 12	Plantations in China	MFA shows a decreasing trend from pith-to- bark and also declines rapidly with height Mean clonal MFA at the breast height ranges from 15.2 to 21.0° It varies from 7.8 to 28° between growth rings at breast height	[50]
Populus clones	6	Plantations in Washington, DC, USA	Wood density at 1.5 m is 0.37 g/cm ³ during the first 3 years. It decreases somewhat at age 4 or 5 years and then increases to an average of 0.45 g/cm ³ at 9 years L increases from 0.57 mm at age 1 to nearly 1.0 mm at 9 years	[51]

In an extensive review that defines and discusses the extent, occurrence and characteristics of JW in pine plantations (mostly *Pinus taeda*), from southern USA, the authors define so-called 'transition' tracheids, which are neither springwood nor summerwood and often comprise the major part of extremely wide juvenile rings. The wide zone of transition tracheids is not only a distinguishing characteristic of JW but also a major determinant of wood quality. In mature Loblolly pines, the MFA is small, averaging about 5-10°. In JW, the fibril angle is large, averaging 20-35° and often up to 50° in rings next to the pith, decreasing outward in the juvenile core. The decrease in fibril angle often continues well beyond the juvenile core. As a general rule, fibril angles are larger in wide rings than in narrow ones [17].

A schematic representation of the variation of MFA at different ages or in different parts of the stem, in normal and reaction wood is shown in Figure 3.4.



Figure 3.4 Representation of the variation of S2 MFA in normal wood at different ages or in different parts of the stem, and of G-layer MFA in reaction wood

As shown in the figure, the MFA in JW is more comparable to the MFA in compression wood than that in normal wood. Nevertheless, JW is different from compression wood in morphology and chemical characteristics, such as tracheid shape and length, sugar composition and lignin structure.

When compared with normal MW of *Pinus taeda*, juvenile and compression wood result in a higher consumption of chemicals during pulping and lower pulp yield. Moreover, differences in L between the juvenile compression wood and mature compression wood have not been found, although juvenile compression wood fibres were thinner than those of mature compression wood [27].

In kraft pulping trials of 8 year-old *Populus hybrids*, the authors concluded that at this age the papermaking properties do not change significantly. This conclusion can be further corroborated by the fact that once the trees are 8-years-old, the amount of wood containing long fibres (0.9 mm and longer) is approximately four times in volume compared with the wood containing short fibres (0.7 mm) [52].

The property profile of JW is somewhat less advantageous for chemical pulp production in most cases. An important exception is eucalyptus pulps where MW gives fibres which are too coarse to compete with JW [2]. In a study of kraft pulps of 15 year-old *E. nitens* and *E. globulus*, the authors found a 5° variation in MFA between the inside and the outside of the stem, concluding that it could not be the reason for the difference in sheet properties [45].

Studies on *Populus tremuloides* kraft, neutral sulfite semichemical, thermomechanical pulping (TMP) and chemithermomechanical pulping processes, demonstrated that pulps produced from chips of JW were of somewhat lower quality than those from MW [53].

The disadvantages of pine species, especially the southern pines, for mechanical pulping include: large thick-walled fibres, high fibre coarseness, high extractives content and low brightness, and can be reduced by choosing juvenile southern pine wood (for example, thinnings). Brightness may be improved and pitch problems ceased by selecting younger trees with a lower proportion of coloured, resinous heartwood [54]. Comparison of the performances of *Pinus taeda* JW and MW in unbleached TMP showed the sheets made from TMP of Loblolly pine JW exhibited superior strength

and optical properties than those from MW. The results also show that screened TMP of JW consumed a large amount of electrical energy to produce a long-fibred pulp with a low fines content and low coarseness, compared with MW [55]. In the case of stone groundwood pulps from *Picea abies*, the sheets made from JW pulp also showed improved light-scattering properties, tear and tensile strength, and higher sheet density, compared with those formed from MW pulp. Differences were likely related to the manner of fibre processing and development at the ultrastructural level [56].

When refining JW, the thin cell walls of springwood fibres break down more readily than the thick cell walls of summerwood fibres. This results in a pulp with lower strength characteristics and undeveloped coarse fibres. Nevertheless, significant opportunities exist in matching the different properties of wood species, or different JW/MW ratios, to specific attributes required during the processing of paper or for certain paper properties. For example, the recommended method for JW refining is to use low intensity refining, which allows separation and development of the strength properties of thin cell wall fibres [57].

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Characteristics of Cells and Properties of Pulps

4.1 Characteristics of Cells and Biometric Relationships

As much as 90–95% of the volume of softwood consists of vertical or longitudinal tracheids; they are elongated, tube-like cells exhibiting a square cross-section with blunt and closed ends. The average length of mature tracheids for the most common pulpwood species is usually 3–4 mm in mature wood (MW). Spruces have longer tracheids than pines. The tangential width of a tracheid is approximately 30–50 µm. The length-to-diameter ratio of softwood tracheids is of the magnitude 100:1. In most hardwood species, fibres occupy 40–75% of the wood volume; the fibres are long with closed ends and very thick walls. There are two kinds of fibres: fibre tracheids and libriform fibres, the former have bordered pits and the latter have simple pits. The fibre length (L) ranges between 0.7-1.2 mm, and the fibre width (W) between 10-30 µm in most hardwoods. The length-to-diameter ratio is approximately 1:50 [1].

The dimensions of tracheids and fibres vary between tree species, from tree to tree and within the same tree [2]. L also varies along the trunk; in general, it increases up from the base to a point where it starts to decrease. To standardise sampling and avoid comparison problems, the sample is taken at a reference height (1.30 m from the ground) called the diameter at breast height.

In the first half of the 20th century the length of the fibres was taken as the most influential feature on the properties of paper. It was found that hardwoods generally produced pulps of lower strength than softwoods, in particular, lower tear strength. The tear strength was especially inferior in the short-fibre pulps with a larger difference in tear rather than tensile strength between hardwoods and softwoods.

Subsequent work showed that all fibre dimensions, or rather the relationships between them, were associated with different kinds of pulp strength. At the beginning of a series of evaluation work concerning the aptitude of tropical timber for pulp and paper, biometric relationships were presented as an issue of special attention as indexes of quality in forest studies. It was stated at that time that the 'flexibility coefficient (F)' (l/W) has a positive influence on the tensile strength, whereas the 'felting power (FI)' (L/W) has a positive influence on the tear strength [3].

It is generally considered that the most favourable paper properties are obtained when the percentage of latewood (LW) fibres and the average value of the wall fraction (WF), are both below 50%. Dense hardwood species that have a fibre WF much above 50% are poorly suited for pulping. The Runkel ratio (R) ($2 \times$ fibre cell wall thickness (w)/lumen diameter) was also introduced at the time as an expression of WF [4]. The R shows good correlation with basic density [5].

Other unusual relationships were eventually used, such as the solids factor (a representation of fibre volume), but did not result in a good single predictor of kraft paper properties in a study performed on Loblolly pine [6].

If operating conditions remain the same, the quality of the produced paper depends on the colour, length, diameter, flexibility, strength and other properties of the fibres used [7].

Several studies have established the effect of wood fibre dimensions and fibre morphology on certain mechanical properties of paper. In summary, the relationships between the basic properties of fibres which have significance on the quality of chemical pulps are:

• FeI, also named 'slenderness ratio', is the relationship between the length and width (both in µm) of the fibre:

$$FeI = (L/W) * 100$$
 (4.1)

• F is the ratio between the diameter of the lumen and the width of the fibre:

$$F = (l/W) * 100 \text{ or } [(W-2 \text{ w})/W)] * 100$$
 (4.2)

• R is the ratio of twice the thickness of the cell wall and the diameter of the lumen:

$$R = 2 w/l$$
 (4.3)

• WF is the ratio between the w and the radius of the fibre:

$$WF = (2 w/w) * 100$$
 (4.4)

• Solids factor is a representation of the volume of the fibre:

Solids factor =
$$(W^2 - l^2) * L$$
 (4.5)

Where:

F: Flexibility coefficient

FeI: Felting index

L: Length

l: Lumen

W: Fibre width

w: Wall thickness

w/w: Weight/weight

The reference values of biometrical ratios are:

- FeI 70-80, satisfactory pulp strengths, especially tear.
- F > 50, good pulp strengths, mainly tensile and burst.
- R < 0.25, excellent fibres for papermaking.

- 0.25 < R < 0.50, very good fibres.
- 0.50 < R < 1.00, good fibres.
- 1.00 < R < 2.00, mediocre fibres.
- R > 2.00, thick walls, the fibre does not collapse and retains its tubular shape leading to poor adhesion.
- WF < 50%, suitable fibres for papermaking.

Biometric parameters and coefficients were widely applied in the study of tropical hardwoods (mostly African species) in the 1950–1970s. Important work investigating the correlation between basic morphological features, such as L and WF, with paper properties has demonstrated that more than 80% of the variations in paper quality could be related to those parameters [8–11]. More recently, they were used to assess the suitability of forested implanted species for paper production as, for example, the studies focusing on *Pinus* [12–15], *Eucalyptus* [16–24], *Populus* [25–27] and *Salix* [28].

Numerous woody species from the five continents have been characterised lately using these parameters, with the aim of establishing their suitability for the manufacture of cellulosic pulp [29–41]. The growing interest to find new raw materials has also pushed the study of numerous nonwood species such as: rice straw [42, 43]; paper mulberry inner bark [44]; giant reed (*Arundo donax*) [45]; *Eulaliopsis binata* [46]; switchgrass (*Panicum virgatum*); *Miscanthus* and others [47]; palm [48]; palma rosa grass [49]; *Hibiscus* [50]; kenaf (*Hibiscus cannabinus*) [51, 52]; *Cannabis* [53]; *Saccharum* [54, 55] and sugarcane bagasse [56, 57]; cotton linters [58] and cotton stalk [59]; bamboo (*Gigantochloa*) [60]; (*Bambusa*) [61, 62]; wheat straw (*Triticum aestivum*) [63] and so on.

Some examples of dimensions of tracheids and fibres of hardwoods, softwoods and nonwood species from subtropical and temperate climates, extracted from the cited references have been analysed. Data are shown in Table 4.1 and the calculated biometrical ratios are presented in Table 4.2.

Table 4.1 Dimensions of tracheids and fibres of selected softwood,					
Selected species	L (mm)	W	w (µm)	1 (μm)	Reference
		(µm)			
Acacia hybrid	1.07	18.8	2.51	13.8	[35]
Acacia mangium	0.98	19.4	2.55	14.3	
Acacia auriculiformis	0.88	16.7	2.81	11.1	
<i>Eucalyptus grandis</i> (4 years-old)	1.06	19.2	3.20	12.8	[24]
<i>Eucalyptus grandis</i> (5 years-old)	0.93	13.0	3.40	6.20	[22]
<i>Eucalyptus grandis</i> (9 years-old)	0.92	16.1	3.50	9.10	
<i>Eucalyptus grandis</i> (18 years-old)	0.98	15.1	3.50	8.10	
Eucalyptus grandis	0.96	13.6	3.20	7.20	[23]
Eucalyptus globulus	0.92	14.6	3.20	8.20	
Eucalyptus viminalis	0.73	12.2	2.60	7.00	
Eucalyptus dunii	0.81	13.8	2.90	8.00	
Eucalyptus cinerea	0.78	11.9	3.00	5.90	
Populus tremuloides	1.05	32.8	3.20	26.4	[25]
Populus tremuloides	0.94	21.0	5.00	11.0	[29]
Populus mexicana	1.45	21.0	4.00	13.0	
Populus simaroa	1.30	29.0	6.00	17.0	
Salix Clone 13-44	0.86	13.6	2.71	8.18	[28]
Salix Clone 250-33	0.95	15.2	3.01	9.18	
Salix Clone 131-27	0.84	15.8	2.88	10.0	
Salix Clone 131-25	0.94	16.2	2.27	11.7]
Salix Clone 26992	0.84	14.5	2.62	9.26]
S. babylonica var. sacramenta	1.07	15.9	3.34	9.22	

Pinus elliotti EW ring 1	2.22	40.6	6.18	28.2	[13]
Pinus elliotti LW ring 1	2.25	38.3	6.56	25.2	
<i>Pinus elliotti</i> EW ring 13	4.05	45.8	7.56	30.7	
<i>Pinus elliotti</i> LW ring 13	3.95	38.2	11.10	16.0	
<i>Pinus elliottii</i> (10 years-old)	3.32	38.8	3.88	31.0	[14]
<i>Pinus taeda</i> (10 years-old)	3.07	36.7	3.86	29.0	
<i>Pinus caribaea</i> (7 years-old)	2.44	54.2	6.62	41.0	[12]
<i>Pinus caribaea</i> (15 years-old)	2.64	59.1	6.47	46.2	
Arundo donax (node)	1.20	16.9	5.30	6.30	[45]
Arundo donax (internode)	1.16	14.6	4.60	5.40	
Gigantochloa scortechinii	1.75	17.2	4.30	8.60	[60]
Hibiscus cannabinus (bast)	2.48	24.2	4.85	14.5	[51]
Hibiscus cannabinus (core)	0.72	31.9	4.08	23.7	
Miscanthus×giganteus	0.97	14.2	4.10	6.00	[47]
Panicum virgatum L.	1.15	13.1	4.60	3.90	
Saccharum Officinerum-Co 89003	1.51	21.4	7.74	5.92	[54]
Sugarcane bagasse	1.59	21.0	5.63	9.74	[56]
Triticum aestivum pbw-343 l	1.18	13.6	3.96	5.68	[63]
Eulaliopsis binata	2.40	10.0	2.11	5.75	[46]
Bambusa tuda	1.89	17.0	6.78	3.44	[61]
EW: Earlywood					

Table 4.2 Biometrical ratios of tracheids and fibres of selected softwood, hardwood and nonwood species from selected references					
Selected species	R	FeI (%)	F (%)	WF (%)	Reference
Acacia hybrid	0.36	57	73	27	[35]
Acacia mangium	0.36	51	74	26	
Acacia auriculiformis	0.51	53	66	34	
<i>Eucalyptus grandis</i> (4 years-old)	0.50	55	67	33	[24]
<i>Eucalyptus grandis</i> (5 years-old)	1.10	72	48	52	[22]
<i>Eucalyptus grandis</i> (9 years-old)	0.77	57	57	43	
<i>Eucalyptus grandis</i> (18 years-old)	0.86	65	54	46	
Eucalyptus grandis	0.89	71	53	47	[23]
Eucalyptus globulus	0.78	63	56	44	
Eucalyptus viminalis	0.74	60	57	43	
Eucalyptus dunii	0.73	59	58	42	
Eucalyptus cinerea	1.02	66	50	50	
Populus tremuloides (1)	0.24	32	80	20	[25]
Populus tremuloides (2)	0.91	45	52	48	[29]
Populus mexicana	0.62	69	62	38	
Populus simaroa	0.71	45	59	41	
Salix Clone 13-44	0.66	63	60	40	[28]
Salix Clone 250-33	0.66	63	60	40	
Salix Clone 131-27	0.57	53	64	36	
Salix Clone 131-25	0.39	58	72	28	
Salix Clone 26992	0.57	58	64	36	
S.babylonica var. sacramenta	0.72	67	58	42	

Pinus elliotti EW ring 1	0.44	55	70	30	[13]
Pinus elliotti LW ring 1	0.52	59	66	34	
<i>Pinus elliotti</i> EW ring 13	0.49	88	67	33	
<i>Pinus elliotti</i> LW ring 13	1.39	103	42	58	
<i>Pinus elliottii</i> (10 years-old)	0.25	86	80	20	[14]
<i>Pinus taeda</i> (10 years-old)	0.27	84	79	21	
<i>Pinus caribaea</i> (7 years-old)	0.32	45	76	24	[12]
<i>Pinus caribaea</i> (15 years-old)	0.28	45	78	22	
Arundo donax (node)	1.68	71	37	63	[45]
Arundo donax (internode)	1.70	79	37	63	
Gigantochloa scortechinii	1.00	102	50	50	[60]
Hibiscus cannabinus (bast)	0.67	102	60	40	[51]
Hibiscus cannabinus (core)	0.34	23	74	26	
Miscanthus × giganteus	1.37	68	42	58	[47]
Panicum virgatum L.	2.36	88	30	70	
Saccharum Officinerum-Co 89003	2.61	71	28	72	[54]
Sugarcane bagasse	1.16	76	46	54	[56]
Triticum aestivum pbw- 343 l	1.39	87	42	58	[63]
Eulaliopsis binata	0.73	241	58	42	[46]
Bambusa tuda	3.94	111	20	80	[61]

Characteristics of Cells and Properties of Pulps

Usual hardwoods in the pulp and paper industry have unimodal distributions of WF, whereas softwood distributions of WF are bimodal. Figure 4.1 shows cross-sections of *Pinus elliottii* wood (Misiones, Argentina), highlighting the cell wall of EW tracheids (a) and of LW tracheids (b). The behaviour of both kinds of cellular elements in a chemical pulp is also shown (c). Whilst the thin walls of EW tracheids cause them to collapse completely, LW tracheids (like hardwood fibres) preserve their pipe-like shape during the refining and sheet-forming processes as a consequence of their thick cell walls, which results in a low interfibre bonding area.





Figure 4.1 Cross-sections of *Pinus elliottii* wood (Misiones, Argentina). a) EW; b) LW; and c) EW and LW fibres in a chemical pulp

The WF of *Pinus* in Table 4.1 varies from about 25% (R: 0.25 and F: 80%) for thin-walled fibres to 35% (R: 0.50 and F: 65%) for the thick-walled LW fibres of the 13^{th} ring. As a consequence, the

percentage of EW and LW in softwoods is very relevant; however, all these pines can be considered suitable for papermaking.

Measurements of the biometric data in EW and LW of different annual rings of 5 trees of 14-year-old *Pinus elliottii* from São Paulo, Brazil, show that there is a variation of L, W and w in the same ring. Differences, which can reach 50%, are clearly more noticeable in the outer rings (which correspond to MW) than in the inner rings (which correspond to JW). The proportions of EW/LW (v/v) tissues were 75/25 in the 1st ring and 43/57 in the 13th ring, so LW becomes relevant on the periphery of the stem [13].

Fibrous and biometric global data, at breast height, of 10 year-old *Pinus* from Misiones, Argentina [14], show longer and more flexible tracheids than those of the São Paulo plantation [13]. The reason could be the subtropical climate in the Misiones region, whereas the São Paulo area has a temperate climate due to its altitude, but it could also be a result of genetic manipulation of the species, since there are about 20 years between the two studies. Unfortunately, there was no recent work found on these species in this region.

Almost all hardwood fibres in **Table 4.1** have a WF between approximately 20-50% of the total fibre cross-section, a F between 50-80%, and a R between 0.25-1.00; however, only the *Acacias*, the 4-year-old eucalyptus and one of the poplars are below 0.5. Only *E. cinerea* would exhibit a poor papermaking performance.

The dimensions of *Eucalyptus grandis* show differences of about 30% between trees of ages 5, 9 and 18 years [22].

Comparative data of *Salix clones*, including *S. babylonica* var. *sacramenta*, named 'American willow', and 5 hybrids implanted in 2 culture sites in Argentina [28] evidence that the variability in biometric characteristics between *Salix clones* is similar or more than those between *Eucalyptus* species [23].

Of all nonwoods included in Tables 4.1 and 4.2, only *Hibiscus* cannabinus (bast and core fibres), *Gigantochloa scortechinii* and

Eulaliopsis binata, have a WF and R below 50% and l respectively, and a F above 50%.

The initially adopted premise was that average L below the critical value of 700–900 µm, invalidated the relationships between the biometric parameters and pulp properties [3]. According to the data from Table 4.1, the species *Hibiscus cannabinus* (core), *Eucalyptus viminalis, Eucalyptus cinerea, Eucalyptus dunii, Salix Clone 131-27, Salix Clone 26992, Salix Clone 13-44* and *Acacia auriculiformis* have L in that range. Nevertheless, these species, mostly *Eucalyptus viminalis, E. dunii* and *Salix clones* are extensively used in the pulp and paper industry.

When analysing all the data from Table 4.1 (Figure 4.2), it is found that the R correlates inversely with the F (correlation coefficient r = -0.92) and directly with the WF (r = 0.92), indicating that all three ratios represent the same property. Considering R values between 0.2 and 1.00, the relationship is linear (r = 0.98 for the WF and r = -0.98 for the F). These values are consistent with the recommended values of a R (<1), WF (<50%) and F (>50%).



Figure 4.2 Relationship between the R, F and WF from Table 4.2

The relationship between the R and FeI for hardwoods, softwoods and nonwoods from Table 4.2 are shown in Figures 4.3 and 4.4.



Figure 4.3 R *versus* FeI of hardwood species from Table 4.2



Figure 4.4 R *versus* FeI of different commercial pines and nonwood species from Table 4.2

Most hardwoods are found in the quadrant circumscribed by FeI values of 50–70%, and only *Eucalyptus grandis* [23], *Populus mexicana* and *Salix clones 13-44* exhibit FeI values of over 70%. *Populus mexicana* and *Salix babylonica* var. *sacramenta* have the best combination of R and FeI (Figure 4.3). Mexican poplars, *Populus simaroa* and *Populus tremuloides* [29], show the highest fibre rigidity

and lowest FeI. *Acacia* and *Salix clones* present a lower R and FeI than the evaluated *Eucalyptus*.

Unlike hardwoods, most nonwood species (except the core fibres of *Hibiscus cannabinus*) possess FeI higher than 70%, whereas their R are less acceptable (Figure 4.4). *Eulaliopsis binata* and *Bambusa tuda* were not included in the figure because their biometric parameters are too far from the standards.

Among the pines, tracheids of *Pinus caribaea* aged 7 and 15 years, as well as tracheids from EW and LW from the ring l of *Pinus elliottii*, show FeI values below 70%.

4.2 Relationship between Fibre Characteristics and Pulp Properties

In fact, fibre properties in pulps are not directly related to fibre dimensions in the tree. There are many factors which affect the weight and dimensions of fibres, such as the pulping process (mechanical, chemimechanical, semichemical and chemical pulping), the pulping yield, the consistency and intensity of refining and so on.

Intrinsic fibre strength must be taken into account together with fibre characteristics that influence conformability and fibre-to-fibre bonding. A pioneering work has demonstrated that the zero span tensile test, when properly employed, provides a valid measure of the intrinsic strength of the fibres [64]. 'Fibre quality' has been defined as the ratio between the zero span tensile strength and the basic weight, and this index has been used as a measure of fibre strength [65]. As early as 1965, an extensive review of the works published over the previous 30 years defined that the three principal factors controlling paper strength were fibre density (w or percentage of summerwood), L and fibre strength [66].

The tensile strength of the paper is controlled by the tensile strength of fibres and by the strength of interfibre bonding. In addition, the

strength of interfibre bonding depends on the relative bonded area (RBA), i.e., the area of contact between two fibres and the bond strength per unit of bonded area (b). Any increase in fibre strength, RBA or b. will increase the tensile strength of the sheet. These concepts were concentrated in a formula which uses the zero span tensile strength, as a measure of the intrinsic fibre strength in the sheet, and fibre dimensions (mean values of cross-section, perimeter and length) [67].

Coarseness is defined as the weight per unit length of fibre expressed as milligrams per 100 m [68]. For example, the coarseness range of eucalyptus pulp fibres is 4.5–11 mg/100 metres. The value depends on the species, tree age, the improved clone and so on, because it is a function of the individual length of each fibre, the w, the fibre diameter and therefore is connected with the average weight of each fibre [69]. Reference values for eucalyptus bleached kraft pulps are 9–11 mg/100 m (high coarseness for use in tissue and filter papers) and 4.5–6 mg/100 m (low coarseness, for other uses) [70].

It has been demonstrated that the coarseness depends greatly on the pulping process, the degree of delignification and the refining, since as these properties increase, the weight of the fibre decreases, reducing the coarseness. Therefore, the coarseness serves as a predictor of mechanical properties only when considering the same kind of pulp or pulps from similar species, otherwise, the results of the relationship of coarseness properties may be different from the assumed ones [71]. Actually, the coarseness is ideal for monitoring or controlling a process in the factory, since any significant alteration in the process could alter the weight of the fibre. However, it cannot be used to compare raw materials or to establish the suitability of a species for papermaking.

A representation of idealised fibres with different coarseness is shown in Figure 4.5, demonstrating that fibres with very different characteristics may have similar coarseness [72–74].

Table 4.3 Biometrical characteristics and coarseness of tracheids and fibres of selected softwood and hardwood species from selected references						
Species	L (mm)	W (µm)	w (µm)	1 (µm)	Coarseness (mg/100 m)	Reference
Pinus radiata LW	3.00	31	7.0	17.0	26	[72]
<i>Pinus</i> <i>sylvestris</i> kraft pulps (sawmill chips)	2.42	35	6.1	22.8	25	[73]
<i>Pinus</i> <i>sylvestris</i> kraft pulps (thinnings)	1.87	35	5.1	24.8	18	
Pinus radiata EW	3.10	31	4.0	23.0	16	[72]
Fagus sp.	1.30	15	5.0	5.0	12	
Populus sp.	0.90	16	2.6	10.8	7.0	
Eucalyptus sp.	0.74	15	2.7	9.5	6.0	[74]
Acacia mangium	0.65	14	2.0	10.1	4.6	



Figure 4.5 Examples of different idealised fibre coarseness [72-74]

Accurate determination of fibre coarseness was difficult prior to the appearance of automatic measuring equipment, such as the Kajaani or the Fibre Quality Analyser [75–80]; these technologies facilitate

the complete characterisation of the biometric parameters of fibrous raw material and pulps, and are increasingly used [15, 20, 27, 61, 63, 81–92].

Biometric parameters can define the possibility of using certain species in chemical pulping, but their usefulness is relative in mechanical pulping because fibrous features, basically those related to fibre stiffness, have different consequences.

Mechanical pulps from spruce have excellent properties due to their fibre characteristics, whereas hemlock and balsam fir respond well to refining but result in lower tear strength and somewhat higher bulk characteristics than spruce. USA southern pines proved to be more difficult to pulp than spruce when using mechanical processes, as they have a higher proportion of summerwood fibres, with thick cell walls, resulting in high reject rates during screening and therefore require higher energy to refine the rejects. Hardwoods were initially considered unsatisfactory for mechanical pulping, but this has been solved using chemical pretreatments [93].

The mechanical behaviour and optical properties of mechanical pulps can be explained by the delicate equilibrium of the three fractions that form these pulps (fibres, fines and fibre bundles), and also by their function in pulp web formation. Strength values are related to the ratio of whole fibres, fibrillation of their walls, average L and shape of the particles, and by the characteristics and proportion of fines [94–98]. Beyond the cut and fibrillation, refining produces alterations in fibre, such as curls and kinks, related to the bending of the fibre, but these topics exceed the scope of this chapter.

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5 Final Remarks

The proliferation of plantations has marked the last decade, and it is expected to be continued and intensified in the near future.

The remarkable proliferation of forest genetic breeding assays, generally aimed at increasing productivity and shortening the age of rotation, results in trees entering the mill which contain an increasingly important proportion of juvenile wood (JW).

With some exceptions, especially in very aged trees (with strong duraminisation), the characteristics of JW are lower density, shorter and wider fibres, and a higher microfibril angle than mature wood. This produces alterations to yield, intrinsic strength, elongation, and results in the shrinkage of fibres. JW fibres have characteristics that reduce the quality of chemical pulps, whereas they may be suitable for mechanical pulps. The use of JW is also recommended in cases where raw materials have thick fibres.

In this survey of more than a decade (13 years), it has been found that the suitability of new species for papermaking is continuously tested, particularly in the Middle East and Asia.

The Runkel ratio combined with the felting index (FeI) remains the most relevant and used fibrous relationship in the assessment of fibres; most other relationships are redundant.

Gathered data show that pines have a Runkel ratio (R) of between 0.2-0.5 and a very variable FeI: lower than 50 for Caribbean pine, 50–60 for Brazilian pine (São Paulo) and 80–100 for Argentina (North-east) pines, but these data are isolated, and cannot be taken as a reference.

Most hardwood species currently in commercial use (eucalyptus, and hybrids of willows and poplars) have a R between 0.5-1, and FeI between 50-70%, which deviates from the premises established in the 1960s.

Most nonwoods recently tested possessed a R between 1-2, and FeI between 60-100%. Fibres from sugar cane bagasse are widely used for paper production in Central and South America. Although there are several varieties of sugar cane, the ones reported in this work fall within these ranges.

Fibrous parameter determination has undergone a revival due to the appearance of automatic equipment. These devices have certainly facilitated the determination of the monitoring fibre characteristics from pulping and refining processes, such as coarseness, curl, kink and so on.

Abbreviations

b	Bond strength per unit of bonded area
BHKP	Bleached hardwood kraft market pulp
DP	Degree of polymerisation
EW	Earlywood
F	Flexibility coefficient
FAO	Food and Agriculture Organization of the United Nations
FeI	Felting index
G-layer	Gelatinous layer
JW	Juvenile wood
L	Fibre length
1	Fibre lumen
LW	Latewood
MFA	Microfibril angle
ML	Middle lamella
MW	Mature wood
Р	Primary wall
R	Runkel ratio
RBA	Relative bonded area
S1	Secondary wall outermost layer
S2	Secondary wall middle layer
S 3	Secondary wall innermost layer
TMP	Thermomechanical pulping

v/v	Volume/volume
w	Cell wall thickness
W	Fibre width
WF	Wall fraction

ndex

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Firs and pines dominated the global picture of raw materials used by the paper industry until the 1950s. At that time, the interest in introducing new species, mostly hardwoods, led researchers to intensify efforts investigating the fibrous characteristics, and their combinations, which could represent the relationship between fibres, pulp and paper.

The pulp and paper industry has shown, mainly in the last two decades, a strong North-South displacement. This is to a large extent due to the favourable climate, which promotes tree development. Similarly, the paper fibres have gone from being almost exclusively softwoods from natural forests of the cold regions of the northern hemisphere, such as spruce and birch, to fast-growing species of short fibres, such as eucalyptus, and willow and poplar hybrids from plantations.

These new species, which are beginning to dominate the paper panorama, not only differ from classic species in fibre length, but they present particular characteristics, such as large amounts of juvenile wood, different fibrillar angle and so on, because trees are increasingly used at a younger age.

This leads us to question whether the old paradigms concerning the relationships between fibre characteristics and pulp properties are still valid or should be reviewed and updated, in which case, the basic fibre parameters, their influence in pulping and refining, and their impact on paper quality should be redefined.

The purpose of this book is to survey publications of the last decade, to verify which morphological characteristics of the fibres authors currently consider relevant, in order to establish the state of the art for this topic. Relatively recent data were surveyed because of the continuous changes that occur in the species due to genetic improvement.



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