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Cellulose microfibril angles in a spruce branch and mechanical implications

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The nanostructure of the wood cell wall and, in particular the tilt angle of the cellulose fibrils versus the longitudinal cell axis (microfibril angle, MFA), are known to play a key role in determining the mechanical properties of wood. A variation of microfibril angles during growth may therefore be regarded as a means to adapt to different loading situations. In the present study, a branch of Norway spruce (*Picea abies*) was used as a model system. The change of microfibril angles with increasing age and size of the branch and therefore increasing gravitational load was systematically investigated. Small angle X-ray scattering (SAXS) was applied to obtain a map of MFA all over the branch as a function of the distance from the trunk within each annual ring. It was found that in compression wood the MFA decreased continuously from the trunk towards the tip in all annual rings. In opposite wood, however, the course of microfibril angles was found to change considerably with the age of the branch: in the outer annual rings, very small microfibril angles occurred in the middle part of the branch. The results are discussed in view of the mechanical implications of different microfibril angles.

1. Introduction

The remarkable mechanical performance of plant bodies and their ability to cope with varying external stresses is a field of growing interest [1]. Trees, for example, are able to reach a considerable size and stability with a minimum of material and adapt to changing loading conditions. Since wood is a hierarchically structured material, the adaptation occurs on various length scales. On the macroscopic level the shape of stem and branches is optimized to avoid local stress concentrations [2]. On the micrometer level, wood is a cellular tissue [3], and on the nanometer level, the complex cell wall architecture influences the material properties of wood [4, 5].

One of the strategies to cope with particularly high local stresses is the formation of reaction wood. In conifers, the term reaction wood is used for tissue under predominantly compressive load as it is found on the lower side of branches and leaning stems [6–8]. It is also termed compression wood, whereas the tissue on the tension side is referred to as opposite wood. Due to a larger growth ring width in compression wood, the cross sections of leaning trunks and branches show a pronounced radial eccentricity (Fig. 1). Compression wood is known to differ from normal wood in composition and cell anatomy [6, 9–11]. Moreover, the tilt angle of the cellulose fibrils with respect to the longitudinal cell axis (microfibril angle, MFA) was found to be larger in compression wood than in normal wood [7, 12, 13].

It is most notable that a change in microfibril angle can result in a change of mechanical properties, such as stiffness and extensibility, of more than one decade [4, 5, 14, 15]. In a recent study the course of the MFA in stems of different species has been interpreted in terms of mechanical optimization [16]. It was suggested, that different microfibril angles may be a means of optimizing the material properties of wood. However, the correlation between mechanical stimuli (stresses and strains) and the tree's reaction through a particular microfibril angle is not clearly established. In order to investigate the relationship between a mechanical trigger

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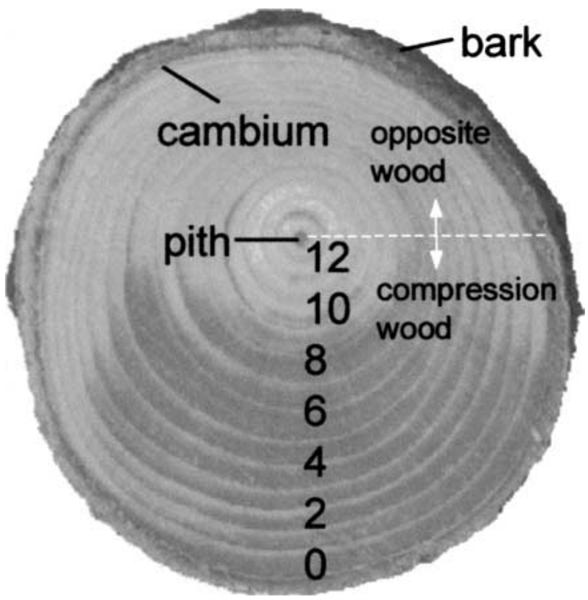

Figure 1 Cross-section of the branch at a position close to the trunk. Note the pronounced eccentricity of the cross section with compression wood on the lower side. The following notation was chosen to designate the annual rings: the outermost and thereby latest formed ring is called 0, followed by 1, 2 and so on.

and structural changes one has to find a system as simple as possible. In the present paper, we chose a conifer branch, grown in a calm environment and, in first approximation, subjected to gravitational forces only. This gives a precisely defined loading pattern with tensile stresses on the upper side and compressive forces of the lower side of the branch.

Since wood grows by adding annual ring by annual ring, it is possible to reconstruct the size, weight and shape of the branch at every time during its life span. Assuming only gravitational forces, this allows one to reconstruct the entire loading history of the branch. Given this information, one should be able to correlate typical stresses occurring in the cambium (the thin layer between wood tissue and bark where the new wood cells are formed) with the microfibril angle of the newly built wood cells.

In the present paper, the microfibril angles in a Norway spruce branch were investigated systematically across the annual rings at several distances from the stem, using small-angle X-ray scattering (SAXS).

2. Materials and methods

2.1. Sample preparation

Samples were taken from a branch of Norway spruce (*Picea abies*), grown in calm environment in the Wienerwald, near Vienna, Austria. The branch measured 5 m in length and its diameter at the trunk was 76 mm. It was cut into five discs in one meter distance from each other. The largest disc contained 20 annual rings, 18 rings were found in the second one, 13 in the third one, 9 in the fourth and 4 in the fifth and smallest disc. The following notation was chosen to designate the annual rings: the outermost and thereby latest formed ring is called 0, followed by 1, 2 and so on, see Fig. 1. This implies that annual ring 0 (the latest built) is shared by all discs, whereas this is not the case for the inner annual rings.

200 μm thick slices were cut from the discs in radial direction using a microtome. The slices in were stored wet condition (above the fiber saturation point) and encapsulated in plastic foil to keep the wood from drying in the vacuum chamber of the X-ray device.

2.2. Measurement

The microfibril angle was determined with position resolved small-angle X-ray scattering (scanning SAXS [17]). An X-ray generator with a rotating copper anode served as radiation source (wavelength $\lambda = 0,154$ nm). The device was equipped with crossed Göbel mirrors as monochromator. The beam size at the sample position was 200 μm. The scattering patterns were recorded by a multiwire proportional detector with an imaging area of 11.5 cm in diameter and a resolution of 200 μm (Nanostar, Bruker AXS). The sample was mounted on a computer controlled movable stage that could be scanned across the beam in two orthogonal directions. The samples were aligned such that the longitudinal cell axis was oriented horizontally and at right angles to the incident beam. Scattering patterns were recorded at selected positions on the sample. In order to obtain reasonable statistics each scattering image was accumulated during one hour.

2.3. Data evaluation

The scattering images contain information about thickness and arrangement of the cellulose fibrils [18–20]. Fig. 2a shows an example of a typical scattering pattern resulting from rectangular cells. The scattered intensity is displayed in a pseudo-gray scale: bright colors indicate high intensity and dark colors indicate low intensity. One can see three streaks of high intensity, the black area in the center is the shadow of the beam stop keeping the very intense direct beam from hitting the detector. In order to determine the MFA quantitatively, the scattered intensity was integrated over the scattering vector q and plotted versus the azimuth angle χ. The resulting curve showed three peaks that were fitted with three Gaussians of equal width, see Fig. 2b. In the case of rectangular cells, the microfibril angle is directly given by the distance of each of the outer peaks to the central peak [13, 21]. In case of round wood cells, however, the scattering pattern consists of two streaks only and the evaluation procedure is slightly more complex. For details refer to [16] or [21].

3. Results

The MFA obtained from the SAXS-measurement were correlated with the position of measurement, yielding a map of microfibril angles all over the branch. In Fig. 3, the microfibril angles are plotted versus the length of the branch for the annual rings 0 to 8. Fig. 3a shows the curves for opposite wood and Fig. 3c for compression wood. The schematic longitudinal section in Fig. 3b illustrates the positions in the branch where the measurements were performed.

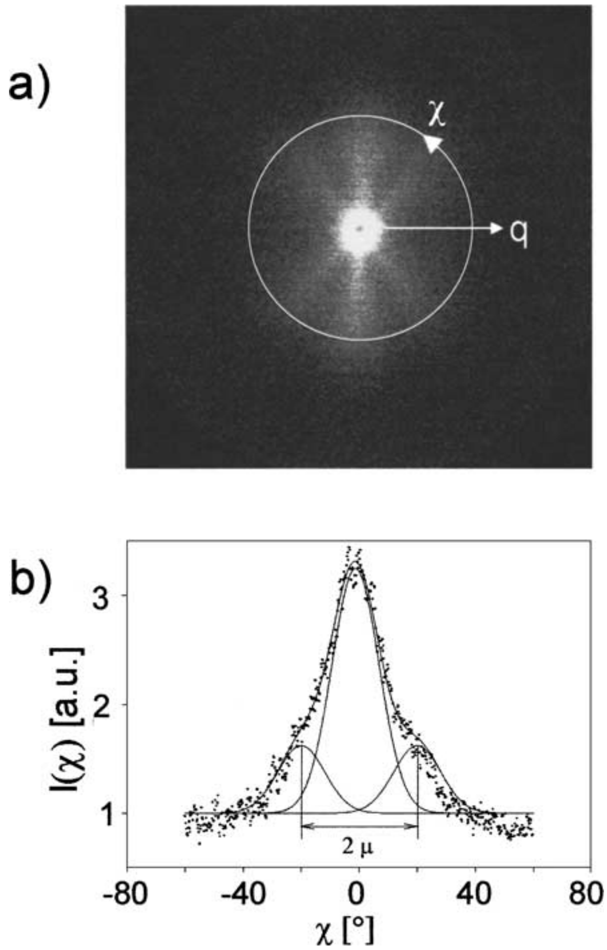

Figure 2 (a) shows a typical scattering pattern obtained from spruce compression wood. The longitudinal cell axis was oriented horizontally. (b) The scattered intensity was integrated over the scattering vector q and plotted versus the azimuth angle χ. The curve was fitted with three Gaussians of equal width. In case of rectangular cells, the microfibril angle μ is given by the distance of the outer peaks.

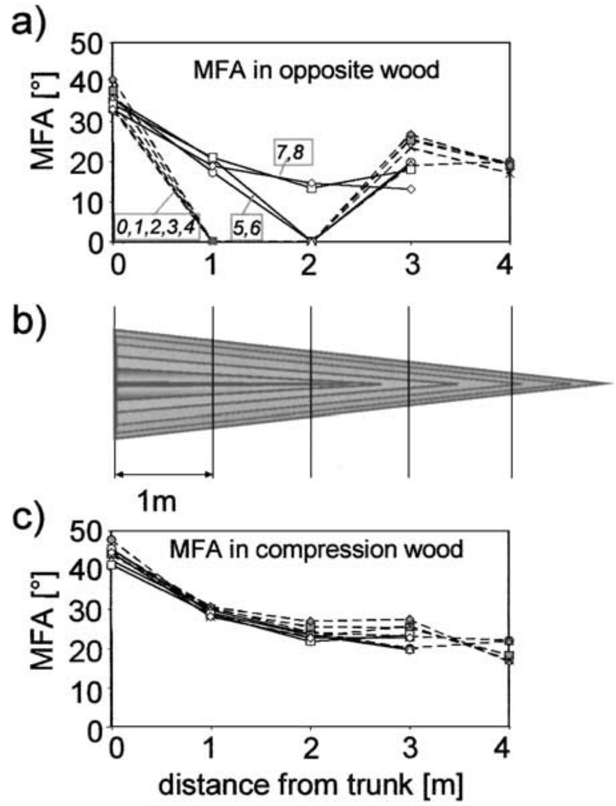

Figure 3 The MFA of each annual ring was plotted versus the length of the branch. (a) As a consistent trend in all annual rings the MFA in compression wood was found to decrease continuously from about 45° near the trunk towards 20° near the tip of the branch. (b) Due to the additive growth of trees, the branch consists of a set of cones as shown schematically. This implies that the cross-sections contain different numbers of annual rings, at 4 m distance from the stem the disc contains only annual rings 0–4. (c) On the tension side, a great variation of the course of microfibril angle with the age of the branch was observed: in the first two annual rings of the young branch (rings 7 and 8, near the pith) a behaviour similar to compression wood was found: the microfibril angle decreased continuously from the trunk towards the tip, only the MFA-values very generally lower. In annual rings 5 and 6 the MFA dropped to very small values around zero at 2 m distance from the trunk. In annual rings 0, 1, 2, 3 and 4, very small MFA were found at even two positions: 2 and 3 m from the stem.

In compression wood (lower side of the branch) the MFA was found to decrease continuously from about 45° near the trunk to about 20° at the tip of the branch in all annual rings. Quite in contrast, on the opposite side, the course of the MFA with position changed with the age of the branch: in the innermost two annual rings (ring no. 7 and 8, corresponding to the first annual rings of the young shoot, see Fig. 3b) the decrease of microfibril angles was observed to be continuous, as found in compression wood. Only the values of MFA were generally lower than in compression wood: they decreased from about 35° near the trunk to 17° at the tip. In annual rings no. 5 and 6, the course of microfibril angles changed and very small values of MFA occurred at 2 m distance from the trunk. In the outermost annual rings (i.e. that were built last) 0, 1, 2, 3 and 4, the MFA dropped to almost zero between one and two meters from the trunk. The results are summarized in Table I.

4. Mechanical considerations

Several conclusions can be drawn from the distribution of MFA in the branch (Fig. 3). First, rather large MFA's are found at all ages of the branch in the region close to the stem where the largest bending moments occur. This clearly indicates that stiffness cannot be the primary optimization principle since high stiffness is achieved with low MFA's [5]. On the other hand, MFA's close to zero are gradually appearing with age on the upper side of the branch, at some distance from the stem, which certainly leads to some stiffening.

4.1. Stiffness or flexibility?

From the function of the branch both flexibility and stiffness are required:

(I) the branch should not bend excessively under its own weight to keep the needles exposed to sunlight.

(II) the branch should be flexible enough to allow bending until heavy loads (e.g. snow) will slide off.

In order to estimate the consequence of these two conflicting requirements, one may use a very crude model for the branch, describing it as a cylinder with constant diameter D and length L and with constant mechanical

TABLE I Cellulose microfibril angles (in degrees) within the last 9 annual rings. The five discs (1–5) were taken at distances 0, 1, 2, 3 and 4 meters from the stem

		Disc 1 MFA	Disc 2 MFA	Disc 3 MFA	Disc 4 MFA	Disc 5 MFA
Opposite wood	Ring 0	33.05	0.00	0.00	19.10	20.25
	Ring 1	33.61	0.00	0.00	25.70	19.57
	Ring 2	37.90	0.00	0.00	25.45	19.00
	Ring 3	40.78	0.00	0.00	26.90	19.00
	Ring 4	35.00	0.00	0.00	23.52	
	Ring 5	36.00	17.40	0.00	19.79	
	Ring 6	36.00	20.71	0.00	19.38	
	Ring 7	34.54	21.05	13.47	18.00	
	Ring 8	33.30	18.94	14.71	13.11	
Pith						
Compression wood	Ring 8	44.50	28.20	22.68	21.50	
	Ring 7	41.32	28.82	21.84	23.40	
	Ring 6	42.55	29.87	23.58	19.83	
	Ring 5	45.42	29.20	23.48	19.67	
	Ring 4	45.44	28.16	22.91	25.80	
	Ring 3	43.18	30.68	26.89	27.41	16.50
	Ring 2	44.18	30.09	25.50	25.23	18.15
	Ring 1	45.00	31.35	24.11	23.00	22.00
	Ring 0	47.56	28.48	24.25	20.00	22.56

properties (implying also a constant MFA) over the whole branch. Under these conditions, in the region close to the stem where the largest bending moments occur, the bending moment due to gravity alone can be written as

$$M = \frac{1}{2}L^2\frac{\pi}{4}D^2\rho g \qquad (1)$$

where ρ is the mass density and g the gravitation constant. Condition (I) implies that the radius of curvature of the branch should always stay larger than some value R_0, which means that $E_L I/M > R_0$, where E_L is the Young's modulus parallel to the wood cell axis and $I = D^4\pi/64$ the moment of inertia [22].

A lower limit for the aspect ratio D/L results from this condition:

$$(D/L)^2 > 8\rho g R_0/E_L \qquad (2)$$

condition (II) has already been shown previously [16] to lead to an upper limit for the aspect ratio

$$D/L < 2\,\varepsilon_0/\lambda_0 \qquad (3)$$

where ε_0 is the fracture strain of the material and λ_0 a constant depending only on θ_0, the critical angle where heavy loads, such as snow, are sliding off.

$$\lambda_0 = (\cos\theta_0)^{1/2}\int_{\theta_0}^{\pi/2}(\cos\theta_0 - \cos\theta)^{-1/2}d\theta$$

The main reason for this upper limit is that the bending of the branch is limited by the fracture strain [16]. The inequations (2) and (3) can only be satisfied simultaneously if

$$\frac{1}{2}{\varepsilon_0}^2 E_L/\rho > g\, R_0\, {\lambda_0}^2 \qquad (4)$$

All material parameters are on the left side, while R_0 and λ_0 define the functional requirements for the branch, according to the conditions I and II. First, a low mass density ρ is advantageous. In fact, due to its cellular structure, wood has a very low density. For a honeycomb-like structure, such as wood, the elastic modulus (parallel to the direction of the cells) is known to be proportional to ρ [23]. Calling ρ_0 the mass density and E_L^* the Young's modulus of the cell wall material, then $E_L = E_L^*\rho/\rho_0$ and Equation (4) can be rewritten

$$W_0(\mu) = \frac{1}{2}E_L^*{\varepsilon_0}^2 > \rho_0 g R_0 {\lambda_0}^2. \qquad (5)$$

$W_0(\mu)$ can be understood as a measure for the specific fracture energy of the cell wall material. It is, in fact, the only parameter appearing in the Equation (5) which depends on the microfibril angle, μ.

Both, E_0 and ε_0 have been measured as a function of μ in previous work using tensile testing [5]. Taking this data, $W_0(\mu)$ can be determined and it turns out that W_0 first increases with μ and then has a broad maximum in the range $\mu = 30$–40° [16, 24]. Therefore, it is not surprising that the MFA is close to this value at all ages in the region close to the junction into the stem where the bending moments are a maximum.

4.2. Compression wood

The model discussed above is, of course, much too simplistic to describe the real situation of a branch. First, the branch is not cylindrically shaped but rather conical, the MFA does not have the same value everywhere (as shown in Fig. 3) and, above all, the loading patterns are very different on the upper and the lower side of the branch. This is important since a honeycomb-like structure is known to behave very differently under tensile and under compressive load along the cell [23]. Due to buckling of the cell tubes, yield strength under compression is usually much smaller than under tension.

This effect is also observed for wood. Hence, the optimization of MFA will generally be different for wood loaded under tension or under compression.

On the lower side of the branch, the microfibril angles near the trunk were found to be larger than those in opposite wood, up to 50°. A larger microfibril angle, corresponding to a slower helix of cellulose fibrils, is considered as beneficial regarding stability of the wood cells upon longitudinal compression [25]. Local failure of the matrix allows the compression of the helix to a certain extent without damaging the cell completely. In contrast, small microfibril angles (vertical cellulose fibrils) provide higher cell wall stiffness, but once the compressive force exceeds a certain limit, the cellulose fibrils will kink and the cell will undergo catastrophic failure. Indeed, in compression wood generally particular large microfibril angles are found [7, 9, 12, 26]. In the present study the microfibril angles were found to decrease from the trunk towards the tip. This may be due to the fact that also the bending moment and the compressive stresses caused by the branch's own weight decrease from the trunk to the tip.

5. Discussion

The systematic investigation of microfibril angles in a spruce branch showed that the MFA was not constant across the branch but changed considerably with position. Near the trunk, microfibril angles of about 45° were found, which is in good accordance with the literature [9, 12, 26]. The microfibril angle is known to be closely correlated with the mechanical properties of wood. Tensile tests had shown that small microfibril angles result in a high stiffness and strength, whereas large microfibril angles correlate with a low stiffness, but a high extensibility and toughness [12]. Therefore, the high microfibril angles adjacent to the trunk in compression wood as well in opposite wood (Figs 3a and c) indicate material with a relatively low stiffness and strength, but a higher flexibility and toughness. The gradual decrease of microfibril angles over the length of the branch in compression wood corresponds to a slight stiffening of the material from the trunk towards the tip. In opposite wood, a similar trend was observed in the first two annual rings. In the following two annual rings, the sudden occurrence of microfibril angles below 5° at 2 m from the trunk results in a considerably increased local stiffness at the upper side of the branch. Small microfibril angles in the last five annual rings at even two positions indicate an increased stiffening of the upper side of the branch with increasing age.

This finding is surprising for two reasons: First, the great variation of microfibril angles suggests a structural adaptation to mechanical stresses. However, in conifers, the reaction to large bending forces has up to now been mostly associated with the formation of cells with different anatomy (round cells, thicker cell walls) and different composition (higher lignin content) on the compression side [3, 8, 10]. The present study shows that in contrast to the adaptation on the microscopic level, the structural adaptation on the nanometer level is not restricted to compression wood. Quite on the contrary, it seems to be even more pronounced in opposite wood.

Secondly, the above calculations for wood under bending stress suggest that larger angles on the tension side may be beneficial in order to allow the branch to bend and shed external loads. With increasing age of the branch, however, very stiff material is deposited on the tension side.

An increasing size of the branch also makes the compressive as well as tensile stresses and strains on the surface of the branch increase. Since it is well known that wood is twice as strong in tension than in compression, bending will lead to failure on the compression side rather than on the tension side. One may speculate that above a certain size of the branch, the danger of compressive failure due to the branch's own weight may become critical and further bending is therefore counteracted by stiffening of the tension side. This may, in part, explain the peculiar course of microfibril angles in opposite wood. It is, however, not clear, why such a stiffening should occur in the middle part of the branch only and not near the trunk. Quantitative models of the real load pattern of the branch should help clarify these questions. Moreover, the influence of other parameters, like the increased lignin content of compression wood compared with normal and opposite wood [27], or a possible influence of different cell anatomy in compression and opposite wood is still unclear.

6. Conclusions

The systematic investigation of the nanostructure of a branch of *Picea abies* showed that on the tension as well as the compression side, the MFA is generally larger than in the stem. Moreover, in both, opposite and compression wood, the MFA changed as a function of the position. Whereas in compression wood, we observed a continuous decrease of microfibril angles from the trunk to the tip of the branch in all annual rings, in opposite wood the course of microfibril angles changed considerably with the age of the branch. In the first annual rings, large microfibril angles were found providing high flexibility to the young branch. In the subsequent annual rings a gradual stiffening of opposite wood through very small microfibril angles in the middle part of the branch was observed. The results indicate that it is not only the so called reaction wood on the compression side that responds to different load patterns but also the "normal" wood on the opposite side. On the nanometer level, opposite wood even shows a more obvious response to changing mechanical requirements with increasing age and size of the branch. It is suggested, that the stiff regions in opposite wood with very small MFA may act as an effective reinforcement, keeping the branch from further bending under its own weight and, thus, protecting the branch from the more critical danger of compressive failure.

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