Determination of Microfibril Angle in Compression Wood By Means of X-Ray Diffraction Technique

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Abstract

The anomalous physical and mechanical behaviour of compression wood (CW) has to be mainly ascribed to the microfibril angle (MFA) arrangement within the cell wall. Studies have also highlighted that at least two different severity classes can be established according to the intensity (darkness) of the CW colour. The aim of this work has been that of establishing possible relationships between CW severity classes and MFA. Measurements were performed on samples of both compression and normal wood using the X-ray diffraction technique. Results showed different types of intensity diagrams between normal and compression wood, which were characterised by two or three well-separated peaks.

Introduction

Compression wood represents the physiological answer of the tree to the action of external compression stresses, and it is also considered one of the most important defects for softwood species, as it affects both physical behaviour and ability to process sawn timber. It has been the subject of many studies covering its anatomical and ultra-structural properties.

Compression wood can occur in different grades, ranging from mild to severe, on the basis of its anatomy;

The first class (*severe compression wood*) is characterised by very thick cell walls, higher lignin content in the S_1 and the S_2 layers (S_{2L}) (particularly in the outermost region of the S_2 layer around the cell perimeter), rounded cell outline with intercellular spaces at the corners, distinct helical cavities, distorted bordered pit apertures, and the absence of the S_3 layer.

The second class has poorly developed helical cavities, intercellular spaces could be present or not, the lignin content is high and the cell walls are still thick.

In the third class (*mild compression wood*) the wall is still thicker than in tracheids of normal wood as well as the lignin concentration in S_{2L}, the cell outline is round; the S_3 is normally missed out. generally there are no more intercellular spaces and tracheids lack their helical cavities.

Donaldson et al. [1] also noticed that an increase in MFA was associated with severity of CW, except within the first few rings from the pith, where the presence of juvenile wood already determine a larger MFA. It is therefore of some technological importance to develop methods that are as simple and rapid as possible for the determination of MFA. Over the years some direct methods have been developed like, for example, polarized light microscopy, fluorescent microscopy, drying checks and iodine staining, ultrasonic checks, angle of slit pits, etc. All of these methods are laborious and very time consuming as the extreme variability of the material demands that a large number of fibre elements has to be measured in order to give meaningful average values.

X-ray diffraction (XRD) technique, on the other hand, can provide a mean diffraction pattern of several hundred elements in a single exposure, at little cost in terms of preparation and observation time. But this method requires the use of complex X-ray apparatus not generally available and a large amount of electronic computer and operator time in the analysis of the diffraction patterns [2].

In this work[1] wood of European larch (*Larix decidua* Mill.) was studied by means of XRD technique. The aims were to analyse (002) diffraction patterns and intensity profiles given by compression wood samples, to compare them with those obtained from normal wood, and to determine the average MFAs of the individual specimens.

Material and methods

For MFA determination the samples were taken from different European larch stems, sampled in two stands located on Alp Mountains in Italy.
Each specimen was obtained from a single annual ring, suspiciously containing compression wood, separated into earlywood (ew) and latewood (lw) and sampled from juvenile and mature wood.
As the average ring with of the disk was quite large, the dimensions of the samples were 0.5 mm x 0.5 mm (radial and tangential directions in the wood) x 10 mm (longitudinal direction) each containing only earlywood or latewood.

The XRD set up was used in perpendicular transmission mode. A diffractometer for single crystal at three circles with a copper rotating anode generator was used. The radiation source was a CuKα (λ = 1.542 A) with a 0.5 mm aperture incident beam.
The wood sample was mounted on the goniometer (motorized three-axis positioning head) with a sample holder that held it perpendicular to the incident X-ray beam, which passed through the radial (or tangential) face. The time of exposure was 180-240 seconds for each sample. Data was then collected by a SIEMENS X-ray scattering system, and the diffraction patterns were recorded by means of a CCD area detector and showed on the computer screen. The pattern consists of a series of arcs that are spaced apart by a number of well-defined concentric circles. The radii (2θ) of the concentric circles are indications of the spacing of the crystalline planes within the cellulose crystalline fibrils [i.e. 2θ = 22.6° for (002) planes and 2θ = 34.5° for (040)] (Fig.1). The strongest reflection of (002) planes was used for MFA analysis[2]. The intensity of the (002) crystallographic plane arcs was recorded and integrated by using GADDS software. The resulting curve is the intensity profile displayed versus the azimuth; it usually shows two curves connected with the two (002) arcs. In the data analysis every curve is presented as a sum of Gaussian functions, which represent MFA distribution in the different cell walls of the fibres that constitute the wooden sample. Positions, widths and intensities of the Gaussians were obtained by fitting the model to the experimental data, according to the *Paakkari and Serimaa method* and using a MATLAB program [3] (Fig. 2).

Results and discussion

Examples of the experimental diffraction patterns and intensity profiles are reported in Fig. 3.

The curve fitting method for obtaining MFA mean values was based on the assumption that the diffraction curve measured on the (002) reflection is a superposition of Gaussian functions. This theoretical curve was then fitted to the measured intensity distribution and the numerical results are shown in Table 1.

Wood samples with lower values of MFA (<20°) gave diffraction patterns characterized by (002) arcs of small width. The intensity profiles along them showed two single narrow peaks, as there was a superposition of contributions from radial and tangential walls. On the other hand for high MFAs (>20°) diffraction patterns were characterised by wide (002) arcs, often

[1] The work has been carried out in the framework of the EU Project Compression Wood in Conifers Coordinated by Forestry Commission – Contract QLK5-CT-2001-00177

[2] Theoretically MFA distribution could be directly measured using the reflection (040), which reflecting planes are perpendicular to the longitudinal microfibril axis. Unfortunately the (040) signal is contaminated by other reflections from other close-by sets of planes [4].

separated into two or three sub-arcs. The integration along these arcs gave multi-peak intensity profiles, because of the distinct contributions from radial and tangential cell walls.

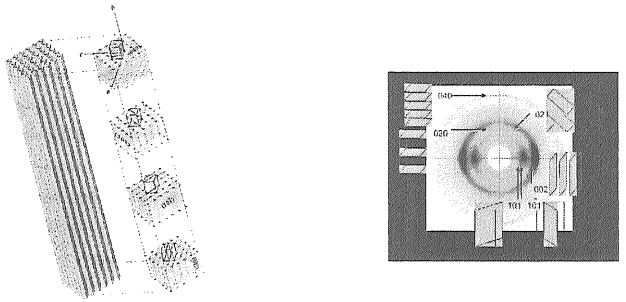


Fig. 1 Microfibril of cellulose and its unit crystalline cell, with the most important sets of crystallographic planes with an example of a diffraction pattern

Table 1. Mean microfibril angle of different samples

Provenience	Number of stem	Ring No.	mean MFA ew	mean MFA lw
Alessandria	13-5	~20	30.95(a)	27.28(b)
Souze d'oulx	11-8A	7	38.02(c)	33.99(d)
Alessandria	8-11	9	34.98(e)	31.42(f)
Souze d'oulx	2-4A	40	30.96(g)	17.84(h)
Alessandria	4-14A	21	23.59(i)	20.78(l)
Souze d'oulx	2-9A	31	20.12(m)	15.34(n)

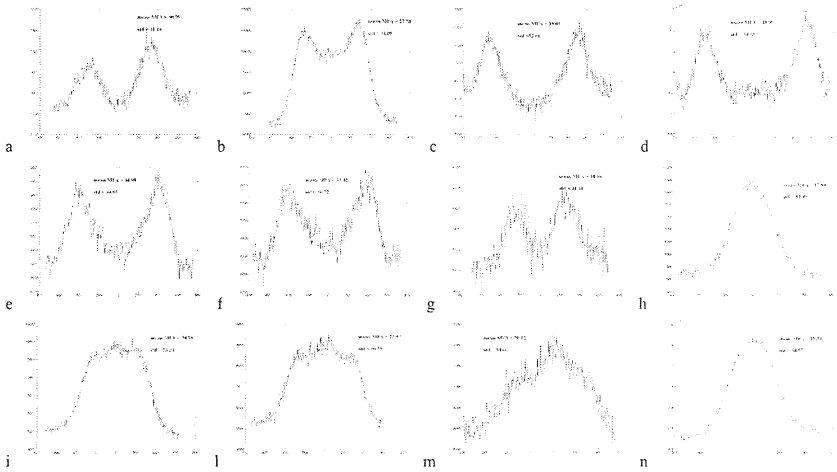


Fig.2. Experimental results and fitted curves obtained from each sample (MATLAB processing)

When spiral angles were lower, the shape of the intensity diagram showed only minor variation with the rotation of the sample around its longitudinal axis, whereas, for high MFAs, the number of peaks in the intensity curve increased. In this case, when the x-ray beam was perpendicular to the radial walls, each half of the profile consisted of three peaks where the central one was the contribution of tangential walls, and the lateral ones represented the contribute of the radial walls. As the sample was rotated, the contributions from tangential walls moved apart while those from radial walls moved inward up to 45°. Here the contribution of radial and tangential wall overlapped and the profile was again symmetric with a clear depression at the centre of each peak (Fig. 4).

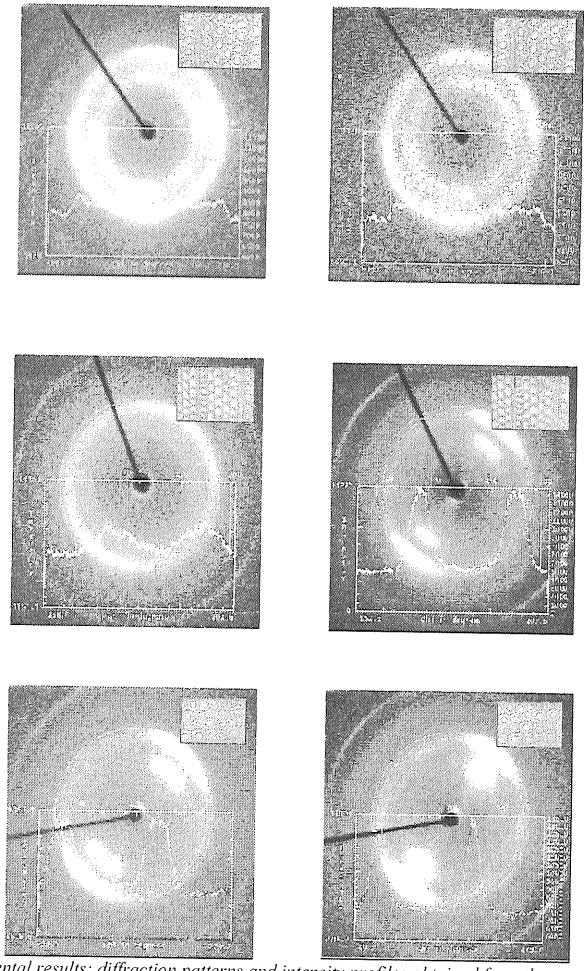


Fig.3 Experimental results: diffraction patterns and intensity profiles obtained from three specimens (left figures early wood - right figures late wood) - 1 upper: compression wood - 2 middle: probable normal wood - 3 lower: normal wood (certain).

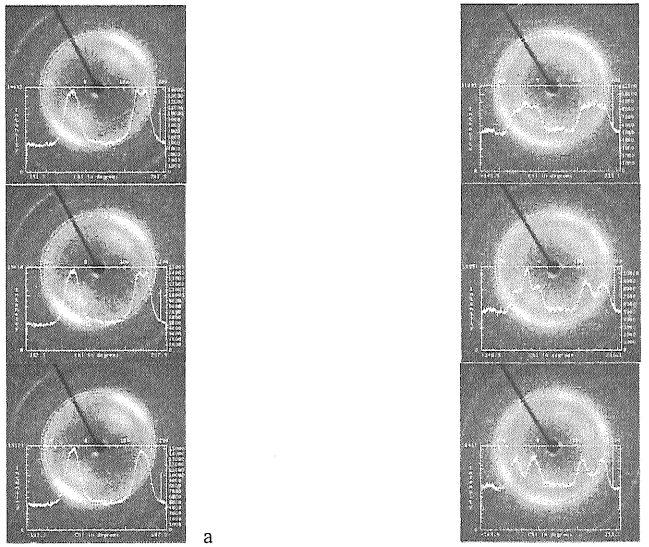


Fig.4. Effect of axial rotation on (002) profile. a) sample with mean MFA<20° b) sample with mean MFA>20°.

In theory this is true for specimens with fibres of square cross-section, as cell walls with perfect circular cross-section (maybe only in theory) would exhibit a profile consisting of two peaks that does not change during axial rotation (Fig. 5) [5].

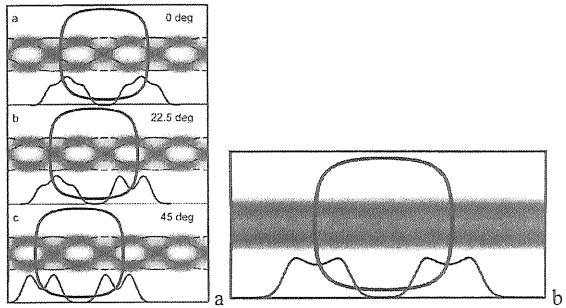
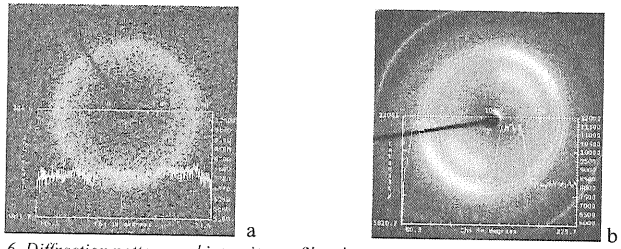


Fig.5. Theoretical effect of fibre rotation about its axis on the (002) intensity profile. a) square cross-section b) circular cross-section. [5]

According to the results obtained in this work it is possible to affirm that:
- wood rings which gave profiles consisting of single peak (in the early and late zones), are certainly formed by normal wood;
- wood rings which gave multi-peak changing diagrams only in the early zone (always characterized by higher mean values of MFA) and a single peak in the late one, are probably made by normal wood as well;
- wood rings which gave multi-peak changing diagrams in the early and latewood zones, would be compression wood or juvenile wood if the ring is close to the pith.

In theory the intensity profile should change with the longitudinal rotation of the sample as show in Fig. 4: *a)* for juvenile wood and *b)* for compression wood. As compression wood tracheids are not always perfectly circular, the experimental effect of rotation is very similar to that of a juvenile specimen. A possible solution for identifying the presence of compression wood in rings close to the pith, is to maintain a shorter exposure time (60-90 seconds): in fact compression wood normally gives worse signals than those of normal and juvenile wood (Fig.6). A partial explanation of this behaviour could be the lower presence of crystalline cellulose in CW (the index of cellulose crystallinity is 20-30% for compression wood and 40-50% for normal wood), also confirmed by lower relative maximum values in the intensity profiles. Even if it has to be considered that the parameter corresponding to the crystallinity is the total diffracted intensity (the area under the peaks, not the peak height) and if the MFA increases, giving broader arcs, the peak height decreases accordingly.



a b

Fig. 6. Diffraction pattern and intensity profile; short exposition time; a) compression wood; b) juvenile wood.

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Heat and moisture transfer in laminated wooden beams

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Abstract

This paper deals with a coupled heat and mass transfer problem for a glulam beam exposed to natural climatic conditions. The numerical analysis of transient response of moisture and temperature distribution across the cross-section is preformed by using a finite difference method. Results for one-dimensional problem are compared to experimental results and to analytical solution obtained from the literature (Chang, Weng [1], Keylwerth [2]).

Introduction

When analysing numerically the mechanical behaviour of load bearing wooden beams in natural environment, the contributions of shrinkage-swelling, temperature, viscous creep and mechano-sorptive strains are of major importance. The development of all the mentioned strains is strongly affected by the actual temperature and water content field of the beam. In this sense the determination of the spatial and temporal distribution of water content and temperature over the element according to relative humidity and temperature of the surrounding air is the first key phase of the analysis.

Experiments prove the mutual effect of the temperature and water content gradients in porous materials, but it is rarely taken into consideration in the computational analysis. In this paper the non-stationary heat and water transfer over a glulam beam exposed to natural climatic conditions is considered. The governing equations of simultaneous heat and moisture transfer in porous materials were provided by Luikov [3].

Assuming the homogeneity of the humidity and temperature field along the beam, the 2-D Luikov equations are solved for the cross-section of the glulam beam. Due to rectangular cross-section the finite difference method using an equidistant mesh of difference points is chosen for the solution. For the spatial integration the symmetric formulae based on quadratic shape functions are introduced, whereas for the time-integration linear shape functions are employed.

Governing equations

Heat and mass transfer is governed by Luikov's equations [3]. The first equation describes heat conduction with respect to the effect of phase-change and heat of adsorption and desorption, which are determined by the speed of moisture changes. The second equation describes water diffusion with respect to the effect of temperature gradients on moisture transport. Furthermore, isotropy and homogeneity of material properties across the cross-section are assumed. The equations can be written as a set of two coupled partial differential equations

$$\frac{\partial T}{\partial t} = L\left(\frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2}\right) + \nu\frac{\partial w}{\partial t} \qquad (1)$$