Deformation mechanisms of wood at the ultrastructural scale

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Abstract

This paper investigates the deformation mechanisms of wood at the ultrastructural scale. At this level, wood is composed of a periodic alternation of amorphous and crystalline cellulose fractions, embedded in a soft hemicellulose-lignin matrix. The mechanical response of wood is calculated under tensile loading conditions by means of the computational homogenisation of a representative volume element (RVE) of material. Three potential mechanisms of failure are suggested: axial straining of the crystalline fraction of cellulose, accumulation of plastic strain in the amorphous portion of cellulose and tensile rupture in the hemicellulose-lignin matrix due to cellulose fibres separation. In order to validate the present multi-scale framework, we assess successfully our numerical predictions for ultimate strains at the instant of failure with experimental values.

1 Introduction

Wood microstructure can be understood as the result of an optimisation process developed by nature over hundreds of millions years of evolution. One of its main features is its hierarchical nature distributed across multiple spatial scales, from submicrometer dimensions to macroscopic scales. This important feature has been a subject of intensive research over the last few years in applied and computational mechanics circles [1, 2, 3]. Nevertheless, despite the increasing interest in this subject and the considerable effort devoted to its description, the complete understanding of the deformation and failure mechanisms of this material at very small scales, and their implications on the macroscopic response, is still an issue which remains open at present.

The constitutive description of wood at several scales has been widely investigated by means of computational multi-scale constitutive models. In the context of elastic response, several works have been presented. Holmberg *et al.* [1] studied the mechanical behaviour of wood by means of a homogenisation-based multi-scale procedure, incorporating growth rings, irregularity in the shape of cells and anisotropy in the layered structure of cell-walls. Hofstetter *et al.* [4, 5] suggested five elementary phases for the mechanical characterisation of wood. These were hemicellulose, lignin, cellulose, with its crystalline and amorphous portions, and water. They proposed a multi-scale model and validated their numerical predictions with experimental data. Qing and Mishnaevsky Jr. [6, 7] proposed a model taking into account several scale levels and investigated the influence of microfibril angles, shape of the cell cross-section and wood density on the elastic properties of wood. Recently, Qing and Mishnaevsky Jr. [3] extended their model by incorporating progressive damage to the homogenised elasticity matrix. Additional works in this field can be found, for instance, in Ref. [8].

In spite of this extensive work, a review of the current literature shows that little research has been done in the context of irreversible processes and microscopic dissipative phenomena taking place in wood at several scales. In Ref. [2], the authors investigated the non-linear irreversible behaviour of wood cell-walls. By adopting a finite element-based computational multi-scale approach, it was shown that one important mechanism of dissipation under tensile loading is the shear plastic deformation in the hemicellulose-lignin matrix due to the reorientation of cellulose fibres induced by their alignment with respect to the loading direction.

Due to its relevance in the macroscopic mechanical response of wood and wood-based materials, our main objective in this paper is to investigate the ultrastructural mechanisms of deformation and failure in wood under tensile loading conditions by means of a computational multi-scale approach. We study the local mechanisms of deformation in wood for a wide range of initial orientation of fibres. Here, we study the failure mechanisms associated with axial straining of the crystalline cellulose fraction, accumulation of plastic strain in the amorphous portion of cellulose and tensile rupture in the hemicellulose-lignin matrix due to cellulose fibres separation. We validate the present model by comparing our numerical predictions for the strains at the instant of failure with experimental data for wood under tension.

2 Ultrastructural mechanical properties of wood

At the ultrastructural scale [9, 10, 11], the wall of wood cells contains three fundamental constituents: cellulose, hemicellulose and lignin. These constituents form a spatial arrangement called microfibril which can be represented as a periodic unit building block of rectangular cross-section (refer to Figure 1).

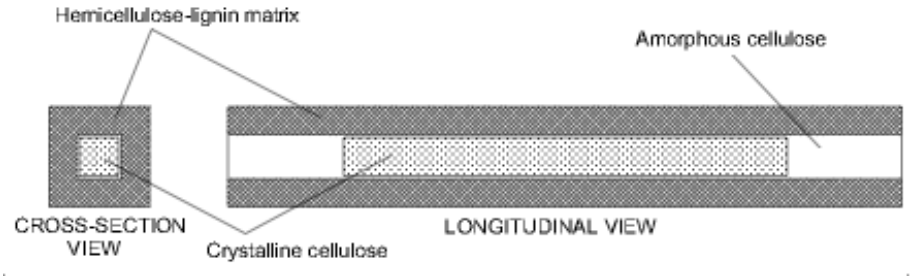

Figure 1: Schematic representation of the microfibril and basic constituents. The cross-section on the left of the figure shows the crystalline cellulose in the centre covered with an outer surface layer made up of amorphous cellulose [12], embedded in the hemicellulose-lignin matrix [13]. The longitudinal view on the right illustrates a typical periodic arrangement of crystalline and amorphous cellulose along the length of the microfibril.

Cellulose, hemicellulose and lignin constitute approximately 30%, 32.5% and 37.5%, respectively, of the total volume of wood substance for *compression wood* cells [2]. The cellulose is a long polymer composed of glucose units which is organised into periodic crystalline and amorphous regions along its length [14] (refer to Figure 1 for a representative portion of this periodic amorphous-crystalline cellulose arrangement). This periodic arrangement is further covered by an outer surface made up of amorphous cellulose [12]. The (volumetric) degree of crystallinity is defined as the ratio between the volume of crystalline cellulose and the total volume of (crystalline and amorphous) cellulose. Normally, its value varies between 0.49 and 0.60 in wood cells of Scots pine and Norway spruce, with

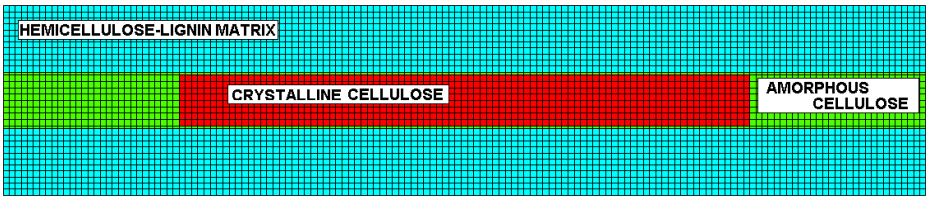

Figure 2: Typical RVE finite element mesh adopted for the computational homogenisation of wood at the ultrastructural scale.

an average value of 0.52 [15]. The specific orientation of microfibrils with respect to the longitudinal cell axis is called the microfibril angle (MFA) and is one of the most important parameters controlling the balance between stiffness and flexibility in trees. Hemicellulose is a polymer with little strength built up of sugar units, with mechanical properties highly sensitive to moisture changes. Despite its partially aligned molecular structure, it can be modelled isotropically. Lignin is an amorphous polymer whose purpose is to cement the individual cells together and to provide shear strength. Its amorphous structure makes possible the assumption of isotropy. Since the distribution of the hemicellulose and lignin in the matrix is not important, it is possible to adopt a single equivalent material for the description of the lignin-hemicellulose matrix [13]. In Ref. [16], the authors showed how the hemicellulose-lignin matrix in wood tissue and in individual cells undergoes large shear strains without apparent damage. This self-healing mechanism present in wood cells allows the assumption of no limit for the maximum shear strain in the hemicellulose-lignin matrix. Nevertheless, a maximum tensile strain of 15% can be adopted for the matrix [17], as a limit strain for the separation between cellulose fibres. For further information about the mechanical properties of the wood cell-wall constituents, we refer to Refs. [2, 19, 18].

3 Multi-scale finite element model

In this section, we consider the fully coupled two-scale finite element modelling of wood at the ultrastructural scale, subject to in-plane tensile loading conditions. The computational homogenisation scheme adopted here corresponds to the *periodic boundary displacement fluctuations* model [20], typically associated with the modelling of (heterogeneous) periodic media.

The procedure described in the following consists of modelling the mechanical response of wood at the ultrastructural scale by means of one single layer, with cellulose fibres oriented in one single direction.

The (microscopic) RVE consists of a two-dimensional periodic arrangement of crystalline and amorphous cellulose fibres embedded in the hemicellulose-lignin matrix. Figure 2 shows a typical RVE finite element mesh adopted for the computational homogenisation of the microstructure. It contains 4768 F-bar four-noded quadrilateral elements with a total number of 4950 nodes. To eliminate volumetric locking, the F-Bar methodology [21] is adopted throughout. For all of the finite element analyses we assume plane strain conditions under large strains regime.

The crystalline cellulose is assumed to be elastic. The amorphous cellulose and the hemicellulose-lignin matrix are modelled with an elastic-perfectly plastic von Mises law. For details about the mechanical properties of the basic constituents, we refer to [19].

The macroscopic problem consists of a 10×10 mm portion of material whose constitutive law is defined by the computational homogenisation of the above microstructure. Since

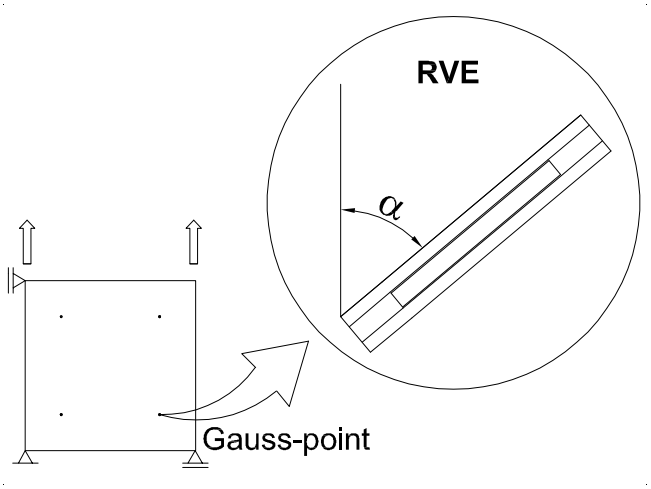

Figure 3: Macroscopic finite element mesh adopted for the computation of the actual tensile strain state at a Gauss-point.

we are interested in the mechanical response of the material, one single F-bar four-noded quadrilateral element is adopted to determine the actual tensile strain state at a macroscopic Gauss-point as illustrated in Figure 3. We note that the same approach was used by Govaert *et al.* [22] to investigate the time-dependent failure behaviour of off-axis loaded composites. Appropriate boundary constraints are imposed on the element as shown in Figure 3. In addition, since the in-plane shear strain is prevented in the wood cell-wall composite due to the interlocking between two or more adjacent cells in the wood tissue [16], the shear deformation is also prevented here by enforcing identical displacements in the x-direction at the two nodes on the left of the element (in all of the cases, x and y-axes coincide respectively with the horizontal and vertical directions).

The loading programme consists of applying a prescribed displacement in the y-direction, sufficiently large to fail the material. The total prescribed displacement is applied in 20 incremental steps. However, when no convergence is detected in the solution of the RVE equilibrium problem at any macroscopic Gauss-point, smaller load increments are taken to ensure the success of the whole macroscopic loading programme. During tensile loading conditions, the (initial) MFA will evolve into a smaller angle α as shown in Figure 3. In order to investigate the reorientation of the cellulose fibres in wood, we select a wide range of MFAs for our study, between 0 and 90^o with respect to the loading direction.

In this investigation, the total failure of wood at the ultrastructural scale is assumed to be associated with the local failure of one of the basic constituents (crystalline or amorphous cellulose, or hemicellulose-lignin matrix). When the (microscopic) strain exceeds the maximum value or ultimate strain in at least one of the constituents, the whole cell-wall composite is assumed to have failed and the corresponding numerical simulation is stopped.

The following potential mechanisms of failure [23] are suggested in this investigation. The first failure mechanism is associated to the longitudinal straining of the crystalline cellulose fraction. In this particular failure mode, the longitudinal tensile strain in the crystalline cellulose reaches the ultimate strain of 0.0014 [19]. For each load step, the longitudinal tensile strain is computed in the crystalline fraction of the microscopic finite element mesh as the change in length per unit *reference* length.

The second potential mechanism of failure is the accumulation of plastic deformation in the amorphous fraction of cellulose. The condition of failure is assumed to occur when the volume averaged equivalent plastic strain reaches the value of 0.13 [19].

A third type of failure mechanism can occur under a dominant state of tensile deformation in the hemicellulose-lignin matrix, which is normally represented by the condition of cellulose fibres separation. Here, the failure is assumed to occur when a maximum total tensile strain of 0.15 is found [19].

4 Numerical results

Our purpose in this section is to validate the present multi-scale model with published experimental data. For all the cases, the variation of the MFA during straining is computed at each load step as the current orientation of the crystalline fraction of cellulose with respect to tensile loading axis. Similarly, the macroscopic strain is calculated at each load step as the current prescribed displacement divided by the initial length of 10 mm.

Figure 4 illustrates the maximum (macroscopic) strains obtained from our multi-scale

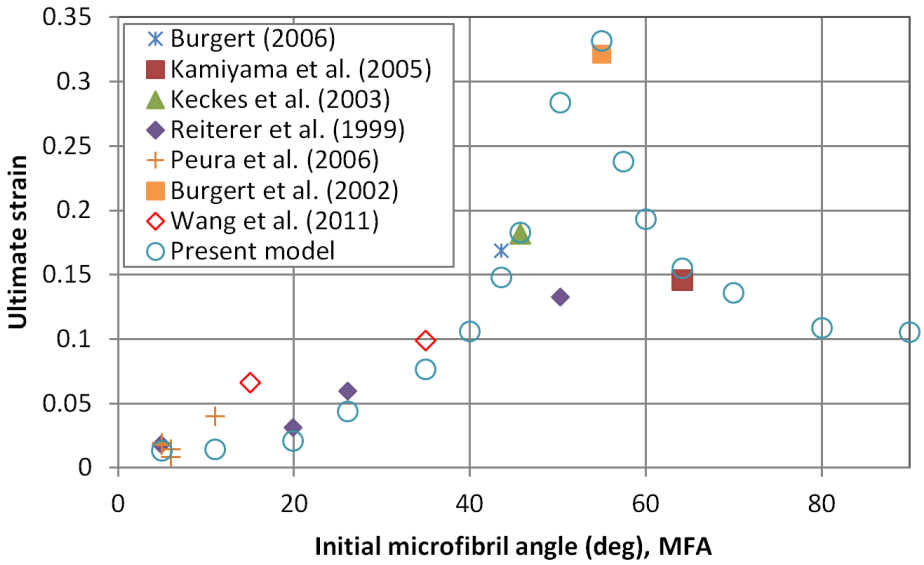

Figure 4: Ultimate tensile strains versus the MFA. Numerical predictions and some experimental results obtained from Refs. [24, 25, 16, 26, 27, 28, 29].

finite element model at the instant of failure along with some experimental values reported for a wide range of initial MFAs.

Despite the large scatter found in the mechanical properties of wood, our numerical model reveals a reasonably good agreement with the reported data. For a very small initial MFA, equal to 5^o, our numerical model predicts an ultimate strain of 0.013, which is very similar to the experimental value reported by Peura *et al.* and Reiterer *et al.*, as shown in Figure 4. Similarly, for a MFA close to 20^o, our model predicts an ultimate strain of 0.02, which is in good agreement with the result provided by Reiterer *et al.*, for the same initial MFA. We note that, the results shown in Figure 4 have been calculated with a volume fraction of cellulose equal to 50% (following Refs. [30, 31] for S_2-layer) when the MFA is smaller or equal than 20^o. For larger MFAs, we have adopted a value of 30% [32, 33].

For larger MFAs, between 20 and 45^o, our numerical results match the experimental data reasonably well. We emphasise the almost perfect matching with those values calculated for initial MFAs close to 45^o, when compared with Burgert [34] and Keckés *et al.* [16]. In fact, the exponential trend described by our multi-scale model coincides qualitatively with the trend shown by the experimental data, with small fracture strains for small MFAs

and with larger strains for MFAs approaching 45^o. It is also interesting to note that for initial MFAs smaller than 30^o, the dominant mechanism of failure is related to the axial tensile straining of the crystalline cellulose.

We note that the published experimental data for MFAs larger than 45^o is very scarce. When compared to the value reported by Reiterer *et al.*, for an MFA close to 50^o, our model overestimates the response with an ultimate strain of about 0.28. However, when compared to the fracture strains published by Burgert *et al.* and Kamiyama *et al.*, our proposed model provides an almost perfect matching. For an angle of 55^o, the predicted maximum strain is near 0.33, and for 64.2^o, the predicted value is 0.155, which are in good agreement with the experimental results shown in Figure 4.

Unfortunately, tensile experiments on wood samples with MFAs larger than 65^o have not been reported to the best of our knowledge. Therefore, more information on fracture strains as function of the MFA is needed here, probably on tissue of the S_1-layer, in which the MFA is near 90^o [35].

For very large MFAs, over 70^o, the prevailing failure mode is associated with the tensile rupture of the matrix due to the separation of cellulose fibres. In general, for values of MFA between 30 and 70^o, the failure mechanism is associated with the inelastic yielding of the amorphous portion of cellulose.

As suggested by Reiterer *et al.* [26], one of the main reasons for larger fibrillar angles in the wood cell wall is the optimisation of extensibility. For instance, the ability to develop large deformations is a fundamental feature in the branches of a tree. In order to allow large strains without failure, plants have evolved in time by adapting their microstructure to withstand mainly wind loads, self-weight and snow. At the same time, their microstructure must facilitate growth strains and adaptation to environmental conditions. For example, *compression wood* cells are typical fibres which can be found in branches of conifers. Normally, their MFA is larger than 45^o [36]. Burgert *et al.* reported initial MFAs for *compression wood* cells between 50 and 60^o [28]. Reiterer *et al.* [26] and Gindl *et al.* [37] published a value of 50^o. Burgert *et al.* [38] determined by X-ray scattering a value of 52^o. Remarkably, by observing Figure 4, we can find that our model predicts the maximum ultimate strains within this interval, at about 50-55^o. Any initial MFA out of this range will result in a smaller ultimate strain and therefore, in reduced extensibility. Consequently, our model is able to provide new clues into the understanding of how trees and plants optimise their microstructure in order to develop larger strains without apparent damage. The requirement of extensibility is of paramount importance when the material is deformed by the combined action of internal growth processes and environmental loading conditions.

5 Conclusions

This paper has investigated the deformation mechanisms of wood at the ultrastructural scale by means of a computational multi-scale approach. Three potential mechanisms of failure have been suggested under tensile loading conditions. These are: axial straining of the crystalline cellulose fraction, accumulation of plastic strain in the amorphous portion of cellulose and tensile rupture in the hemicellulose-lignin matrix due to cellulose fibres separation. Our numerical predictions for the ultimate strains in the material have been validated successfully with published experimental data. For initial MFAs smaller than 30^o, the dominant mechanism of failure is related to the axial tensile straining of the crystalline cellulose whereas for very large MFAs, over 70^o, the prevailing failure mode is associated with the tensile rupture of the matrix due to the separation of cellulose fibres. For inter-

mediate values of MFA, the failure mechanism is associated with the inelastic yielding of the amorphous portion of cellulose. We believe that the present modelling strategy, with the appropriate support of experimental work, can provide a robust platform for further investigations with a particular view to clarify unsolved issues with the microscopic dissipative mechanisms, and their influence on the macroscopic response, which still remain open at present.

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