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Constitutive modeling of a paper fiber in cyclic loading applications

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ABSTRACT

The tensile response of dense fiber-based materials like paper or paperboard is mainly dependent of the properties of the fibers, which store most of the elastic energy. In this paper, we investigate the influence of geometrical and material parameters on the mechanical response of the pulp fibers used in paper manufacturing. We developed a three-dimensional finite element model of the fiber, which accounts for microfibril orientation of cellulose fibril, and the presence of lignin in the secondary cell wall. The results showed that the change in the microfibril orientation upon axial straining is mainly a geometrical effect, and is independent of the material properties of the fiber, as long as the deformations are elastic. Plastic strain accelerates the change in microfibril orientation and thus makes it material-dependent. The results also showed that the elastic modulus of the fiber has a non-linear dependency on microfibril angle, with elastic modulus being more sensitive to the change of microfibril angle around small initial values of microfibril angles. Based on numerical results acquired from a 3D fiber model supported by available experimental evidence, we propose an anisotropic-kinematic hardening plasticity model for a fiber within a beam framework. The proposed fiber model is capable of reproducing the main features of the cyclic tensile response of a pulp fiber, such as stiffening due to changing microfibril angle. The constitutive model of the fiber was implemented in a finite-element model of the fiber network. By using the fiber network model, we estimated the level of strain that fiber segments accumulate before the typical failure strain of the entire network is reached.

1. Introduction

Paper consists of fibers and other components (such as fines, fillers, coatings and pigments) that are subjected to a complex loading history during its manufacturing process with varying tension applied at continuously varying moisture content. The inelastic features of the tensile behavior of paper are mainly governed by fibers [1,2]. However, most of the numerical analyses performed on paper neglect the load history by assuming stress-free fibers and by using the mechanical properties of fibers measured outside the network. Once the fibers are inside a dense fiber network, probing their tensile mechanical properties experimentally is no longer possible. Therefore, independently quantifying the effects on the fibers brought by the paper-making process is practically impossible. In this work, we advance in addressing a problem of accounting for the loading histories in micro-mechanical analysis of paper material starting from a fiber level. First, we propose a detailed model of a fiber that was used to quantify the changes in mechanical properties depending on the initial micro-fibril angle. Secondly, we propose a constitutive model that can capture a wide range of mechanical responses that fiber may exhibit. Finally, we incorporate the constitutive model in the fiber network model, in order to test the range of strain that can be attained by the fibers prior to network failure. As a result, by observing the changes in micro-fibril orientation, one can now relate those to the corresponding changes in the constitutive behavior of a fiber in tensile loading.

Paper pulp fibers used in paper manufacturing have a composite structure of their own. The technical process of separating fibers from raw wood destroys the original *wood* fiber structure. Pulp fibers are classified based on the extraction process; for example, chemical pulp (kraft pulp), thermomechanical (TMP), chemi-thermomechanical (CTMP) pulp, etc. Depending on the process, the original geometry and the chemical composition of the wood fibers is altered.

The original natural wood fiber consists of several layers, some of which are called S1, S2 and S3 cell wallslayers (the letter "S"

Abbreviations: MFA, microfibril angle; FEM, finite-element method; TMP, thermomechanical pulp; CTMP, chemi-thermomechanical pulp; SSC, stress–strain curve; CS, cross-section.

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standing as shorthand for "secondary"). The S2 layer is the thickest one, and represents the greatest proportion of the fiber wall thickness [3–5]. Cell walls contain not only cellulose but also a polymeric substance comprising of hemicellulose and lignin, which are significantly more compliant than cellulose, and, therefore, often treated as a matrix [6–8]. During the chemical pulping process, some of the hemicelluloses and ligninare are removed from the fiber, drastically affecting its composition. In contrast, mechanical pulping retains most of the fibers' volume apart from the surface layer. In both cases, the morphology of the pulp fiber can be defined by the remaining S2 layer and may thus be expected to describe fiber properties fairly adequately [9,10]. Thereby, the fiber's structural stiffness can mainly be characterized by the properties and the amount of cellulose microfibrils embedded in a polymeric matrix of the S2 layer, as well as their orientation along the fiber. Microfibrils are wound around the axis of the fiber and form a helical assembly in bundles. The key feature is the orientation angle of the microfibrils, also called the microfibril angle (MFA), and measured with respect to the fiber axis. This orientation angle governs the mechanical properties of the fiber (e.g. [3,5,11–13]).

In line with experimental work, there has been a strong interest in developing theoretical models that describe fiber mechanical properties in terms of MFA or cell-wall layer properties. Cave and Hutt [10] were the first to introduce a single S2 layer model for analysis of the fiber's elastic properties, followed by Mark and Gillis' [4] "two-wall" fiber model, with the allowance for different transverse strains for radial and tangential walls in coupling torsional behavior of wood fibers. A single S2 layer model with non-uniform microfibril angle along the fiber axis was also used by Navi [13] in computing the effective elastic modulus of the fiber based on its microstructure. De Young et al. [14] suggested a geometrical theory of a collapsed fiber torsional response to humidity changes, based on the observed angle of twist as a function of MFA. Torsional properties of fibers, including the effects of fiber twisting due to hygroexpansion, were measured and analyzed by Kolseth [15]. Another approach of modeling fiber, incorporating several cell-wall layers, was performed by Persson [16] taking account of essential transverse shrinkage properties of the subsequent cell wall layers. At the same time, due to the microfibril helical assembly, a fiber subjected to axial load is coupled with the relative twist (or untwisting of its helix) [17]. While the models explain much of the observed strong correlation between the microfibril angle of the cellulose fibrils in the S2 layer and structural response such as elastic modulus, extensibility of the cell wall and stress–strain behavior, there has been little attention on the quantification and integration of the coupling effect between axial and torsional deformations into analytical models.

Geometrical aggregation of microfibrils that are oriented in helical paths can also be found in other materials such as yarn [18] or textile [17]. In paper mechanics, the attempts on connecting changes of microfibril orientation upon straining resulted in simultaneous measurements of these variables with different techniques. However, it has been concluded that experimental methods involving X-ray diffraction methods for measurement of MFA are probably the most reliable [3], leaving the progress of the methods controlled by the development of the equipment [19]. In this manner, softwood samples were monitored *in situ* with a synchrotron X-ray diffraction method to account for the changes of microfibril angle, as a function of the total strain, by [20]. The results of these experiments were later used in mechanical modeling of the deformation of the wood cell wall based on shearing of the non-cellulosic matrix by Fratzl et al. [21], but were not explicitly linked with the mechanical response of paper.

In order to relate the mechanical response of fibers and paper, we first investigate the response of a single fiber as a function of fibril orientation and a fraction of crystalline cellulose. Based on the results, we propose a material model for an entire fiber, which is incorporated into a fiber network model, where each fiber is meshed with beam elements and fiber bonds are modeled through beam-to-beam contact [22]. The proposed fiber model is based on the framework of plasticity, and it is capable of reproducing the main features of the tensile response on the fiber level, such as stiffening due to changing MFA, and the observed hysteresis in cycling loading. The model is consistent with available experimental evidence and numerical results from a 3D fiber model. Apart from pure tensile behavior, we also investigated the coupling between translational and torsional motion of the fiber, which arises due to its helical structure.

2. Computational model

2.1. 3D finite-element model of fiber

In order to derive appropriate relationships needed for constitutive law, we started by considering a 3D finite-element model of a pulp fiber that accounts for the presence of microfibrils, assuming their helical orientation with a specified initial microfibril angle MFA_0 (Fig. 1). Similar models have been used in other studies focusing on either the elastic properties of the fiber [16,23] or the bonding properties [24,25]. We assumed the shape of fiber to be a hollow cylinder with a circular or elliptic cross-section, as shown in Fig. 1a and b, respectively. MFA is measured with respect to the *Z*-axis of the cylinder, and the helix angle is initially constant through the fiber.

We assumed fiber hierarchy consisting only of an S2 layer. This is a reasonable assumption for a pulp fiber in which the outer wall is partly destroyed by fiber processes [10]. The structural architecture of the S2 layer includes an assembly of cellulose, lignin and hemicellulose. A number of investigators have shown that microfibrils aggregate into bundles, which may also be classified as macrofibrils [6,26,27]. Thus, these ultrastructural components, representing cellulose contribution, are modeled with adjacent helixes. The rest of the geometrical volume of the modeled cell wall layer is filled with non-cellulosic matrix material, i.e. assuming the impact of hemicellulose, lignin and other minor constituents.

The way the components of fiber's chemical composition are arranged varies from fiber to fiber due to the nature of the wood fibers. In a quantitative analysis, Cave and Hutt [10] concluded that the cellulose volume ratio varies with the mean microfibril angle in the S2 layer. Previous studies have reported that the cellulose volume fraction in the S2 layer is roughly 50% [4,27–29]. It has been shown by Hänninen et al. [30], that the ratio between cellulose and lignin is similar in common wood species, but that the volume fraction of cell-wall constituents differs. The volume fractions are further affected by the pulping process, in which fibers undergo mechanical and chemical treatment. We took this information into consideration and compared fibers based on the pulping process. We considered chemical (bleached kraft) and thermomechanical pulp (TMP) fibers. The number of fibrils in the model was estimated according to a given cellulose volume fraction and the total number of finite elements. To ensure a random distribution of the elements and the exact proportions of cellulose/matrix volume fraction, the Fisher-Yates shuffle algorithm was used [31]. This algorithm corresponds to an iterative regression procedure, which generates a random unbiased permutation of a finite set, thus providing exact proportions of cellulose/matrix volume fractions. The two constituents of the fiber cell wall were then distributed randomly in circumferential patterns. Thus, the cell wall architecture espouses the traditional concept of concentric lamellar structure [19], as shown in Fig. 2. The employed values of cellulose versus matrix ratio are summarized in Table 1.