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Fungal degradation of softwood cell walls: Enhanced insight through micromechanical modeling

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ABSTRACT

Fungal degradation is among the greatest hazards for standing trees as well as timber constructions. Herein we aim at gaining more detailed insight into the degradation strategies of wood destroying fungi and the consequences on the mechanical performance of wood. At the macroscale, the occurring losses of mass and of mass density mask effects of altered chemical composition and microstructure. Thus, it is necessary to step down the hierarchical organization of wood to the cell wall scale in order to resolve these changes and their mechanical impact. We present a multiscale micromechanical model which is used to estimate the stiffnesses of the S2 cell wall layer and the compound middle lamella of fungal degraded wood. Data from a detailed chemical, microstructural and micromechanical characterization of white rot and brown rot degraded Scots pine sapwood is analyzed. Comparing predicted cell wall stiffnesses with measured ones confirms the suitability of the approach. The model enables to establish structure–stiffness relationships for fungal degraded wood cell walls and to test hypotheses on yet unknown effects of fungal decay. The latter include the evolution of porosity, modifications of the cell wall polymers resulting in changes of their stiffnesses, as well as increasing cell wall crystallinity. The model predictions in general showed good agreement with the predictions not considering pores in the cell wall. However, this finding does not rule out the formation of porosity. Other degradation related effects like modifications of the cell wall polymers as well as increased crystallinity have the potential to account for stiffness decreases upon the formation of pores.

1. Introduction

Fungi play a major role in the degradation processes of all biological materials in nature and thus, also in wood decay. Fungal degradation is among the greatest hazards for the integrity of standing trees as well as of interior and exterior timber constructions.

The macroscopic mechanical behavior of wood is determined by its inherent microstructure which is hierarchically organized. The levels of organization have been identified as the growth ring structure consisting of earlywood (EW) and latewood (LW), the individual wood cell walls, the cell wall layers, and the arrangement of the wood polymers in these layers in the fashion of a fiber-reinforced composite (Page, 1976; Fengel and Wegener, 2003).

The different degradation strategies of wood decaying fungi and the consequences on the wood cell wall composition and microstructure have been the subject of many research studies (Blanchette, 1984; Eriksson et al., 1990; Goodell, 2003; Lee et al., 2007; Howell et al., 2009; Yelle et al., 2011; Fuhr et al., 2012; Bader et al., 2012a). Wood rotting fungi are categorized according to their mode of degradation into brown rot, white rot or soft rot (Eriksson et al., 1990; Schwarze, 2007), whereby only brown rot and white rot are relevant for above ground applications. Brown rot fungi directly utilize the more nutritious wood polysaccharides, in initial stages especially pectic polysaccharides and hemicelluloses, and modify lignin only to a limited extent (Goodell, 2003). White rot fungi are further categorized into selective white rot fungi, which first cleave lignin to get access and then decompose the polysaccharide components in the wood cell wall, and simultaneous white rot fungi degrading all wood polymers at the same time (Blanchette, 1984).

The mechanical consequences of fungal decay have been investigated as well (Wilcox, 1978; Winandy and Morrell, 1993; Curling et al., 2002; Bader et al., 2012b). These studies almost exclusively dealt with the overall macroscopic stiffness and strength loss, which was linked to the altered wood composition

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after fungal degradation. While showing statistical correlations, such efforts do not deliver causal relations between changed microstructure and loss of mechanical properties. Such structure-stiffness relationships start to be understood for sound wood (Cave, 1968; Niklas, 1992; Bergander and Salmén, 2002; Hofsetetter and Gamstedt, 2009; Salmén and Burgert, 2009). However, it is unclear whether they also hold for deteriorated wood.

Fungal degradation of wood always features mass losses and mass density losses, which can be highly variable (Curling et al., 2002; Bader et al., 2012a). This makes investigations of structure-stiffness relationships of decaying wood difficult, as the occurring mass loss often masks effects of changing composition and microstructure in the remaining wood cell walls on the effective wood properties. Thus it is necessary to step down the hierarchical organization of wood to the cell wall scale where the fungal degradation actually occurs. This can be achieved by multiscale modeling, which is the main focus of this paper. By developing such a model based on homogenization strategies, we aim at enhancing the current understanding of the mechanical consequences of fungal degradation and to derive currently missing structure-stiffness relationships for decayed wood.

Recently we presented results from an experimental campaign, gathering microstructural as well as mechanical data of fungal degraded wood cell walls, separately for EW and LW (Wagner et al., 2014b). The investigated material (Scots pine sapwood) had been deteriorated by either brown rot (BR) or selective white rot (WR) up to mass losses of 10%. In contrast to the common observation of decreasing stiffness upon fungal degradation at the clearwood level, we found increased stiffnesses at the wood cell wall scale. Links between altered composition, microstructure and these cell wall stiffnesses were established by means of multivariate data analysis (Wagner et al., 2014b). Multiscale modeling will allow us to raise these relations from a statistical to a physical level, and to finally derive the sought causal structure–stiffness relationships. It enables to identify the effects of individual altered compositional and microstructural characteristics of wood cell walls on their respective mechanical properties. Moreover, it also allows testing hypotheses on fungal degradation mechanisms, by comparing experimentally observed stiffness changes with predictions of the model obtained for specific assumptions of altered composition and microstructure.

2. Materials and methods

2.1. Wood cell wall structure

Wood cells exhibit a multi-layered cell wall, comprising a primary cell wall (P-layer), three secondary cell wall layers (S1, S2 & S3-layer), and the middle lamella (ML). The latter connects individual wood cells and ensures the load transfer between them (Page, 1976; Fengel and Wegener, 2003). The S2 cell wall layer comprises about 80% of the total cell wall volume (Fengel and Wegener, 2003; Salmén and Burgert, 2009). Thus the cell wall layers mainly responsible for the mechanical behavior of wood are the S2 cell wall layer and the ML. Following conventions of Fengel and Wegener (2003), we consider the ML together with the adjacent P-layer and refer to it as the compound middle lamella (CML). The microstructures of the S2 layer and of the CML, respectively, exhibit different levels of organization. The S2 cell wall layer is described as a natural fiber-reinforced material, with cellulose fibers embedded in a lignin-hemicellulose matrix (Page, 1976; Salmén, 2004). The cellulose fibrils contain crystalline and amorphous regions, with typical crystallite lengths and thicknesses of 30–40 nm and 3–4 nm, respectively (Andersson et al., 2003; Peura et al., 2008). The cellulose fibrils are surrounded by glucomannan which appears to be oriented along the cellulose fibrils (Stevanic and Salmén, 2009). The so formed bundles, referred to as cellulose aggregates, range from 15 to 20 nm in diameter and are oriented in parallel and inclined to the cell axis by the so called microfibril angle (MFA) (Salmén, 2004; Fahlén and Salmén, 2005; Donaldson, 2007). The surrounding matrix material consists of lignin and xylan, which have been reported to be only partly oriented, if at all (Stevanic and Salmén, 2009). In view of the lack of qualitative data on the anisotropy, the entire matrix material will be considered to be isotropic, whereas the resulting S2 cell wall layer material is assumed to be transversely isotropic. The CML constitutes a lignified pectic layer between the cells, with some randomly oriented cellulose fibrils intruding from the adjacent P-layers (Hafrén et al., 2000; Fengel and Wegener, 2003). This structure suggests isotropic behavior of the CML. Water and extractives are incorporated in the whole cell wall. 80% of all extractives are reported to be located in the S2 layer. The amount of water in the wood cell wall is determined by the equilibrium moisture content (EMC), which depends on temperature and relative humidity (Fengel and Wegener, 2003). According to the specific composition of each cell wall layer the individual moisture contents (MC) in the S2 layer and the CML are different (Gloimüller et al., 2012).

2.2. Homogenization theory – continuum micromechanics

Homogenization theories aim at predicting properties of macroscopically homogeneous materials from their respective heterogeneous microstructure. A wide range of homogenization methods has been developed, ranging from rather simple rules of mixture to the numerical analysis of repetitive unit cells with complex internal structures. When only general statistical data about the microstructure is available but no detailed morphological information, mean field methods have proven powerful and efficient for the estimation of elastic properties (Suquet, 1997; Zaoui, 2002). The level of microstructural detail represented by mean field approaches matches well the level of knowledge of the wood cell wall microstructure, particularly in its degraded state. This motivates the use of mean field approaches in the current study. There, quasi-homogeneous subdomains, so called material phases, have to be chosen within representative volume elements (RVE). The characteristic length of the RVE l has to fulfill $l >> d$, d standing for the characteristic dimension of the inhomogeneities within the RVE, and $l << L$, L being the characteristic dimension of the geometry or loading of a structure made up by the material defined by the RVE ("separation of scales"). The mechanical properties within one RVE can then be estimated from its phases by their inherent mechanical properties, their dosages, their shapes and their interactions, using the solutions for Eshelby-type matrix-inclusion problems (Eshelby, 1957). Depending whether the phase arrangement within the RVE is either fiber-matrix like or dispersed, the Mori-Tanaka method (Mori and Tanaka, 1973; Benveniste, 1987) or the self-consistent scheme (Zaoui, 2002) is applied, respectively. If a single phase exhibits a heterogeneous microstructure itself, the mechanical properties can be estimated by introducing RVEs within this phase, with characteristic lengths $l_2 << d$, comprising smaller phases with characteristic dimensions $d_2 << l_2$. Repeatedly applied, this leads to a multiscale homogenization scheme.

2.3. Multiscale micromechanical model for sound wood cell walls

Our modeling efforts build upon an existing multiscale micromechanical model for wood stiffness (Hofstetter et al., 2005), which will be extended and adapted for degraded material. The existing model has already undergone extensive validation for sound wood