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Effects of an impregnation procedure for prevention of wood cell wall damage due to drying

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Abstract Drying of wood may lead to readily observable macroscale cracks. Recently observations were made indicating that also at the level of cell walls, damage occurs due to drying. A method is presented where green wood is impregnated using a solution of water and a bulking compound such as glycerol. Tensile strength parallel to the grain for wood impregnated in the green state was compared with that for ordinary dried wood and for wood impregnated after drying. Data demonstrate significantly higher strength for wood impregnated in the green state. It is postulated that this is due to damage in the cell walls of non-impregnated wood where the damage is induced by the drying stresses. Support for this hypothesis is also presented in the form of fractography results. For wood impregnated in the green state, damage development during drying is limited. This is because the impregnating chemical (glycerol in the present case) in the cell wall substitutes some of the moisture and therefore limits the drying stresses.

Introduction

The drying process of wood commonly leads to macroscale cracks which are readily observable. Recently we presented data in support of damage due to drying also in wood cell walls, Kifetew et al. (1998). Two sets of very thin *Pinus sylvestris* specimens were prepared and subjected to uniaxial tensile tests. One set was in the green state whereas the other set was subjected to drying and then resoaked to wet condition. The purpose of resoaking the material was to allow testing of materials with similar moisture content. Substantial differences were observed at the fracture surfaces. The dried/resoaked specimens showed a flatter and more brittle appearance. This is in support of the hypothesis that drying leads to cell wall damage.

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In a related study, Thuvander et al. (2000), hygroscopic stresses were analyzed at the scale of the individual S_1, S_2 and S_3 layers. The layers were assumed to be bonded to each other in a laminate structure. Assuming linear elasticity and using classical laminate plate theory, it was demonstrated that drying of the cell wall leads to substantial stresses in individual layers. These stresses were caused by the substantial dimensional change in individual cell wall layers if they were free to expand or contract due to changes in moisture content. In this context we introduce the term "free shrinkage" to describe the dimensional change for imaginary cell wall layers (S_1, S_2 or S_3) subjected to drying, but not bonded to neighboring layers. Although significant stress relaxation is expected in real wood, the high levels of hygroscopic stress computed in Thuvander et al. (2000) provide support for the suggested mechanism.

Recently, additional results supporting cell wall damage due to drying were obtained, Wallström and Lindberg (2000). As green specimens were impregnated with glycerate and silver nitrate, silver was found to be evenly distributed throughout cross-sections of the cell wall. However, as dried and resoaked samples were subjected to similar treatment, a more non-uniform silver distribution was observed. It was assumed that cell wall damage causes the more non-uniform distribution of silver in the dried/resoaked samples.

The objective of the present study is to study how an impregnation procedure in the green state affects failure of *Pinus sylvestris*. Such an impregnation procedure may reduce the extent of cell wall damage during drying since the free shrinkage is expected to be reduced. Our previous data demonstrate that conventional drying of wood is harmful at the level of individual cells, even one single cycle of drying leads to changes in the fracture mechanisms Kifetew et al. (1998). For this reason, bulking of the cell wall with a chemical compound is performed on green wood. The chemical replaces some of the moisture in the cell wall and thus limits the extent of shrinkage as the wood is subjected to drying.

Experimental

Green sapwood of Swedish pine (*Pinus sylvestris*) was cut by a saw to dimensions easy to handle. A conventional sledge microtome was then used to cut thin sections of thicknesses 0.2–0.8 mm in the direction apparent in Fig. 1. Each individual slice was then cut, so that the separating crack was in the longitudinal direction. This created paired specimens of similar microstructure (from the same annual ring).

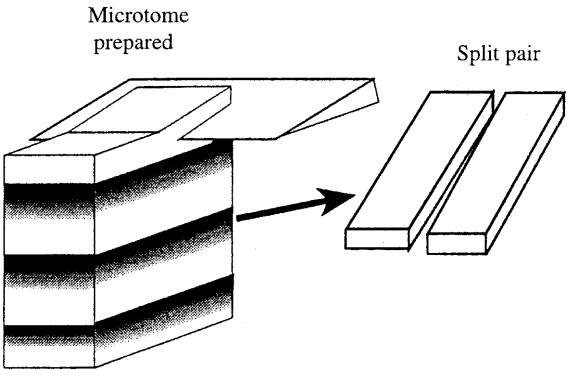

Fig. 1. Schematic illustration of specimen preparation by microtome cutting

Two separate treatments were used. In the first, one specimen was soaked in a solution of water and glycerol ("green/impr") whereas in the second, the neighboring specimen was just stored in water ("green"). The specimens were approximately 30 mm in length and 4 mm in width. Vacuum was applied during 30 minutes to the green/impr specimens in order to facilitate removal of air from the specimens subjected to impregnation. A pressure of 0.5 MPa was then applied to the container for 12 h. Both groups of specimens, green and green/impr, were then subjected to oven drying at 103 °C until no further weight loss was detected.

Specimens were then tested in uniaxial tension in the longitudinal direction using a Reith tensile stage at a displacement rate of 0.015 mm/min. Force and displacement were registered and specimens were tested until final failure due to separation. Tensile strength was calculated from the peak load and the measured cross-sectional area of the specimen. Fracture surfaces were coated by a thin gold layer in a sputter coater and observed by scanning electron microscopy.

In a second set of mechanical testing, the experimental fragmentation procedure reported in Thuvander et al. (1998) was used. Thin wood samples ($130 \times 2 \times 0.2$ mm) were bonded between plasticized PVC sheets in order to form specimens for fragmentation experiments. A similar splitting procedure as just described was used. Thus, paired specimens of similar microstructure were created with the longest wood dimension in the loading direction. As a specimen was loaded in uniaxial tension, the wood sample fractured by the formation of a crack transverse to the loading direction whereas the PVC matrix remained undamaged. Because of the high strain to failure of plasticized PVC as compared with wood, continued loading caused formation of a second fracture at the next weakest location in the wood. Several wood fracture sites were thus formed. The advantage of this method is that we obtain many data from one specimen. In Thuvander et al. (1998) the data were found to correlate well with a Weibull distribution. The major failure parameter measured directly in the experiment is strain to failure.

For the fragmentation experiments, one of the samples was kept in the green state. The other sample in the pair was dried at 103 °C for 45 minutes and then soaked in water. Vacuum was then applied to the container for 15 minutes in order to remove air. Both the dried/resoaked and the green samples were then impregnated using the glycerol/water solution and the procedure described earlier. The specimens were then dried again as previously. Fragmentation specimens were then fabricated and tested.

Results and discussion

Bulking agents can be used in order to limit dimensional changes in wood due to changes in wood moisture content (Stamm 1977). The idea is to replace moisture in the cell wall with the bulking agent. Shrinkage of the wood is then reduced due to bulking, since there is simply less moisture to be lost. An interesting example is the Swedish wooden battleship Vasa which sunk in the harbor of Stockholm in 1628 but was recovered in 1961 (Håfors 1990). In order to prevent cracking of the structure due to drying, the ship structure was subjected to a water-based solution of polyethyleneglycol (PEG) for many years. The stabilization mechanism is that PEG-molecules are present in the cell wall in order to reduce the extent of cell wall swelling and shrinkage due to variations in moisture content in the air. However, the most common industrial bulking procedure is to impregnate wood which was already dried. In the present study, we performed the impregnation step on wood in the green state in order to

Table 1. Tensile strengths parallel to grain of paired specimens. Green/dried were dried in the green state. Green/impr/dried were impregnated in the green state according to procedure in experimental section and then dried

Sample pair	Longitudinal tensile strength (Mpa)	
	Green/dried	Green/impr./dried
1	37	80
2	25	52
3	25	47
4	25	55

avoid any cell wall damage. Although there are patents describing green wood impregnation, Hudson (1951) and Stamm (1977) also discusses beneficial effects from impregnation of wood in the green state, we have not encountered any study where effects are discussed at the level of the cell wall.

The first experimental results presented are those for the tensile tests of two sets of specimens. The first set was in the green state and was then dried and tested. The second set was impregnated by the water/glycol solution in the green state and then dried and tested. In Table 1, strength data for four paired specimens are presented where each pair has a similar microstructure. The variation in strength between individual pairs of specimens is most likely due to differences in the fraction of strong, high density latewood. For each of the four specimens, the impregnated sample has at least twice the longitudinal tensile strength as compared to the specimen which was not impregnated prior to drying. It is difficult to succeed in the fabrication of good specimens, for this reason the number of specimens is limited. However, the higher strength of the impregnated specimens is apparent.

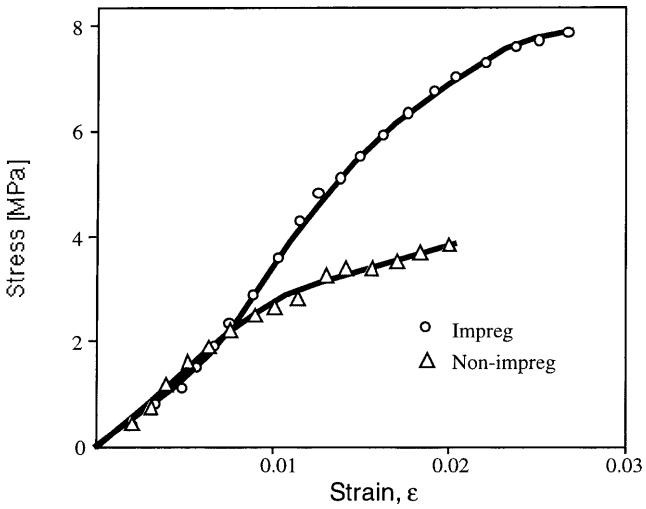

Fig. 2. Uniaxial stress-strain curves for impregnated and non-impregnated Pinus sylvestris specimens tested parallell to grain in dry state

In Fig. 2, a typical stress-strain curve for a pair of dried specimens, sample 1 in Table 1, is presented. The initial modulus is about the same for these paired specimens. This seems reasonable since they should have a similar microstructure. At a strain of close to 0.5%, the stress-strain slope suddenly decreases for the non-impregnated specimen. This indicates the development of damage entities which influence the stiffness of the specimen. One possibility is microscopic cracks in the cell walls. The non-impregnated specimen then fails at a lower strain and its strength is also less than half the strength of the impregnated specimen. Thus, not only the strength but also the work at fracture is higher for the impregnated specimen. Based on our observations of the stress-strain slopes we may also speculate that any preexisting damage due to drying primarily acts by initiating microscale cracks which form during deformation.

Although the higher strength of the impregnated specimens indicates beneficial effects from the impregnation, we desire some insight regarding the differences in failure mechanisms. In Fig. 3, fractography micrographs are presented for the non-impregnated dried specimens. In Fig. 3a, the surface appears flat and failure appears to be dominated by brittle transwall fracture. At the scale of the cell wall,

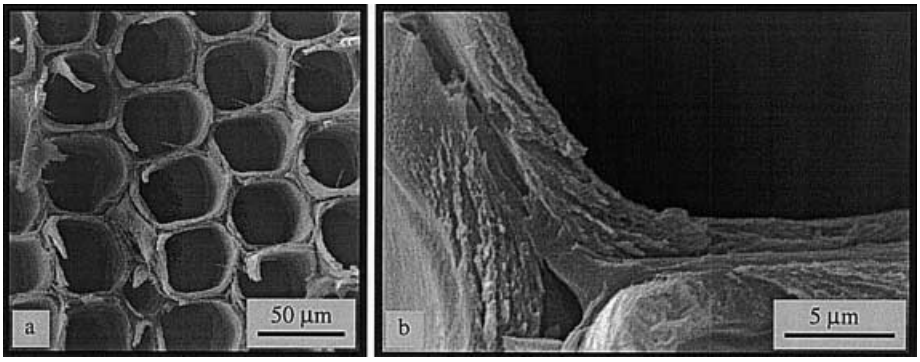

Fig. 3a, b. SEM micrographs of fracture surfaces resulting from test for tensile strength parallel to grain. Non-impregnated specimen tested in the dry state: **a** low magnification revealing cellular structure; **b** higher magnification at the scale of the cell wall thickness

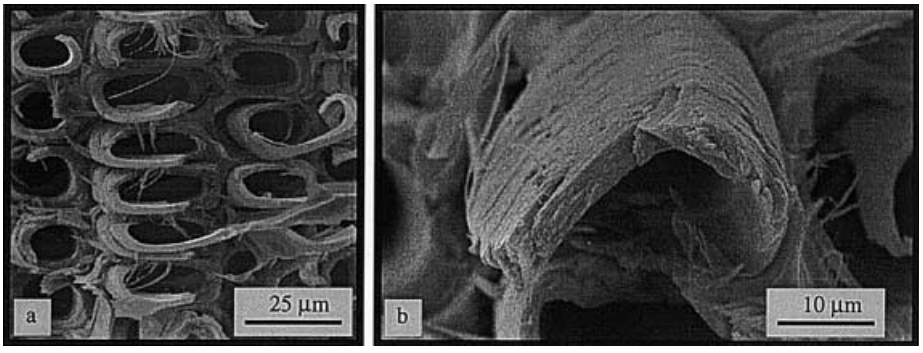

Fig. 4a, b. SEM micrographs of fracture surfaces resulting from test for tensile strength parallel to grain. Impregnated specimen tested in the dry state: **a** low magnification revealing cellular structure; **b** higher magnification at the scale of the cell wall thickness

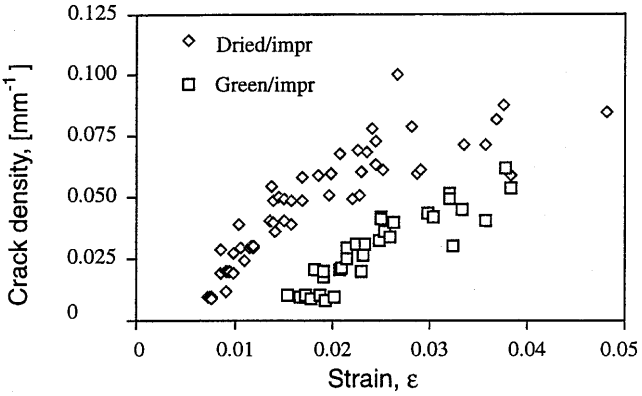

Fig. 5. Crack density (measure of number of fractures) as a function of strain for two wood materials subjected to fragmentation tests. Dried/impr was dried, resoaked, impregnated and dried. Green/impr was impregnated in the green state and then dried

see Fig. 3b, the impression of brittle fracture remains. There are no signs of more ductile tearing mechanisms.

In Fig. 4, the corresponding micrographs are presented for the impregnated and dried specimens. Here, the fracture surfaces demonstrate a very different appearance. Cracks may fail an individual cell wall layer but then often grow between two layers. Individual tracheids appear to have failed in planes away from the major crack plane so that tracheid pull-out takes place. At higher magnification, see Fig. 4b, the impression of higher toughness is preserved. The observed fractography results correlate well with the strength data in Table 1. The weaker non-impregnated dried specimens show a more brittle fracture surface appearance. The tougher appearance of the fracture surfaces for the impregnated material correlates well with the larger area under the stress-strain curve in Fig. 2.

A general problem in studies involving strength of wood is the large scatter in data. One reason for this is the large difference in density between early wood and late wood in combination with differences in annual ring width and the fraction of strong, high density late wood. Other reasons include the sensitivity of strength data to defects such as knots, ray cells etc. In the present study, we use miniature specimens in order to avoid large defects. The microtome cutting procedure is still expected to induce some microscale damage. Due to the expected data scatter, we decided to apply the previously developed technique of fragmentation testing, Thuvander et al. (1998), as described in the experimental section. Many fractures are observed in a single specimen so that each specimen typically provides in the order of ten data. The failure parameter we measure directly is the strain to failure. Because of the non-linear stress-strain curve in wood, calculation of strength will induce significant errors. At high strains, the wood specimen has a tendency to separate from the substrate close to the fracture sites so that the data become less reliable. However, at small strains, the strain in the substrate and in the wood sample are the same. Finite element calculations indicate that interaction between stress field disturbances due to the crack sites will not influence the results significantly for the first ten data. If large scale debonding occurs between the wood sample and the substrate, this is obviously not true.

In Fig. 5, data for crack density as a function of global strain are presented.

Crack density is the number of cracks observed per unit length (mm) of the wood sample. The dried/impregnated wood specimen was dried, impregnated by glycerol and dried again. The green/impregnated wood specimen was impregnated in the green state and then dried. The purpose of this comparison is to investigate if the presence of glycerol itself may have caused the increase in strength observed for the data in Table 1. According to the results in Fig. 5, this is not the case. The green/impregnated wood demonstrates much higher strain to failure than the dried/impregnated wood. The strain at formation of the first crack is about twice as high for the green/impregnated material. This excludes the possibility of any strength reinforcing ability of glycerol itself. It also demonstrates the beneficial effect of the impregnation procedure. Impregnation must be carried out on virgin material in the green state. Although we have no direct observations of cell wall damage due to drying, our microscopy results [here, in Kifetew et al. (1998) and Wallström and Lindberg (2000)] and drying stress calculations (Thuvander et al. 2000), strongly support that drying of the cell wall leads to damage. In the impregnation procedure that has been used here, the bulking agent reduces the extent of cell wall damage by taking some of the sites otherwise occupied by moisture. During drying, the glycerol stays in the cell wall thus reducing the shrinkage as compared with non-impregnated wood. Absolutely essential for the success of this procedure is that impregnation is carried out as the wood is still in its green, undamaged state.

Conclusions

A method was used where green wood was impregnated using a solution of water and a bulking compound (glycerol). Tensile strength parallel to the grain for wood impregnated in the green state was compared with that for ordinary dried wood and for wood impregnated after drying. Data demonstrate that strength and strain to failure can be twice as high for wood impregnated in the green state. The reason is that as soon as wood is dried, significant strength reduction occurs at the level of the cell wall. We argue that this is due to cell wall damage induced by the drying stresses. In addition to strength data, support for the cell wall damage hypothesis is obtained from fractography results. Drying of non-impregnated wood leads to very brittle appearance of the fracture surfaces where transwall fracture dominates.

For wood impregnated in the green state, the impregnating chemical (glycerol in the present case) in the cell wall substitutes some of the moisture and therefore limits the drying stresses. As a consequence, damage is limited so that more of the cell wall ultrastructure in green wood is preserved.

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