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Properties and set-recovery of surface densified Norway spruce and European beech

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Abstract The chemistry and wetting behaviour of surface densified wood were investigated using FT-IR spectroscopy and contact angle analyses. Furthermore, set-recovery of the surface under conditions of fluctuating humidity was measured and quantitative microscopy analyses were undertaken. FT-IR indicated that no significant chemical changes took place during the densification process. However, the wettability of the densified surfaces was significantly lower than unmodified surfaces. Following several high humidity-oven dry cycles, it was found that this densification process was almost completely reversible, i.e., there was full set-recovery.

Introduction

It is well known that many of the mechanical and physical properties of wood are correlated with its density. Being porous, the density of wood can be increased by impregnating it with some material or by compressing it. Furthermore, it is known that the wood cell walls rupture when compressed under normal conditions (RH 65%, 20°C). On the other hand, when above the glass transition point, wood can be compressed in the transverse direction without fracturing the cell walls, thereby effecting densification. Several studies have shown that rather than densifying the entire bulk of a piece of wood, only the surface can be densified (e.g., Tarkow and Seborg 1968; Inoue et al. 1990; Lamason and Gong 2007; Rautkari et al. 2008,

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2009). Tarkow and Seborg (1968) found that in a continuous surface compression process, where the temperature was raised to 260°C and then quickly cooled down, there were differences in the density profiles formed depending upon the wood species used and whether it was sapwood or heartwood being densified. A new surface densification method was presented by Inoue et al. (1990) who found that it is possible to preferentially impregnate the surface layer of wood with water and then to compress only this surface layer. Lamason and Gong (2007) investigated the effect of press closing time, temperature and compression set (densification ratio) on Brinell hardness and found that compression set was the most significant factor. There are obvious advantages modifying the surface only, notably the potential to reduced energy requirements.

Unfortunately, compressed wood springs-back without further treatment because of internal stresses. This phenomenon, known as set recovery, is the tendency for compressed wood to return to its original dimensions. Navi and Heger (2004) explain set-recovery in the following way; after wood has been compressed, significant internal stresses are locked into the helical semi-crystalline microfibrils. These stresses must be relaxed in order to avoid set-recovery. This relaxation can be done by post-treatment at high temperatures in a saturated steam environment. Treatment time reduces exponentially with increasing steam temperature (Navi and Heger 2004). Various studies on compressed solid wood (Inoue et al. 1993; Navi and Gigardet 2000; Dwianto et al. 1998, 2000; Ito et al. 1998; Welzbacher et al. 2008; Morsing 1997) have reported that set-recovery can be almost totally eliminated if a high temperature (180–200°C) post-treatment is carried out, particularly in a closed press system under humid conditions. It is also possible to relax the inner stresses by heating alone, but then the process time is much longer. Morsing (1997) found that it takes 20 h at 190°C to eliminate inner stresses by heating in an oven, but with steaming, it is possible to eliminate the inner stresses within 10 min.

Earlier studies (Rautkari et al. 2008, 2009) on surface densified wood, modified through friction, have shown that process parameters, such as temperature, pressure and time, influence the increase in surface hardness. Furthermore, an increase in the smoothness of friction densified wood surfaces can be achieved. These findings have shown that in a rather short process time, surface properties can be enhanced significantly. The study reported herein continues the earlier studies noted above.

The aim of this study was to evaluate the set-recovery of friction densified wood surfaces during a series of humid-dry cycles. Furthermore, chemical, mechanical and wettability changes to the friction densified wood surface were investigated using FT-IR spectroscopy, light microscopy and contact angle measurements, respectively. Waxed surfaces were used as a reference in contact angle measurements.

Materials and methods

Materials

Clear specimens of Norway spruce (*Picea abies* L.) and European beech (*Fagus sylvatica* L.), with average densities of 0.45 and 0.72 g/cm^3, respectively, were

used. Wood material was obtained from Switzerland and kiln dried. The specimens were cut so that the growth rings were aligned either parallel or perpendicular to the faces of the specimens. The specimen dimensions were 150 mm (longitudinal) × 20 mm (tangential) × 30 mm (radial). All specimens were conditioned for at least 1 month in a conditioning chamber set at RH 65% and 20°C prior to modification through densification. Paired specimens were used for the reference and modification. Just before surface modification, the specimens were abraded with fine sandpaper (grit size 800).

Surface modification

Surface modification was carried out on a modified Branson 2700 linear vibration-welding machine, details of which can be found in Rautkari et al. (2009). Densification was carried out at a frequency of 100 Hz and amplitude of 3 mm. The process/heating pressure was 2.2 MPa. This process pressure produced enough heat within 12 s to soften the wood material. The surface of the lower platen, where the friction was produced, was smooth polished steel. Two densification levels were obtained after the vibration procedure by applying constant pressures of either 0.58 or 2.2 MPa until the surface temperature had cooled down to 60°C, when it was assumed that the surface had solidified.

Compression set

The compression set, or densification ratio, is a measure which indicates the deformation following densification and is therefore a measure of the increase in the density of the wood. The densification ratio can be calculated using Eq. 1.

$$C = \frac{T_0 - T_C}{T_0}, \text{ when } T_0 > T_C \quad (1)$$

where T_0 and T_C are the thicknesses of the specimen before and after the densification, respectively. The thickness of the specimen was measured with a digital indicator prior to and immediately after densification.

Set-recovery

For the measurement of set-recovery two slices, each 10 mm in thickness in the longitudinal direction, were cut from each densified specimen. These specimens were dried in a fan-assisted oven at 50°C following which the dimensions were measured to an accuracy of 0.001 mm, at pre-marked positions on the specimens. The specimens were then placed in a humidity-controlled chamber (RH 97%, 22 ± 2°C) and allowed to reach equilibrium before re-drying in the same manner as described above. A total of five dry-humid cycles were conducted. After each cycle, the dimensions of the specimens were again recorded. The high humidity conditions were obtained using a saturated aqueous solution of potassium sulphate. The duration of the humid cycle was at least 7 days and the dry cycle at least 2 days to ensure equilibrium was attained. Set-recovery was calculated using Eq. 2.

$$S_r = \frac{T_r - T_c}{T_0 - T_c}, \text{ when } T_r \geq T_c \text{ and } T_0 > T_c \quad (2)$$

where T_r is the thickness of the specimen (RH 65%, 20°C) after the 5 cycles, T_c is the thickness of the compressed specimen and T_0 is the initial thickness of the specimen (RH 65%, 20°C).

Optical microscopy

Anatomical changes to the modified specimens were examined using both a Leica DMLM 25-500× epi-illumination microscope equipped with a Leica DFC 320 video camera and a Leica MZ16 7.1-11.5× stereomicroscope equipped with a Leica DFC 300 video camera. Cross-sections, 10 mm in thickness were cut from modified specimens oven dried at 50°C. Prior to inspection, the surfaces of the specimens were abraded with fine sandpaper. The densification depth was measured from 10 spruce specimens (tangential surface); five from both process pressures. It was not possible to measure the densification depth from radial surface. However, the annual ring collapse-angle of the radial spruce surfaces was measured from ten specimens, five again from both densification parameters. All the measurements were processed using Leica IM 1000 software.

Contact angle measurements

Contact angle measurements were performed with a Krüss DSA 10 contact angle-measuring instrument and analysed by a drop shape analysis program (Krüss DSA v 1.80). Distilled water was used for the drop. Measurements were made on the tangential and radial surfaces. The drop shape was recorded by video camera for 900 s along the longitudinal direction of the specimen. Contact angle was calculated automatically by the software. Measurements were performed only on early wood. Four measurements per sample were carried out and the results averaged. The measurements were made on specimens densified at high pressure (2.20 MPa) and at low pressure (0.58 MPa). The reference specimens were abraded with fine sandpaper and treated with three thin layers of wax (3032, Osmocolor) and polished. Moreover, untreated and abraded specimens were also used as a reference.

FT-IR measurements

Surface densified and untreated spruce and beech specimens were cut to approximately $30 \times 15 \times 3$ mm^3 ($L \times T \times R$) sized specimens. The untreated specimens were abraded with fine sandpaper just before the spectra were collected. Only the tangential grain oriented early wood surfaces were observed. The IR spectra were obtained using a Perkin–Elmer Spectrum 100 FT-IR (Fourier transform infrared) instrument with a universal ATR (attenuated total reflection) diamond crystal. Four IR spectra consisting of eight single scans each were recorded from the same annual rings of reference and densified specimens and averaged. The spectra were recorded in the wavenumber range of 800–4,000 cm^{-1} at resolution of

8 cm^{-1}. The analysis depth was 0.2–2 μm, depending on the wavenumber. The spectra were ATR-corrected before the analyses. Moreover, IR spectra of spruce canal resin were obtained for verifying the absorption peak in the carbonyl region.

Results and discussion

Surface modification

The results of the surface densification process are presented in Table 1 and show that the compression ratios are rather small, because of relatively low pressures and short heating time. Similar compression ratios and temperatures with the same densification parameters were found in previous work (Rautkari et al. 2008). The maximum temperatures obtained during the process, immediately after the vibration ceased, are also presented in Table 1.

Optical microscope analysis

Qualitative and quantitative analyses of the cross-sections of densified spruce and beech wood were undertaken. No changes in the densified beech wood were observed with light microscopy, most probably due to the low-densification ratio obtained and the consequent lack of any permanent cell wall deformation. No quantitative measurements were therefore possible with beech wood. In spruce, with these process parameters only early wood was densified on tangential surface (Fig. 1) and densification of the late wood was not observed. The densification depth (being the average depth measured from the surface where the early wood cells were compressed) of spruce densified in the radial direction (i.e., tangential surface), was measured. The densification depth was 2.32 (0.67) mm, when densified at high pressure and 1.23 (0.26) mm at low pressure (standard deviations in parentheses). The measurements were obtained from $5 + 5$ specimens and averaged. On the radial grain oriented spruce specimens instead of densification depth, the annual ring collapse angle was measured. It was found that there is no relationship between the process pressure (with pressures 0.58 and 2.2 MPa) and the collapse angle. The collapse angle was ranged from 0° to 53°, where 0° indicates no collapse.

Table 1 Compressing ratios in low and high process pressures (standard deviations in parentheses), compression in millimetres, and maximum temperature in the process

Samples	Temperature max (°C)	Compression ratio (%)		Compression (mm)	
		Low pressure	High pressure	Low pressure	High pressure
Spruce, radial	158 (11)	2.3 (1.8)	4.7 (2.6)	0.69	1.43
Spruce, tangential	156 (8)	2.7 (0.5)	7.1 (2.9)	0.82	2.15
Beech, radial	163 (23)	0.8 (0.1)	2.1 (0.4)	0.25	0.62
Beech, tangential	155 (13)	0.3 (0.0)	0.8 (0.7)	0.10	0.24

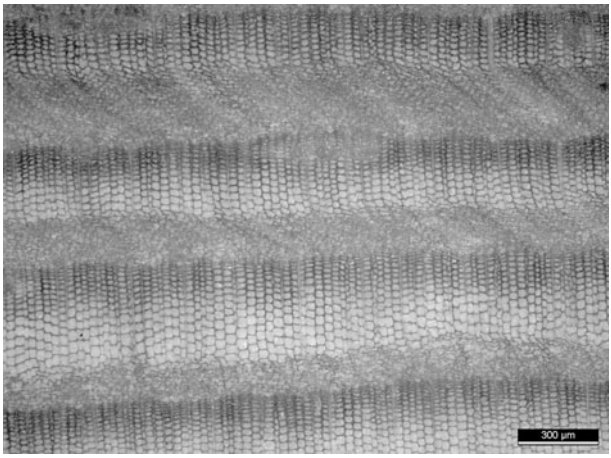

Fig. 1 Densified spruce wood with tangentially oriented annual rings. Compression direction from the top

Set-recovery

From earlier studies (Morsing 1997), a high degree of set-recovery was expected from the friction densified wood because of the relatively short treatment time and low temperatures involved. In an earlier study (Tarkow and Seborg 1968), it was demonstrated that set-recovery of surface densified wood occurs when the modified wood is subjected to conditions of high relative humidity. In this study, set-recovery was measured after five high-low humidity cycles (97% RH—oven dry at 50°C). The results are presented in Table 2. As may be observed, the hysteresis effect of wood and measurement errors have a great influence on the set-recovery measurements, since values of 100% represent total recovery and in beech wood values of more than 300% were obtained.

There are a number of possible sources of error in the measurement of set recovery if wood is locally compressed (e.g., by 0.5 mm), conditioned at high relative humidity and again reconditioned to a lower moisture content. First, there is the problem of wood hysteresis behaviour and secondly a reduction in wood hygroscopicity after repeated humidity cycles. Garćia Esteban et al. (2004) found that repeated high-low humidity cycles reduced wood hygroscopicity and therefore reduced wood swelling. Moreover, the accuracy of measurement and sample stabilization to equilibrium moisture content most probably had an influence on the results as well.

The results of how the high-low humidity cycles affect set-recovery are presented in Fig. 2. The values are presented as percentages from the initial thickness (100%). First step is initial thickness, second is thickness after compression and the following ones are after cycling between oven-dry and conditioning to RH 97% and finally, conditioning again to RH 65%. As the beech samples did not compress largely with these process parameters, the dimensional changes are almost entirely

Table 2 Set-recovery of the samples after five humid-dry cycles

Samples	Set-recovery (%)	
	Low pressure (1.58 MPa)	High pressure (2.20 MPa)
Spruce, radial	68	67
Spruce, tangential	67	71
Beech, radial	327	182
Beech, tangential	1	22

due to the swelling and shrinkage of the wood between the humid and dry cycles. Meanwhile with the spruce samples, especially those densified in the radial direction (tangential surface), the differences in set-recovery due to the high and low process pressures are clearly seen.

Anatomical changes, following soaking the specimens for 7 days and then drying, were also assessed using light microscopy. The anatomical structure of conditioned, densified, spruce wood with radially oriented annual rings is presented in Fig. 3a. The same sample after 7 days soaking in water and then drying in an oven is shown in Fig. 3b. It may be seen that the annual ring collapse following densification is almost fully recovered after the soaking–drying cycle. Densified spruce wood with tangentially oriented annual rings is presented in Fig. 4a. The same sample after 7 days soaking in water and drying in an oven is presented in Fig. 4b. The early wood can be seen to have almost totally recovered.

Contact angle measurements

The wettability of the densified surfaces was assessed by contact angle analysis. Contact angle analyses were also undertaken on waxed and untreated wood surfaces as a reference. According to EN 828 (1997), the contact angle should be measured 2 min after the droplet has been applied to the surface. The results of the contact angle measurements, measured after 2 min of application, are presented in Table 3. The contact angle on untreated wood (spruce and beech) was found to be 0°, which is a result of the rapid absorption of water on this surface. The contact angle on the tangentially and radially oriented annual rings was found to be higher on densified specimen than on the waxed specimen. No significant differences between the high- and low-densification pressures were observed. The contact angle on the densified beech wood was found to have a high standard deviation, providing more evidence that there was no significant densification of the surface. For a better understanding of the wetting of the surfaces, contact angles were plotted as a function of time. Figure 5 shows the relationship between contact and time on the radial surfaces of spruce. It was observed that on the waxed surface the droplet is absorbed more slowly than on the densified surface. Figure 6 shows contact angle as a function of time on the tangential surface of spruce. No significant difference in the absorption rate of the droplet was observed between the surface of spruce treated with wax or densified at high pressure. The surface of specimens densified at low pressure had a significantly greater absorption rate, when compared to the other treatments.

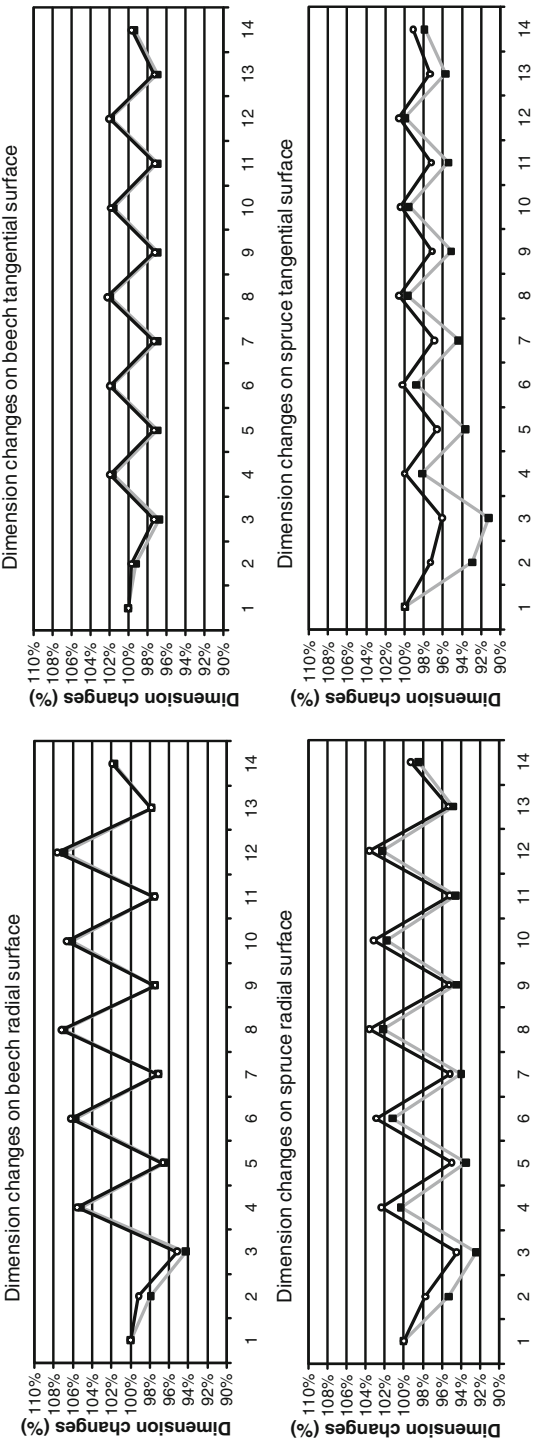

Fig. 2 Dimension changes in the densified samples following high-low humidity cycling. First step is initial dimension, second step is after compression and following ones are dimensions in high-low humidity cycles and the last step is conditioning again at RH 65% and 20°C

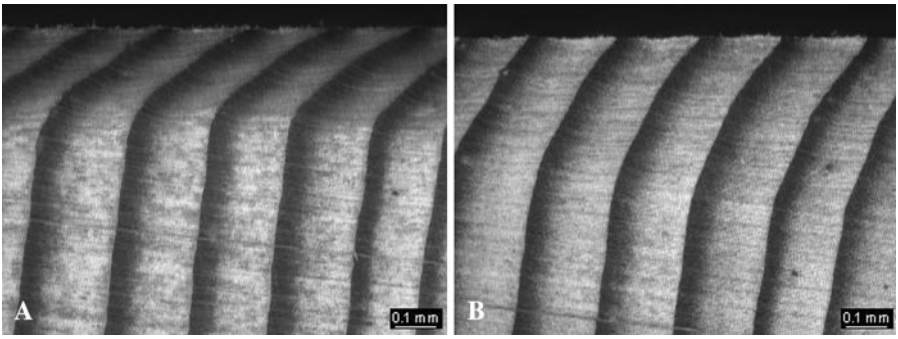

Fig. 3 Surface densified spruce with radially oriented annual rings, before (**a**) and after (**b**) water soaking

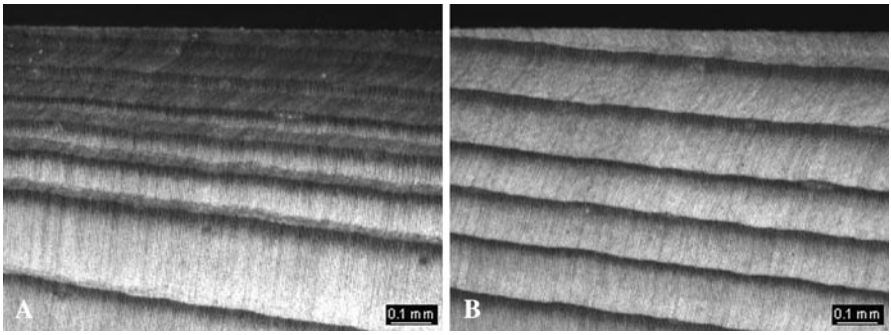

Fig. 4 Surface densified spruce with radially oriented annual rings, before (**a**) and after (**b**) water soaking

The influence of evaporation of the droplet was not specifically measured in this work, however, it is probable that the contact angle results are influenced by evaporation of the droplet to some extent. A study by Panwar et al. (2003) found that the mass of a water droplet evaporates linearly as a function of time in contact angle measurements on glass and polycarbonate due to evaporation.

Previously, contact angle measurements have shown that the surface of densified wood with linear vibration friction technology, oiled wood surfaces and wood surfaces with one layer of polyurethane varnish applied, exhibited similar wetting

Table 3 Results of contact angle measurements according to EN 828 (standard deviations in parentheses)

Samples	Spruce		Beech	
	Tangential	Radial	Tangential	Radial
Ref.	0	0	0	0
Waxed	84 (4)	88 (2)	–	–
High pressure	97 (3)	93 (3)	35 (30)	59 (14)
Low pressure	97 (3)	91 (4)	–	–

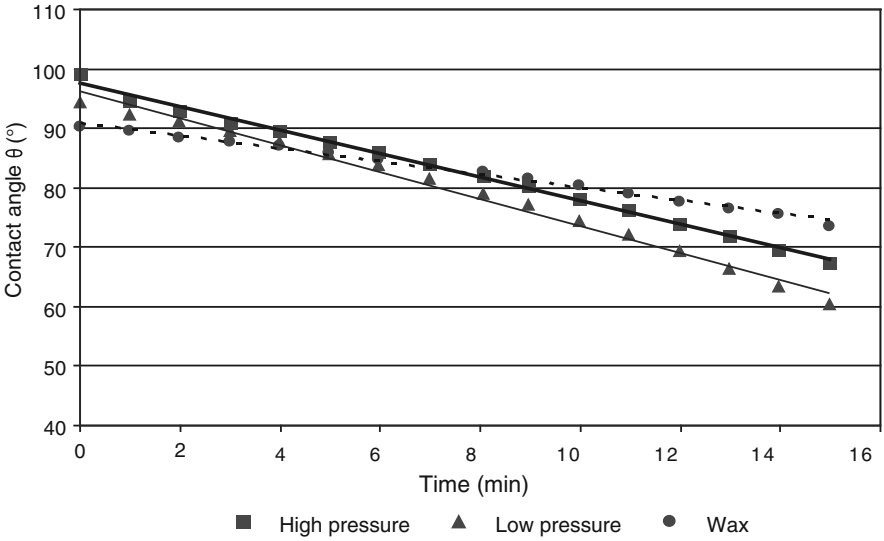

Fig. 5 Contact angle results as a function of time of surface densified spruce and waxed spruce with radially oriented annual rings. The *lines* are linear regression lines

behaviour (Pizzi et al. 2005). In the same study, untreated beech wood was found to have some water resistance after 15 min, whilst in this present study beech wood totally absorbed the water droplet in less than 2 min, differing, therefore, from the previous study. These findings are most probably due to different experimental arrangements. Moreover, in the study by Pizzi et al. (2005), the heat was generated by wood against wood with oil. Herein friction was created between wood and metal without oil.

FT-IR measurements

Densified and untreated spruce and beech specimens were investigated using FT-IR ATR spectroscopy. Figures 7 and 8 show the spectra of the densified and the reference specimens of spruce and beech. The spectra indicate that there is no significant difference between those spectra qualitatively, indicating that there are no chemical changes, detectable by FT-IR spectroscopy, occurring during the densification process. This is perhaps not unexpected given the short treatment time and relatively low temperatures involved. The quantitative differences between densified and reference spectra probably appear because the densified surface has better contact with the ATR crystal and therefore a larger contact area and higher original conduction. An earlier study (Rautkari et al. 2008) reported the glossiness increase of friction densified wood surfaces. In that study the hypothesis was that the high glossiness values might be caused by resins migrating to the surface during densification. This phenomenon was investigated with FT-IR. Densified specimens of spruce and beech were densified as explained before. The temperature on the surface, during the densification was approximately 160°C for a short time (12 s).

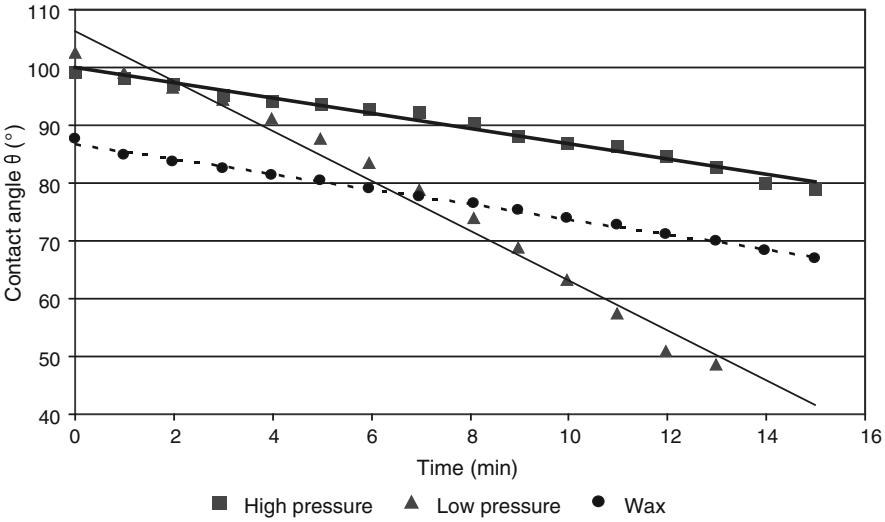

Fig. 6 Contact angle results as a function of time of surface densified spruce and waxed spruce with tangentially oriented annul rings. The *lines* are linear regression lines

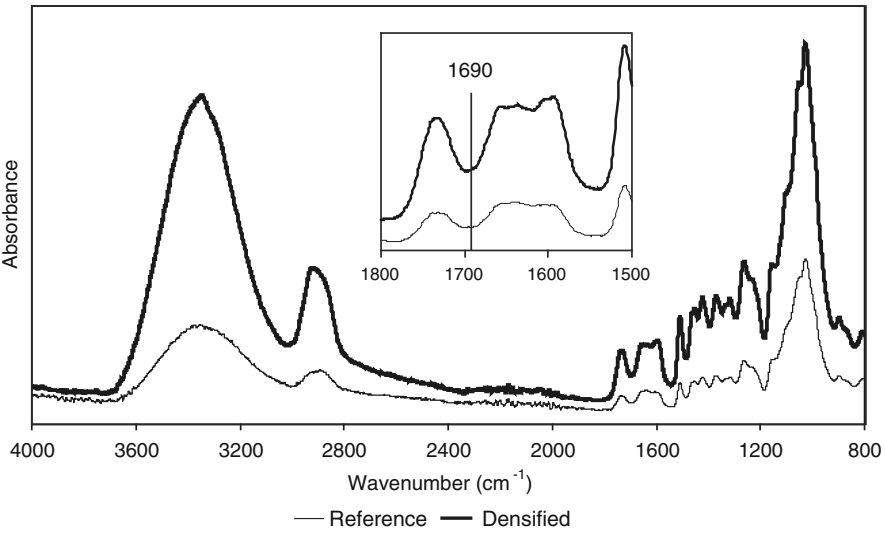

Fig. 7 FT-IR spectra of surface densified spruce and reference

Canal resin exhibited a strong absorption band in the carbonyl region at 1,690 cm^{-1} (Fig. 9). In an earlier study (Nuopponen et al. 2003), tiny resinous spots were observed in the cross-sections of the heartwood of pine heat-treated at 100°C. These resinous spots showed a strong band in the carbonyl absorption region at 1,697 cm^{-1}. The observed canal resin absorption peak at 1,690 cm^{-1} was not found with IR-spectroscopy on the densified spruce, indicating that migration of canal resin to the densified surface was unlikely (Fig. 7).

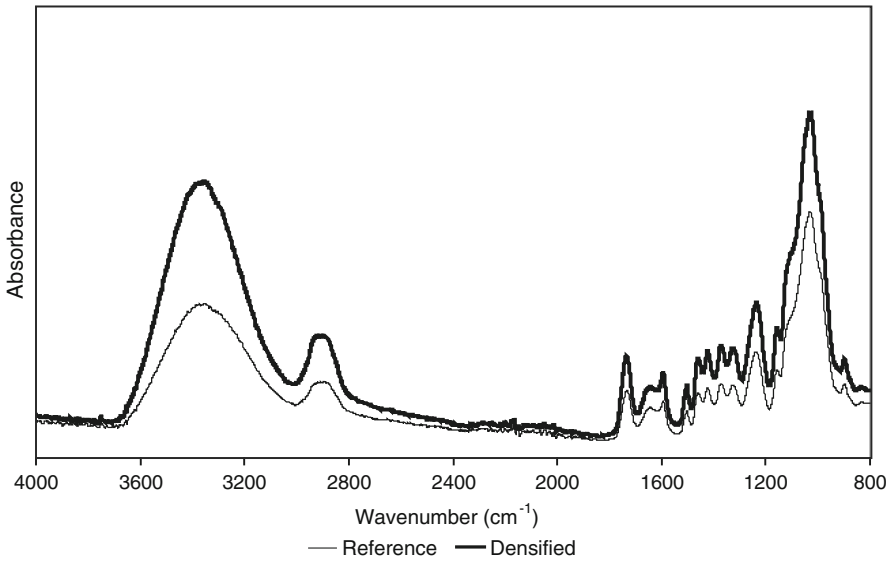

Fig. 8 FT-IR spectra of surface densified beech and reference

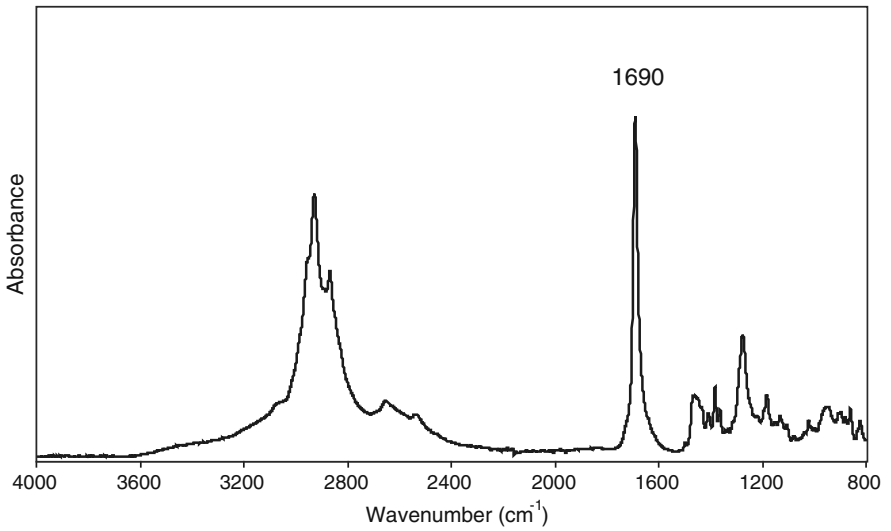

Fig. 9 FT-IR spectra of spruce canal resin droplet

Conclusion

The full set-recovery of the surface densified samples, after exposure to high-low humidity cycles, was observed. The method for calculating set-recovery, which is used for whole wood densification, is not suitable for the local densification due to low densification ratios. The wettability of the surface, as measured by water contact

angle during a 15 min period, showed that surface densified wood has rather good wetting protection when compared to waxed and non-treated wood. FT-IR shows that no significant chemical changes occur during densification. The FT-IR spectra of untreated, densified wood, and canal resin were compared and changes in the carbonyl absorption region were not found on densified surfaces, indication, therefore, that there was no migration of canal resin.

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