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Erstveröffentlichung in / First published in:

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Testing of set recovery of unmodified and furfurylated densified wood by means of water storage and alternating climate tests

Abstract: Densification is a well-known method for improving the mechanical properties of wood. In the present study, unmodified and furfurylated wood samples were densified and submitted to cyclic water storage tests and cyclic alternating climate tests. Swelling coefficients and spring-back data were determined for the evaluation of the quality of densification. The study shows that results depend on the test method applied. Simple water storage tests do not reflect the behavior of densified wood in the high humidity range. The spring-back data of unmodified samples are more influenced by the testing method than those of the furfurylated ones.

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Introduction

Densification of wood is a long-known method for improving its mechanical properties. For example, Seborg et al. (1945) reported about compressed wood with the commercial name Staypak. Compression or densification of wood is still a topic of interest (Jung et al. 2008; Kamke and Rathi 2011; Laine et al. 2013) also in the context of resistance of wood products against fungi (Skyba et al. 2008, 2009). Densification leads to plastic deformation of wood as a whole and its cells. The process is reversible, that is, the shape of untreated densified solid wood in contact with moisture approaches slowly its original form. This behavior is known as "set recovery" or "spring-back effect" (Morsing 2000; Navi and Girardet 2000). The reversibility of densification is attempted to be prevented

by different approaches, and the extent of set recovery can be evaluated by different methods.

Inoue et al. (2008) measured the set recovery by soaking densified samples for 30 min under reduced pressure in water followed by water storage for 210 min at atmospheric pressure then by water storage in boiling water for 30 min and completed by oven-drying. Ito et al. (1998) measured set recovery by means of a similar process. After the samples had been dried for 24 h at 105°C, they were stored for 30 min under reduced pressure in water and then were left in water for 24 h. This process was repeated nine times, and then the samples were treated with boiling water and finally oven-dried. Morsing (2000) vacuum impregnated wood with water (2 h) and let soaking the samples for 24 h. Then, a gentle drying process was conducted. This process was repeated six times followed by a boiling process and completed by drying. All techniques described above are based on reduced pressure for accelerating the soaking process, but there are approaches without vacuum treatment, where soaking time, water temperature, and number of cycles are the essential treatment parameters (Navi and Girardet 2000; Blomberg et al. 2006; Rautkari et al. 2011; Kutnar and Kamke 2012).

Regardless of the method applied, the objective is always the practical description of the densification quality and the behavior of densified wood in contact with water. It should be considered, however, that densified wood would not always be submitted to permanent water contact; an environment with high relative humidity (RH) is also a frequent situation in practice. Wood climatization tests under conditions of different RH are common for evaluating the dimensional stability and the anti-swelling efficiency of modified wood (Lande et al. 2004). However, these tests are seldom performed in connection with densified wood.

In the present study, cyclic water storage tests (WST) and cyclic alternating climate tests (ACT) will be conducted with unmodified and furfurylated (modified) densified samples. Furfurylation is supposed to be effective in terms of reduction of water capacity and set recovery. The furfuryl polymer, developing during the process, acts as binder

between the polymer matrix of the cell wall and fixates its densified shape. One of the objectives of this study is to detect the influence of the test method on the results of swelling parameters, spring-back, and set recovery.

Materials and methods

Beech wood (*Fagus sylvatica* L.) with dimensions of 30 mm (T), 20 mm (R), and 80 mm (L) were tested. Furfurylation was conducted according to Pfriem et al. (2012). The samples were impregnated with a solution of furfuryl alcohol and with 5% maleic anhydride as catalyst. Then, the samples were densified in a hot press at 150°C and left in the press for 1 h. A 24 h post-curing process at 103°C completed the modification process. Control samples (without modification and densification) were also considered.

The compression set (*c*) was performed to 30% and 50%:

$$c(\%)=100(T_{in}-T_0)/T_{in}, \qquad (1)$$

where T_{in} and T_0 are the dimensions in densification direction (R direction) at room temperature before and after compression.

Each test series comprised six samples. Table 1 shows the average densities (oven-dried) of the test series. The samples were cut into two parts: one half was used for WSTs and the other half for ACTs. For evaluation of cell damages due to densification, images were taken with incident light microscopy. To this end, samples were prepared before and after moisture exposure.

The steps of the WSTs were (1) oven-drying, (2) pressure reduction (0.1 bar) for 30 min, (3) addition of 20°C warm water with simultaneous vacuum release, (4) storage of the samples in water for 24 h, (5) determination of mass and dimensions of the samples, and (6) gentle oven-drying (to avoid cracks). The water storage-drying cycle was conducted three times.

For climate stress tests, oven-dried samples were stored in a climate box at 90% RH at 23°C over KNO₃ solution. A higher RH leads to condensation of water within the box and on the samples. Thus, this humidity represents the highest stress possible in atmospheric humidity. All samples remained in the climate box until all samples had reached equilibrium moisture content (EMC). The conditioning lasted up to 20 weeks. After conditioning, the mass and dimensions of the samples were determined and a gentle oven-drying was conducted. This alternating climate cycle was also conducted three times.

The densification quality was evaluated by means of the swelling coefficient R_T in densification direction and the spring-back R_S.

$$R_T(\%)=100(T_S-T_0)/T_0, \qquad (2)$$

where T_S is the swollen thickness and T_0 is the oven-dried, densified thickness in initial state.

$$R_S=(T_{1od}-T_0)/(T_{in}-T_0), \qquad (3)$$

(according to Ito et al. 1998), where T_{1od} is the radial thickness under oven-dried conditions after the first, second, and third wet cycles and T_{in} is the initial thickness of the sample before densification. Figure 1 explains the different thicknesses, which appear in Eqs. (2) and (3), and the reference distances of swelling coefficient and spring-back.

Results and discussion

Figure 2 shows the swelling coefficients of the samples obtained from the WSTs (a) and ACTs (b). In both diagrams, the two upper graphs are the unmodified densified samples. The first long slope indicates the recovery inclusive swelling; the zigzag shows the swelling-shrinkage-movement in radial direction. All values of swelling coefficients in wet state are higher in the case of WST than in the case of ACT. That is not surprising because 100% fluid water is available during WST. Therefore, a complete saturation of cell walls can be reached, whereas, at 90% RH, the cell walls are not saturated completely. Regarding the comparison of samples without densification, the difference between the maximal swelling coefficients ($R_{T,max}$) of 5% to 6% by WST and 4% by ACT can be well explained with the water availability. Water saturation of cell walls is independent of the physical state of water (fluid or gaseous). Concerning the unmodified densified samples, the R_T of WST samples is distinctly higher than of those of ACT. The swelling coefficients of WST samples amount to 36% (*c*=30%) and 73% to 75% (*c*=50%) and those of ACT samples amount to 25% (*c*=30%) and 41% to 44% (*c*=50%). The available amounts and phases of water (water vapor at 90% RH and fluid water) result in different saturation degrees of the cell walls, but the significant differences with this regard cannot be explained
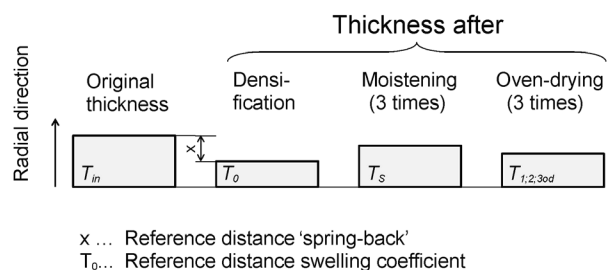
Table 1 Average densities (oven-dried) of samples after densification.

Wood	Densities for the compression sets (*c*)		
	0%	30%	50%
Unmodified (kg m⁻³)	680	910	1180
Furfurylated (kg m⁻³)	–	1120	1360

Figure 1 Illustration of the nomenclature for thicknesses in Eqs. (2) and (3).
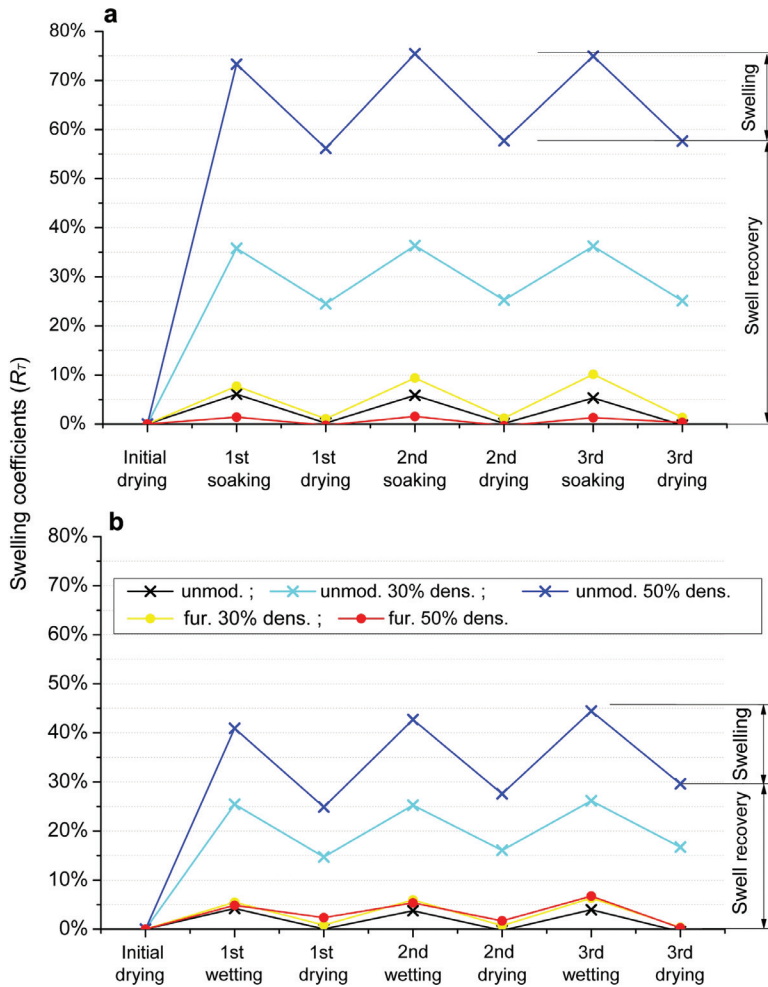
Figure 2 Swelling coefficients of (a) WSTs and (b) ACTs.

solely by this fact. On the contrary, the extent of swelling caused by the different methods (i.e., the swelling-shrinkage movement) does not differ significantly. The unmodified 30% densified WST samples have a range of swelling-shrinkage movement of 11% (after the initial swelling), and 9% to 10%, under conditions of ACT. The unmodified 50% densified samples have a swelling range of 17% and 15% to 16% for WST and ACT, respectively. An interesting fact is the dependence on swelling-shrinkage movement of the compression set. Figure 3a and b exemplarily show the cell tissues of unmodified samples with different compression sets. Obviously, the stronger densification causes more damages. The cells of the 50% densified sample (Figure 3b) suffered considerably more cracks in the cell walls than the cells of the 30% densified sample (Figure 3a). Because of the cracks, a higher radial extension is possible by swelling, which leads to a higher swelling-shrinkage movement.

The only difference between the two swelling coefficient graphs of unmodified densified samples is the initial swelling, which includes the set recovery. The set recovery comprises the decrease of plastic deformation of cells (i.e., the approach of cell and lumen shape to their original shape). In the case of WST, fluid water is soaked into the lumens due to releasing vacuum. Fluid water can immediately loosen bonds between fibrils, which were fixed during plastic deformation. Hence, the lumens are opened and reshaped. Thus, cell structure is recovered to a considerably higher degree than it was possible with the water vapor alone available at 90% RH.

The behavior of furfurylated samples is inconsistent in the case of WST and ACT. Samples with 30% densification behave like unmodified densified samples. The ACT results in lower $R_{T,max}$ (5%) compared with WST (8–10%). The alternating climate also causes a lower swelling-shrinkage movement of 5% to 6%, whereas the
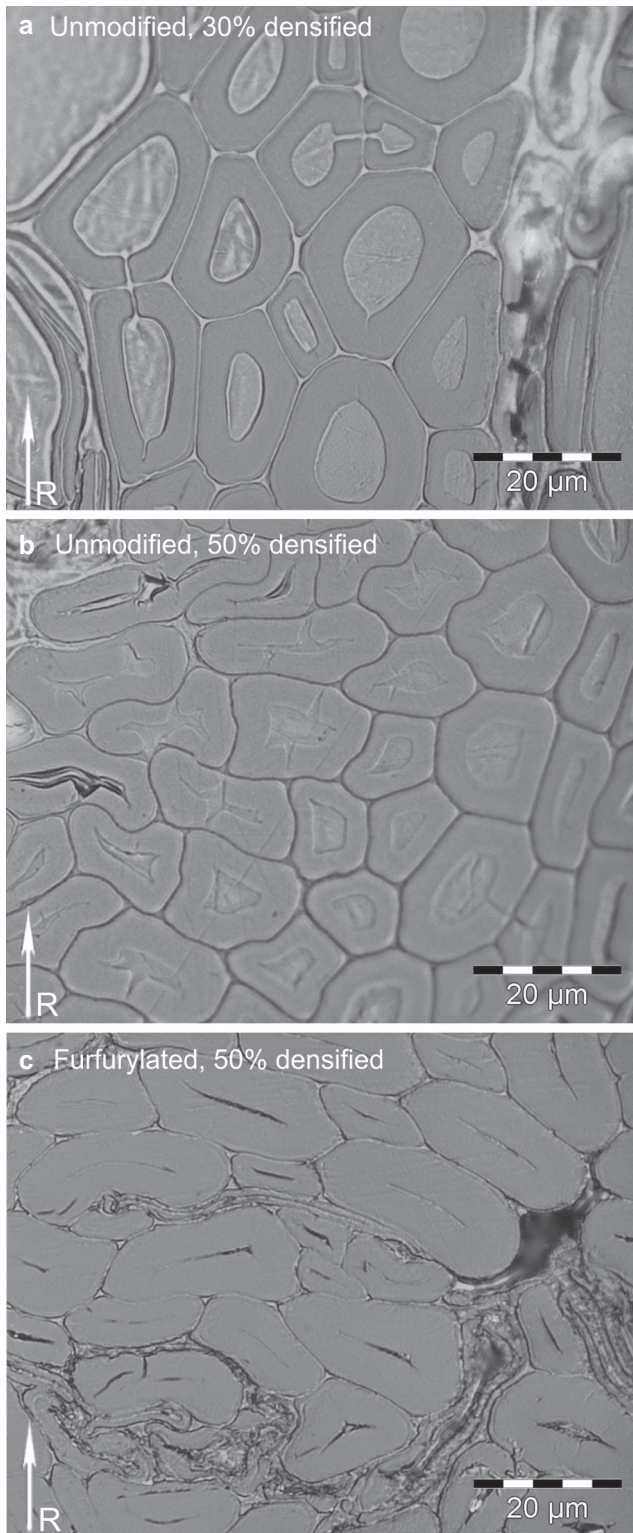
a Unmodified, 30% densified

20 μm

b Unmodified, 50% densified

20 μm

c Furfurylated, 50% densified

20 μm

Figure 3 Incident light microscope images before moisture exposure.
(a) Unmodified cell tissue, 30% densified; (b) unmodified cell tissue, 50% densified; and (c) furfurylated cell tissue, 50% densified. Arrow (*R*) indicates the radial direction.

WST causes a 7% to 8% movement. This behavior could also be well explained by the lower water content available at 90% RH conditions and in the case of 100% availability of fluid water in WST. Regarding the furfurylated samples with 50% densification, the ACT results in distinctly higher values of R_T (5–7% vs. 1.5%) and swelling-shrinkage movement (3–6% vs. 1–2%) compared with WST. Due to furfurylation, the samples behave differently in both tests. In this case, the WST acts more effectively by lower compression sets. If there are cavities, into which fluid water can penetrate, water penetration and interaction is supported by the current pressure differences, similarly to the unmodified densified samples (Figure 3a and b). In the absence of such cavities, the accessibility for fluid water is inhibited (Figure 3c). The microscopic image shows the cell structure of a furfurylated and 50% densified sample. Due to the plasticization by furfuryl alcohol, the wood is compressed to a higher degree compared with unmodified densified wood with c=50%. Most of the cells are compressed so strongly that lumens disappear. Thus, water can hardly penetrate via soaking (i.e., penetration is inhibited by furfurylation and swelling hardly occurs). The situation is different in the case of ACT. Twenty weeks passed until the 50% densified samples reached an EMC, whereas the 30% densified samples have been conditioned within 13 weeks. This fact also proves that the water accessibility is easier with a lower compression set, but this fact also shows that EMC can be reached with furfurylated 50% densified wood. However, more time is necessary for this. It can be stated that a complete swelling of furfurylated 50% densified wood requires longer water storage times.

Figure 4 shows the spring-back of the samples obtained from WST (a) and ACT (b). Regarding the unmodified densified samples, the distinctly higher spring-back due to water storage becomes obvious. The spring-back in the case of WST amounts to 82% to 84% for both 30% and 50% densified samples. Storage in ACT causes a spring-back of 50% to 55% for 30% densified samples and 41% to 48% for 50% densified samples. The reason for this behavior is the same as described above.

The spring-back increases under ACT conditions with increasing cycle number. This means that the more frequently the humidity is alternated, the higher is the densification recovery. The furfurylated samples do not behave in that way. The spring-back of WST samples amounts to 3% for 30% densified samples and 0% for 50% densified samples. Storage in ACT conditions causes a spring-back of 2% to 1% for 30% densified samples and 3% to 0% for 50% densified samples. The contrary is
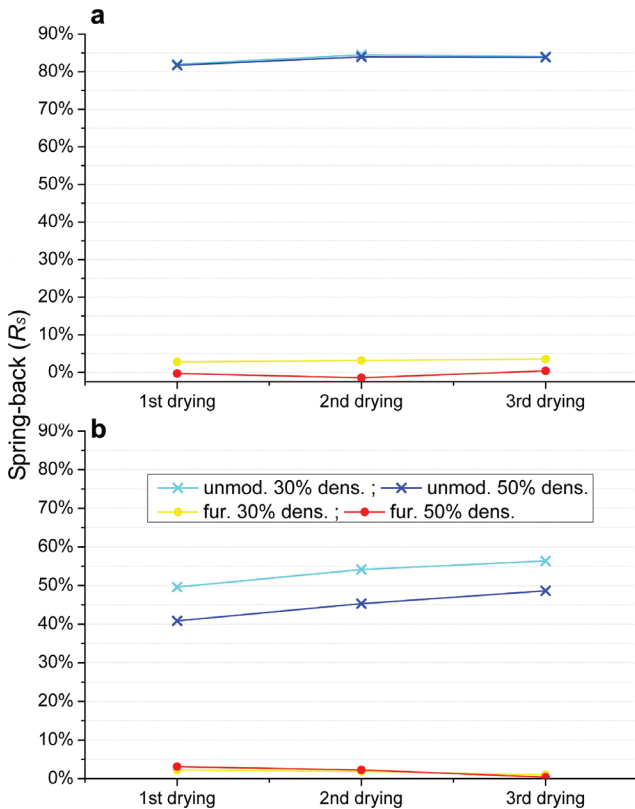
Figure 4 Spring-back data of (a) WSTs and (b) ACTs.
Data of unmodified 30% and 50% densified samples are almost equivalent; one graph superposes the other.

true for both compression sets under different storage conditions, too, but this behavior is not as obvious as in swelling coefficients. Thus, the test method does not influence the spring-back values of furfurylated densified wood considerably.

Conclusions

The storage of densified wood under conditions of high RH results in distinctly lower values of set recovery and spring-back than under conditions of water storage. The swelling-shrinkage-movement of unmodified densified samples is independent of the test method. On the contrary, this parameter is different in the case of furfurylated densified samples in WSTs and ACTs. Thus, it can be concluded that the test method has an influence on the amount of swelling of furfurylated and densified wood. The accessibility of water and water vapor to furfurylated wood is different, so that WST reflects only one aspect of spring-back behavior. Water storage of 24 h is too short for the recovery of furfurylated densified wood. It is not clear whether a 24 h WST of furfurylated 30% densified wood leads to a reliable data. However, WST or ACT gives similar results of spring-back for the furfurylated densified samples. There is no correlation between the data of WST and ACT.

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