

## MICROSCOPIC AND MACROSCOPIC SWELLING AND DIMENSIONAL STABILITY OF BEECH WOOD IMPREGNATED WITH PHENOL-FORMALDEHYDE

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### **Abstract:**

*Three phenol-formaldehydes with varying molecular weight are investigated according to their suitability to improve the dimensional stability of beech wood (*Fagus sylvatica* L.). Focus of the work is to investigate the relation of the bulking effect of the wood caused by the varying phenol-formaldehydes treatments to the corresponding dimensional stability on macroscopic level of small wood blocks compared to microscopic level of single cell walls. Therefore, beech blocks are treated with the certain phenol-formaldehydes, bulking is determined and subsequently the wood is exposed to three soaking-drying cycles. The evaluation was performed according to the anti-swelling efficiency and leaching of the phenol-formaldehydes. On microscopic level, changes of the transverse area of the cell walls due to phenol-formaldehyde treatment and subsequent water soaking are estimated. The obtained results indicate, that penetration into the cell walls are the decisive factor for dimensional stability of the treated wood. Phenol-formaldehyde residuals in the cell lumina contribute negligibly to the dimensional stability.*

**Key words:** wood modification; phenol-formaldehyde; dimensional stability.

### **INTRODUCTION**

Phenol-formaldehyde resins (PF) are widely used in wood working industries due to their beneficial characteristics, e.g. the hydrophobicity and weather resistance (Sontag and Norton 1935, Hill 2006). Impregnated into wood, PF improve the wood dimensional stability and reduce water absorption (Stamm and Seborg 1942, Gabrielli and Kamke 2010).

The primary goal of the present work is the impregnation of beech veneer (*Fagus sylvatica* L.) with PF resins in order to plasticize the treated veneers and enhancing the mouldability. Furthermore, a simultaneous temperature-induced curing is suggested to improve the form stability of the modified veneers compared to the traditionally used steaming or boiling of wood. Amongst others, the concept of cell wall swelling by PF impregnation enabling an improved mouldability has been demonstrated by e.g. Franke (2017). An easy penetration into wood cell walls was found for low molecular weight PF (L-PF) leading to an increased plasticization in contrast to high molecular weight PF (H-PF). These results are in accordance with observations from Furuno et al. (2004) and Kajita and Imamura (1991). For completion, the present study focuses on the dimensional stability of PF modified wood samples when exposed to water. In compliance with earlier studies and in accordance with Buchelt et al. (2012), measurements were conducted on cell wall level using microtome slices as well as on macroscopic level using small wood blocks made from beech. Again, three different PF resins were selected for this study representing a low (L-PF) a medium (M-PF) and a high molecular weight PF (H-PF). In order to achieve a better understanding of the mechanisms of improved dimensional stability of PF treated wood, the differences between swelling after impregnation, bulking, and dimensional stability after three soaking-drying cyclic tests were investigated and compared to the behaviour of treated single cell walls.

## MATERIAL AND METHODS

### Phenolic Resins

The used water-soluble PF are products from Prefere Resins®, Erkner, Germany. Three various PF were selected for the investigations. The main characteristic feature of a PF is the varying molecular weight (Mn) and viscosity. Thereby a low molecular weight PF (L-PF), a medium molecular weight (M-PF) and a high molecular weight phenolic resin (H-PF) were investigated.

Table 1

**Technical specification of the applied phenolic resins**

PF type	PF concentration [%]	Molecular weight Mn	Viscosity [mPas]
L-PF	45	250	13
M-PF	55	450	196
H-PF	45	890	242

### Microscopic Investigations

The microscopic investigations were performed with a light microscope (Olympus BX 41 equipped with a digital CCD-camera).

In order to compare microscopic swelling and dimensional stability of the three various PF, microscopic swelling was determined at four different states of the cell wall. Therefore, specimens were prepared from beech (*Fagus sylvatica* L.) with a transverse section of 10mmx5mm. In a first step, transversal slices with a thickness of 15 ... 20µm were prepared and subsequently dried at 80°C to constant weight. Afterwards, 30 - 40 cell wall dimensions were measured along the middle lamella and the cell wall lumina and the cell wall area was calculated by means of subtracting the outer area minus the inner area. In the next step, same specimens were saturated with deionised water and the cell wall areas were determined as described before for the same cells. In the third step, specimens were impregnated with the particular PF for 1h under vacuum conditions (100mbar). After a subsequent soaking for 24h under room conditions, the samples were cured at 150°C for 90min. In the last step, the cured specimens were saturated again with deionised water and cell wall areas were determined once more.

The area bulking coefficient (ABC) according to Buchelt et al. (2012) was determined according to equation (1).

$$ABC = \frac{Area_1 - Area_0}{Area_0} \cdot 100 [\%] \quad (1)$$

where: ABC is the area bulking coefficient [%], Area<sub>1</sub> is the area of the treated specimen [µm<sup>2</sup>] and Area<sub>0</sub> is the area of the absolute dry state before treatment [µm<sup>2</sup>].

Dimensional stability was expressed throughout the anti-swelling efficiency (ASE) in equation (3) based on the swelling coefficient (SC) displayed in equation (2) according to Rowell and Ellis (1978).

$$SC = \frac{Area_1 - Area_0}{Area_0} \cdot 100 [\%] \quad (2)$$

where: SC is the swelling coefficient [%], Area<sub>1</sub> is the area of the treated and water-soaked cell wall [µm<sup>2</sup>], Area<sub>0</sub> is the area of the absolute dry state after treatment and curing of the PF [µm<sup>2</sup>]. Respectively, SC was also determined for the untreated control.

$$ASE = \frac{SC_{ut} - SC_t}{SC_{ut}} \cdot 100 [\%] \quad (3)$$

where: ASE is the anti-swelling efficiency [%], SC<sub>ut</sub> is the swelling coefficient of the untreated and water-soaked cell wall [µm<sup>2</sup>], SC<sub>t</sub> is the swelling coefficient of the treated and water soaked cell wall [µm<sup>2</sup>].

### Macroscopic investigations

For the macroscopic investigation, beech blocks (*Fagus sylvatica* L.) with dimensions of 25mmx10mmx30mm (radial x tangential x longitudinal) were prepared. Additionally to the origin PF

concentration, PF were diluted in deionised water to a PF concentration of 20%. 12 specimens for each PF (L-PF, M-PF and H-PF) and PF concentration as well as 12 untreated control specimens were used.

Prior to impregnation, all specimens were kiln-dried to constant weight at 80°C. Subsequently, dimensions were determined at two measurement points in radial and two measurement points in tangential direction. Afterwards, specimens were impregnated with the respective PF and PF concentration and respectively with deionised water for the control group. Therefore, a vacuum of -85kPa was applied for 30min followed by a pressure of 500kPa for 90min. In order to realize a maximum penetration of the PF into the wood, specimens were kept submerged in the impregnation solution for 24h. Afterwards, the specimens were cured first at a temperature of 80°C for 90min and subsequently for 60 min at 140°C. Dimensions were measured again and the WPG was calculated according to equation (4). Furthermore, the area bulking coefficient (ABC) was determined as shown in equation (1). Hereby, radial × tangential dimensions [mm<sup>2</sup>] were used instead of the cell wall area.

$$WPG = \frac{m_1 - m_0}{m_0} \cdot 100 \text{ [%]} \quad (4)$$

where: WPG is the weight percent gain [%],  $m_1$  is the weight after curing [g] and  $m_0$  is the absolute dry weight of the untreated specimen [g].

In sum, three cycles of soaking and drying were carried out to determine the dimensional stability and leaching of the certain PF and their concentrations. During soaking the specimens were impregnated for 20min with water at a vacuum of 100mbar and afterwards kept submerged for three days in the water. It should be noted that no vacuum was applied for the first soaking cycle. After soaking, the specimens were dried for three days under laboratory conditions and finally dried in a kiln at 80°C to weight constancy. After each soaking-drying cycle, the macroscopic ASE was calculated based on the SC following equations (2) and (3) based on the radial × tangential changes of swelling in mm<sup>2</sup>. The loss of PF during the leaching cycles are expressed as leaching coefficient (LC) and were determined according to equation (5).

$$LC = \frac{WPG_1 - WPG_0}{WPG_0} \cdot 100 \text{ [%]} \quad (5)$$

where: LC is the leaching coefficient [%],  $WPG_1$  is the absolute dry WPG after the current leaching cycle [%] (tangential × radial) and  $WPG_0$  is the absolute dry WPG of the current leaching cycle of the specimens [%].

## RESULTS AND DISCUSSION

Beech wood treated with 20% L-PF and M-PF display similar WPG after curing (Tab. 2), whereas H-PF treated samples display lower WPG values at 20% PF concentration. It is conceivable that H-PF penetration into the wood is retarded due to a higher molecular weight and a corresponding higher viscosity. After exposure to three leaching-drying cycles, a great amount of the impregnated H-PF has been washed out. The LC displays values of 39.6% (45% H-PF concentration), respectively 27.2% (20% H-PF concentration). In contrast, L-PF and M-PF treated samples displayed a minor weight loss due to leaching (Table 2). A similar leaching behaviour has been observed before for wood treated with same PF exposed to an artificial weathering test (Franke et al. 2017). Furuno et al. (2004) claim that lower molecular weight fractions of PF can penetrate the wood cell wall, whereas fractions of higher molecular weight remain in the cell lumina. Similar suggestions are found by Wallström and Lindberg (1999) for pine (*Pinus sylvestris* L.) treated with polyethylene glycol. Consequently, PF in the cell lumina is leached out more easily, whereas PF in the cell walls might be more stable. Hill (2006) concluded that PF molecules form three-dimensional networks within the cell walls and thus, resist leaching. Additionally, Yelle and Ralph (2016) claimed covalent bonding between lignin and PF. This can also contribute to the stability of PF polymer inside the cell walls.

Considering the penetration into the cell wall, Fig. 1 displays the bulking of the wood blocks (at various PF concentrations) and the cell walls (PF concentration: L-PF = 45%, M-PF = 55%, H-PF = 45%) by means of the ABC. In general, the cell walls show a higher degree of swelling compared to macroscopic swelling of the wood blocks. Furthermore, it is shown that even the H-PF can induce a bulking effect to the wood cell walls, indicating a partial penetration into the cell walls. However, the macroscopic ABC values of the H-PF treated wood are lower than those of the L-PF and the M-PF. L-PF affects highest swelling of the material. The slightly higher concentration of the M-PF impregnation solution (55%) results in a higher WPG but lower ABC values. These results are in accordance with claims from Furuno et al. (2004) suggesting that only smaller fractions of PF penetrate the wood cell walls, whereas greater molecular fractions remain in the cell lumina. A comparison of microscopic ABC and macroscopic ABC values demonstrate a cell wall swelling being at least twice as high as the macroscopic swelling at similar PF concentrations of L-PF and M-PF. In contrast, the H-PF modified samples display a different behaviour. Macroscopic swelling at 45% PF concentration is one sixth lower than the swelling of the cell wall. As previously assumed, the penetration of

H-PF into the wood might be limited due to the high molecular size and high viscosity. At 20% PF concentration L-PF and M-PF treatment show a reduction of the ABC. Yet, swelling values are still around 10%. In contrast, the macroscopic ABC for the H-PF treated samples at 20% PF concentration display negative values resulting from the restricted penetration H-PF into the cell walls and a stress-induced collapse of the wood might occurred during curing of the PF at 140°C. A similar material behaviour of wood impregnated with H-PF was also observed by Furuno et al. (2004).

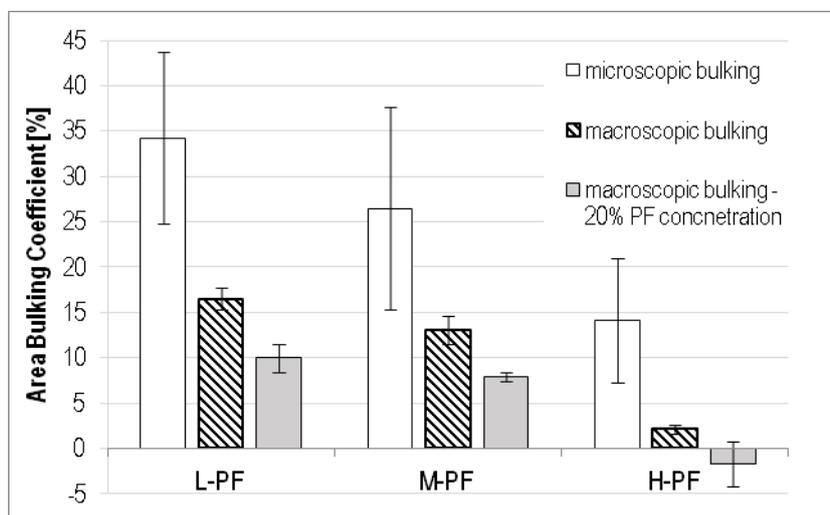


Fig. 1.

**Area bulking coefficient (ABC) of the PF treated beech cell walls (microscopic bulking) and of PF treated beech blocks (macroscopic bulking) at a PF concentration of L-PF 45%, M-PF of 45% and H-PF 45% and wood blocks at 20% PF concentration (macroscopic bulking – 20% PF concentration).**

Table 2

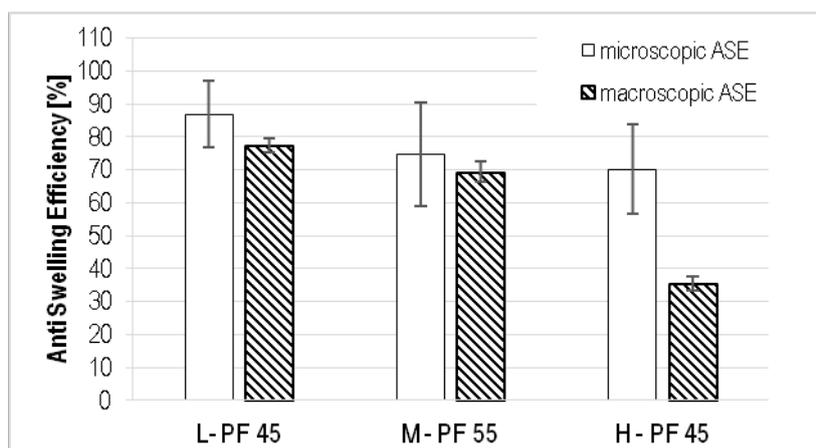
**Weight percent gain (WPG) after curing and after the third soaking-drying cycle, leaching coefficient (LC) and the ASE of beech blocks treated with the various investigated PF at various PF concentrations as well as the anti-swelling efficiency (ASE) after each cycle\***

PF type	PF concentration [%]	WPG after curing [%]	WPG after the 3 <sup>rd</sup> cycle [%]	LC [%]	ASE 1 <sup>st</sup> cycle [%]	ASE 2 <sup>nd</sup> cycle [%]	ASE 3 <sup>rd</sup> cycle [%]
L-PF	45	53.5 (3.0)	51.1 (3.0)	4.6	77.4 (2.2)	72.3 (1.2)	70.6 (1.2)
	20	26.3 (1.0)	24.3 (1.0)	8.4	65.1 (1.1)	62.8 (2.3)	58.6 (3.1)
M-PF	55	59.3 (2.2)	56.0 (6.9)	5.8	69.4 (3.1)	65.7 (1.3)	63.7 (3.5)
	20	25.1 (0.5)	24.1 (6.9)	4.2	60.5 (1.1)	55.6 (1.3)	53.1 (1.3)
H-PF	45	46.9 (4.3)	33.6 (0.5)	39.6	35.5 (2.2)	-2.1 (2.7)	-11.6 (2.9)
	20	22.2 (3.7)	17.4 (3.6)	27.2	23.6 (2.6)	-0.3 (3.1)	-6.3 (4.5)

\*standard deviation in parenthesis

The dimensional stability of the wood blocks and cell walls of the microtome sections after the first water soaking is expressed by the ASE. Results are displayed in Fig. 2 showing that the macroscopic ASE of the wood blocks treated with higher concentrations of L-PF (ASE=77%) and M-PF (ASE=69%) display a similar ASE like on microscopic level (L-PF 86% and M-PF 74%). Furthermore, L-PF treatment affects a higher ASE at microscopic and macroscopic level compared to the ASE of M-PF treated wood. Similar findings were described by Deka and Saikia (2000) and Anwar et al. (2009) claiming similar macroscopic ASE values for various wood species treated with commercial low molecular weight PF. However, significantly lower ASE were obtained from H-PF treated samples. Here, the macroscopic ASE was determined with 35%, whereas the microscopic ASE was appr. 70%. As a result, H-PF treated beech wood displays a considerably higher microscopic dimensional stability compared to the macroscopic dimensional stability. However, a high variation was obtained for the microscopic ASE values and, thus, also for the SC at microscopic scale. Similar trends were mentioned by Buchelt et al. (2012) for furfuryl alcohol treated beech cell walls. These authors explained the finding with the varying restriction of swelling caused by surrounding cells and the varying chemical composition of each cell wall. Additionally, the degree of penetration of the PF into a single cell wall remains unknown and might also contribute to the high variation of the measured cell walls. Thus, the obtained microscopic ASE values should be interpreted with caution. Nevertheless, a trend is observable showing that H-PF can basically increase the dimensional stability due to cell wall penetration. Thus, dimensional stability might be mainly evoked by the PF molecules which are small enough to penetrate into the cell walls (Furuno et al. 2004, Anwar et al. 2009) and minor due to PF residuals in the cell lumina which are suspected to limit liquid flow into the wood. However, during the exposure of the treated wood to cyclic soaking and drying stress, it could be shown that the macroscopic ASE further decreases for all applied PF types at all concentrations (Tab. 2).

As discussed for the leaching of the various PF, where the H-PF treated samples display a high LC during the leaching cycles, the macroscopic ASE of H-PF treated wood result in negative values after the second leaching cycle. At the same time, the L-PF and the M-PF treated samples show only a slight decrease of their initial ASE (appr. 7% for both applied PF concentrations). This finding additionally supports the hypothesis of an insufficient penetration of H-PF into the cell wall. Furthermore, residual PF in the cell lumina hinder the liquid flow of the water through the wood after the first soaking. This effect becomes less pronounced after leaching of the PF. Thus, not only PF inside the cell walls affect the dimensional stability, but also the PF inside the lumina may limit the liquid flow affecting the dimensional stability. However, this effect is not persistent. More work should be carried out on leaching of the certain PF out of single cell walls and its effect on the dimensional stability.



**Fig. 2.**

**Anti-swelling efficiency of the PF treated beech cell walls (microscopic ASE) and of PF treated beech blocks (macroscopic ASE). Where the number stands for the PF concentration of the certain PF.**

## CONCLUSIONS

In this study, three PF with varying molecular weight were investigated in respect to their potential to improve the dimensional stability of beech wood. According to the obtained results, it can be concluded that:

- L-PF and M-PF can penetrate into the wood cell wall to high degree, whereas H-PF basically has the potential to penetrate wood cell walls, but it seems that the penetration into the wood is limited at the macroscopic level.

- The dimensional stability of beech treated with L-PF and M-PF is improved to the same extent at macroscopic and on microscopic cell wall level. Whereas, H-PF displays a clearly lower dimensional stability on macroscopic level than on microscopic level. It can be concluded, that penetration and a sufficient cross-linking of the smaller molecular weight fractions inside the cell walls are the decisive factor for dimensional stability.
- During three soaking-drying cycles, only minor amounts of L-PF and M-PF were washed out, corresponding with a negligible decrease of the macroscopic dimensional stability.
- In contrast, H-PF is leached out in high amounts during the three soaking-drying cycles and simultaneously the dimensional stability results in negative values. This indicates an insufficient interaction with the wood cell wall components. Furthermore, those PF residuals in cell lumina might initially increase the dimensional stability, but this effect is not persistent.

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