The impact of primers for wood bonding on beech wood's Young's modulus

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Abstract

When producing load-bearing timber products with one-component polyurethane adhesives and certain wood species, e.g., beech or larch, primers can be used to obtain stronger and more durable bonds. The active chemicals of three existing primer systems, *hydroxymethylated resorcinol formaldehyde* (HMR), *polysorbate 20* (PS20), and *poly(ethylene glycol)* (PEG), were used to impregnate beech wood. Young's modulus (E) in the radial direction was determined in tensile mode. A modified E can influence the deformation and stress distribution at the wood-glue interface. Compared to the untreated wood, the average E was reduced by 7% for the water-treated reference and the HMR treatment. With the PS20 treatment, the average E was reduced by 16%, compared to the untreated wood, and by 45% with the PEG treatment.

1 Introduction

In today's production of load-bearing engineered wood products (EWPs), e.g., glued laminated timber, one-component polyurethane (1c-PUR) adhesives are widely used, especially due to their short curing times, which allow faster production. Spruce is the predominant wood species in the production of EWPs in Europe. However, ongoing changes in silviculture, away from spruce monocultures to a broader variety of tree species, driven by climate change, bark beetle infestations and biodiversity aims, will reduce the availability of spruce in the upcoming decades. The reduced quantities of spruce increase the interest in producing EWPs from currently less used species, or those regarded disadvantageous (problematic) for constructive bonding. To comply with the legal requirements defined for the bond performance

of load-bearing wood products, with certain wood species, a primer has to be applied prior to the adhesive. While different publications confirm the effectiveness of those primers (Bockel et al. 2020; Böger et al. 2024; Christiansen 2000), the functionality of the primers remains vague. Nowadays, three different primer systems can be identified as relevant. The *hydroxymethylated resorcinol* (HMR) primer is the scientifically most examined system, which is often prepared in a 2-stage method described by Christiansen (2000). The Henkel AG & Co. KGaA (Düsseldorf, Germany) offers two different waterborne primer systems: the LOCTITE® PR 3105 PURBOND and the LOCTITE® PR 7010 PURBOND that contain polysorbate 20 (PS20) and poly(ethylene glycol) (PEG), respectively, as their main active chemical. Böger et al. (2024) observed in swelling experiments indications for a plasticization of the treated wood with those primers, which could lead to a more beneficial distribution of stresses within the bond line region.

This publication provides data on Young's modulus (E) of primer-treated wood, to quantify possible favorable plasticization effects. Therefore, tensile tests with the load in the radial direction, meaning in the LT plane, were conducted. The radial direction was chosen because possible plasticization was expected to occur predominately in the amorphous lignin and hemicellulose domains. Even though lignin is predominantly responsible for compression strength within the wood structure, the impact of an expected plasticization can also be observed in tensile testing. Therefore, the most commonly used longitudinal test direction, which is oriented

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along the fibers, was discarded due to the huge influence of the unaffected crystalline cellulose domains that cannot be swollen by the primers (Salmén 2018).

2 Materials and methods

Defect-free boards from the same beech tree were initially conditioned at 20 °C/65% RH. The boards were planed to 10 mm, and specimens of $10 \times 10 \times 100$ mm³ (L × T × R) were sawn. The specimens were sampled to include minimal variance between specimens' individual density (765 ± 7 kg/m³), similar year ring patterns, and a clear distinguishment between the three main directions of the wood (Fig. 1).

The specimens were divided into five random groups of $n = 30$, with the groups' average density only varying by ± 1 kg/m³. While one group was used unaltered, the other groups were impregnated in (*i*) deionized water, (*ii*) a 50% solution of PS20, (*iii*) a 50% solution of PEG (600 g/mol), and (*iv*) HMR directly after preparing the B-stage (reactive mixture after adding the formaldehyde) as described by Christiansen (2000). For the impregnation, the specimens were placed in a beaker and submerged in 1 L of the respective liquids. The beaker was placed for 2 h into an evacuated desiccator and subsequently for 20 h at ambient pressure. Due to its reactivity in the B-stage in the case of HMR, the time at ambient pressure was reduced to 2 h to avoid the formation of a bakelite-like, solid layer on the specimen's surfaces. After removal from the beaker, the excess liquid on the specimen surfaces was carefully removed with a paper towel. Afterward, the specimens' mass was measured in the wet stage and after acclimatization to mass constancy

at 20 ± 1 °C /65 ± 3% RH, which also was the test climate. In preparation for strain measurement with the digital image correlation (DIC) system, a thin acrylic coating with a stochastic dot pattern of 10×20 mm² (T × R) was spray-painted in the center of the specimen's RT-surface (Fig. 1). Following, the specimens were acclimatized to mass constancy again.

Wedge screw specimen holders were used to fix the specimen into a universal testing machine (UTM) (Zwick-Roell GmbH & Co.KG, Ulm, Germany) with a 40 mm grip-to-grip distance and the spray pattern centered. While clamping the specimen into the specimen holders, the UTM was set to constant force regulation with a set value of 5 N to compensate for stresses caused by the clamping procedure. For stress-strain measurement, a constant test speed of 0.4 mm/min was applied until a load of 500 N was reached. This equals a stress of 5 N/mm², which is below the 7–10 N/mm² of tensile strength in radial direction reported in the literature. The DIC system "*ARAMIS adjustable base 12 MP*" from GOM GmbH (Braunschweig, Germany) was electronically connected and synchronized with the UTM and measured the position of approximately 9000 individual points on the spray-painted area with 2 Hz. From the surface positions, an average normal uniaxial strain in the radial direction (ε) was calculated using the DIC's software "*Aramis Professional 20*". For uniaxial stress (σ) calculation, the specimens' dimensions (width and thickness) taken before the impregnation treatment were used. Poisson's ratio was neglected in the evaluation. The measured stress and strain in the elastic region were used to calculate Young's modulus $E = \Delta\sigma/\Delta\varepsilon$ *via* ordinary least squares regression.
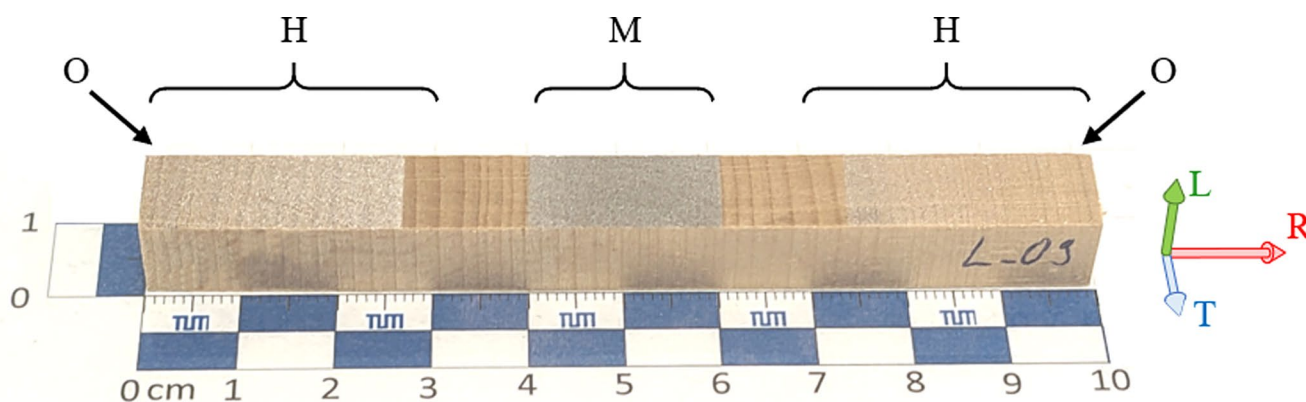
Fig. 1 Beech wood specimen used for Young's modulus measurements in the tensile mode and in the radial (R) direction. A random black-and-white dot spray pattern for strain measurement (M) by a digital image correlation (DIC) system is on the RT-plane. For the area to always be 20 mm long and centered, the adjacent wood was covered with masking tape before painting. Afterwards, the tape was removed, exposing the natural wood below and regions with some overspray (O) on both ends of the specimen. For testing, 30 mm of each end of the specimen was placed in the holder (H) of the universal testing machine

Fig. 2 Young's modulus from tensile testing in the radial direction ($n = 30$) of beech wood without any treatment, impregnated in deionized water, the HMR's reactive B-stage, a 50% solution of PEG-600 (PEG), and a 50% solution of polysorbate 20 (PS20). Note: Box = 25–75 percentile, line = median, star = average, whiskers = 1.5 IQR, cross = outlier. The dotted line gives the average of the water-treated group as a reference. Different letters show significant differences ($p < 0.05$) based on two-sample t-test analysis
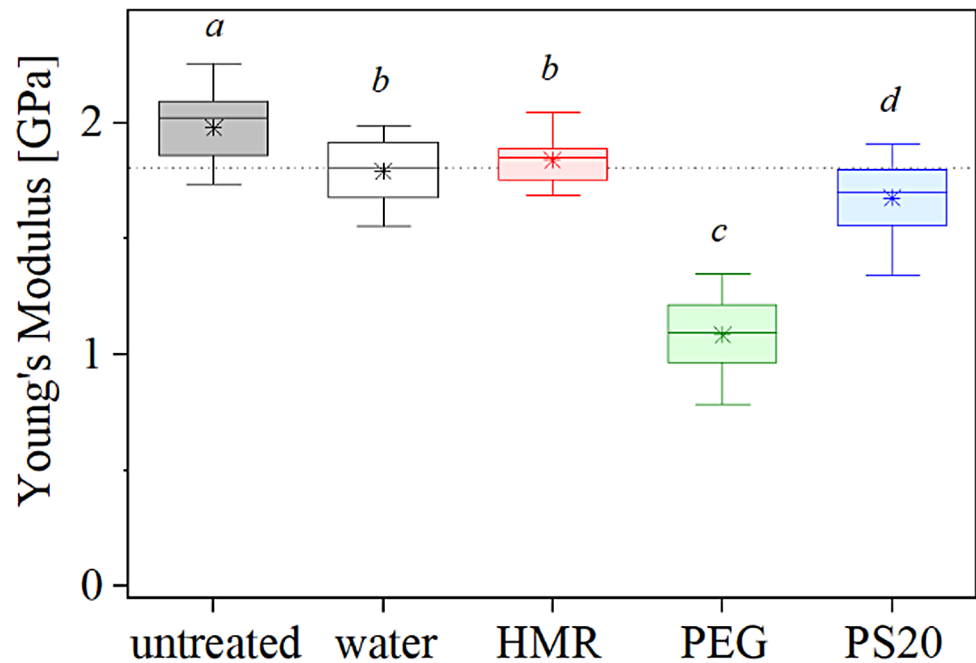
3 Results and discussion

The average mass increase of the specimens caused by the impregnation agents was 0%, 6%, 44%, and 29% after the water, HMR, PEG, and PS20 treatment, respectively. The average E in the radial direction of the untreated beech wood was 1.98 ± 0.14 GPa (average ± standard deviation), while it was 1.79 ± 0.13 GPa after the water treatment. With the HMR primer, the PEG-based primer, and the PS20-based primer, E was 1.84 ± 0.09 GPa, 1.09 ± 0.14 GPa, and 1.68 ± 0.16 GPa on average, respectively (Fig. 2).

The average E of the HMR-treated specimens compared to the untreated beech wood was reduced by 7%. In previous studies, Sun et al. (2005) compared stress relaxation under static strain, which indicated an increase in E by HMR-priming. By dynamic mechanical thermal analysis (DMTA), Son et al. (2005) did not observe a significant impact of the HMR primer on E. However, the authors reported a reduced glass transition temperature (T_g) for lignin using dynamic scanning calorimetry (DSC), which indicated a plasticization effect of the amorphous domains. These different characterizations were likely based on differences in the test set-up, which included the measurement techniques, as well as the reaction time and exposure method of the reactive HMR primer. Based on these differences, simultaneous effects could occur in different locations of the wood structure, which might superimpose each other. The reduced T_g, caused by small quantities of the HMR plasticization of the lignin domains, which is predominantly found in the middle lamella and primary cell wall, could be counteracted by reinforcing effects of the HMR within the secondary cell wall and on the lumen surface. While many plasticizers are liquid, solid plasticizers are also used for technical polymers, e.g., based on sulfonamides (CAS-number 3622-84-2). Therefore, a plasticizing effect of HMR molecules inside the amorphous wood polymers should principally not be omitted. However, the opposite effect, a stiffening, can be expected when the reactive substances in the HMR crosslink and form new, supportive structures in and/or around the cell wall. Even though with a 6% mass increase, the quantity of active chemicals was lower compared to the PEG and PS20, the used concentration of the HMR performed best in a tensile shear comparison by Böger et al. (2024). Overall, it is not possible to conclude if the observed 7% reduction is due to the effect of the primer's water, due to multiple effects of the HMR (partially) superimposing each other, or due to a low amount of active material in the cell walls. This is to be further investigated using other analytic techniques.

Here, with PEG (600 g/mol), a 45% reduction of the wood's average E was observed. This decrease is in line with a 15% reduction in bending stiffness reported by Stamm (1959), who tested Sitka spruce sticks impregnated with PEG (1000 g/mol), reaching a mass increase of 45.5% - similar to the 44% obtained in this study with beech. This deviation in the amount of stiffness reduction can primarily be explained by the different crystallization behavior of PEG 1000 g/mol with respect to PEG 600 g/mol. According to Paberit et al. (2020), the melting temperature and the corresponding enthalpy value are 40 °C and 152 kJ/mol, and 20 °C and 72 kJ/g for PEG 1000 g/mol and 600 g/mol, respectively. This clearly indicates that PEG-1000 is more prone to crystallize upon drying. Therefore, it is less likely

to be deposited in the amorphous wood polymer domains, thus remaining mainly in the lumina.

By the PS20 treatment, E was reduced by 16%. Bockel et al. (2020) used nanoindentation to evaluate the influence of PS20 on E and did not find clear differences between the treated and untreated wood. Due to some structural similarities to PEG, one could have expected a similar reduction in E. The smaller difference in E and the lower uptake during the impregnation could result from its higher molar mass (1228 g/mol) and amphiphilic nature. A statistical significance of the observed effect by PS20 compared to the water reference was confirmed by a two-sample t-test analysis ($\alpha = 0.05$). Prior to the test, it was confirmed that the requirements for the test were fulfilled by having two normally distributed, independent groups with similar variances according to the Levene-test (p: $0.237 > 0.005$).

4 Conclusion

The chosen method of measuring E in the R-direction provides reliable data on the influence of the primers' active chemicals on the mechanical properties of the treated wood.

With the PEG-600 treatment E reduced significantly– from 2.0 GPa to 1.1 GPa. The local plasticization leads to a modified stress distribution in the region of the bondline and, thereby, possibly improving the overall bond performance. With the waterborne PS20, a smaller but statistically still relevant effect– from 2.0 GPa to 1.7 GPa– is observed. However, this rather moderate plasticization appears too small to be the predominant mode of function of the PS20-based primer. Finally, with the HMR primer, a significant impact on E was observed– from 2.0 GPa to 1.8 GPa, but similar to the water-treated wood value of 1.8 GPa. Further research is necessary to identify whether this is due to the absence of a plasticization effect, too few active chemicals in the cell walls, or the superposition of opposing effects.

Data availability No datasets were generated or analysed during the current study.

Declarations

Competing interests The authors declare no competing interests.

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