Distinctive Impact of Processing Techniques on Bonding Surfaces of Acetylated and Heat-Treated Beech Wood and Its Relation to Bonding Strength*

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Abstract

In this study, the tensile shear strength of untreated, acetylated, and heat-treated beech (*Fagus sylvatica* L.) wood joints was investigated as a function of different surficial processing techniques. It was hypothesized that differentiating patterns of surface texture are induced by specific processing techniques directly affecting the bonding performance of adhered assemblies. Surface processing was implemented either by peripheral planing with sharp and dull knives, or by sanding (P100). Process-dependent surface textures were visualized by scanning electron microscopy and a digital light microscope was applied to display the structural integrity of surficial wood tissues. In dependence on wood modification techniques, process-related patterns of surface texture were observed. Laser scanning data of surface morphology was used to derive area-related functional roughness parameters defining complex surface textures quantitatively. For tensile shear testing, lamellae were bonded either with a two-component melamine-urea-formaldehyde adhesive or with a one-component moisture-curing polyurethane adhesive. Single lap-joint specimens were prepared following EN 302-1:2013 by the Deutsches Institut für Normung considering a material-adapted specimen geometry. Bonding strength was evaluated with respect to differentiating regimes of moisture. Specific dependences of modified beech wood properties on surface morphologies subsequent to surface processing and, therewith, on the associated bonding performance could be verified.

As a result, universal relationships between bonding performance and surface processing technique could not be identified. Thus, individual studies of bonding performances in dependence on adherend- and processing-related surface textures are inevitable.

Wood modification systems are process technologies with increasing market share (Hill 2011). Various process technologies in combination with varying species are implemented to produce modified wood, which is mainly used for

outdoor applications such as claddings or deckings. Recently, the use of bonded modification products for structural purposes as well as for nonload-bearing applications such as window scantlings increasingly came into focus (Bongers et al. 2016).

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Bonding of Modified Wood

The many positive effects to the modified wood substrate, such as biological durability, are accompanied by alterations of material properties that directly influence the bonding properties (Frihart et al. 2004). Various studies report that modified wood exhibits inferior bonding properties depending on chemical agents and process conditions, especially in combination with water-based adhesive systems (Jones and Hill 2007, Ntalos et al. 2008). However, another study proves the bonding performance of modified wood to be as good as that of untreated wood, sometimes even better (Hunt et al. 2007). To clarify this contradiction, further analyses of potential impact factors, either being relevant in the bonding process or during service life, have to be analyzed individually in dependence on wood characteristics, modification technique, adhesive system, and testing methodology.

During the bonding process, adhesive wetting, flow, and penetration are essential processing characteristics that determine the bonding performance (Kamke and Lee 2007). Regarding modified wood, bonding can be limited because of lower surface polarities and a reduced capillary uptake, particularly for water-based adhesive systems (Brandon et al. 2005, Hunt et al. 2007, Bryne and Wålinder 2010, Bastani et al. 2015). In modified wood, hydroxyl functionalities are inaccessible or blocked, which restricts the formation of interactions between adhesive polymers and cell wall components (Habenicht 2008, Rowell 2014). Moreover, adhesive curing is influenced by changes in acidity or moisture-related properties (Ormstad 2007, Kariz et al. 2013). Kägi et al. (2006) observed significant losses of bonding strength due to a lower equilibrium moisture content (MC) of the modified wood, which hindered the curing of one-component polyurethane (PUR) adhesives. Numerous studies have demonstrated that adhesive curing is also affected by extractives, which are modified and relocated toward the surface as part of the modification and drying process (Hse and Kuo 1988, Nuopponen et al. 2003, Sernek et al. 2008). Furthermore, as potential factors for bond durability, lower dimensional changes of modified wood during water sorption limit the movement of the adherend and, thus, promote homogenous stress distributions (Frihart et al. 2004, Hofferber et al. 2006, Hunt et al. 2007).

Bonding and Surface Morphology

Wood bonding is the macroscopic phenomenon of complex interactions between wood surfaces and adhesive systems. In the 1950s, bonding performance of untreated wood joints was not supposed to be associated with surface roughness (Marian et al. 1958). Nowadays, it is commonly accepted that surface texture influences bonding characteristics diversely (Stehr and Johansson 2000, Habenicht 2008).

The bonding surface is primarily formed through mechanical processing techniques such as planing or sanding (River et al. 1991). What these technologies have in common is that cutting edges of rakes compress the wood until the maximum strength is exceeded and wood chips are disintegrated (Gottlöber 2014a). Therewith, not only the outermost cells but also the entire wood matrix close to the surface can be influenced, which forms the mechanically influenced (weak) boundary layer of adhesive joints.

Roughness, the accuracy of fit, and the structural integrity of the surface with the corresponding boundary layer are essential for the overall bonding performance (River et al. 1991, Stehr and Johansson 2000).

The manifestation of bonding surfaces and boundary layers, respectively, depends on various wood-related properties, e.g., density, MC, and grain orientation (Murmanis et al. 1983, 1986). Additionally, the type and the associated parameters of mechanical processing techniques, e.g., tool sharpness and rake angle, directly determine the surface morphology (Kollmann 1955, Sinn et al. 2009). The most established planing technique is peripheral planing, by which the wood is machined by cylindrical tools rotating parallel to the processed surface (Grüll et al. 2016). The length and the depth of the plane knocks can be used for quality control (Kollmann 1955). Sharp planing knives produce smooth surfaces and undamaged boundary layers (Jokerst and Stewart 1976), whereas dull planing knives initialize fracture and deformations of cell walls, deteriorating the bonding performance (Seltman 1995, Bustos et al. 2010, Kläusler et al. 2014, Knorz et al. 2014). Abrasive planing or sanding of surfaces is based on multiple cutting edges with tool geometries, which can only be approximated statistically. Although negative rake angles prohibit presplitting, high normal forces intensify cell wall fracture as well as cell wall deformation during mechanical processing. Torn-out fibers increase the roughness of surfaces (Murmanis et al. 1986, Hernández and Cool 2008a, Cool and Hernández 2011). Depending on grit sizes, varying bonding performances were reported (Jokerst and Stewart 1976, Kläusler et al. 2014). However, no information about the impact of surface texture of modified wood on its associated bonding properties is available.

Scope of the Study

The present study relates the interaction between wood modification and mechanical surface processing techniques to the joint strength of bonded assemblies. Because physical and elastomechanical properties of the adherend are changed by wood modification, it is hypothesized that surface processing causes differentiating surface textures. Hence, the associated bonding performance of modified assemblies should be influenced as well.

As a first step, adherend surfaces of untreated, acetylated, or heat-treated beech wood are evaluated after they have been either planed, using sharp or dull knives, or sanded. The surface morphology and its corresponding cross sections of the bulk material are visualized microscopically. Moreover, surface and bulk characteristics are related to functional roughness parameters, which are calculated on the basis of laser scanning measurements.

As a second step, a two-component melamine-ureaformaldehyde (MUF) and a one-component PUR adhesive are applied for bonding. Single lap-joint specimens with a material-adapted geometry are tested to quantify the tensile shear strength (TSS) in varying regimes of moisture.

Materials and Methods

Untreated and modified wood

The investigations were carried out using untreated as well as modified beech wood (Fagus sylvatica L.). Acetylated boards with a weight percent gain of approximately 20 percent (Accoya) were purchased from Accsys

Technologies, Arnhem, The Netherlands. Timura Holzmanufaktur GmbH, Suedharz–OT Rottleberode, Germany, thermally modified boards according to the industrial Vacu³ process (Hofmann et al. 2013). All boards were stored at 20°C and 65 percent relative humidity (RH) until equilibrium MC (EMC) was reached.

Surface processing

Immediately before bonding, one of three different surface-processing techniques was applied on boards. which were previously conditioned at 20°C and 65 percent RH. For peripheral planing, the thickness planer T45 (Otto Martin Maschinenbau GmbH & Co. KG, Ottobeuren, Germany) was used. The knife shaft held four knives. Following Kollmann (1955, formula 61), machine parameters were adjusted to realize plane knocks of about 0.25 mm. The cutting depth was limited to 1 mm. Sharp and dull planing knives were considered. For distinguishability, the degree of tool wear was measured optically with a threedimensional surface metrology system (MicroCADPlus; GFMesstechnik GmbH, Berlin, Germany) and it was analyzed on the basis of software (ODSCAD 6.3 D, including the cutting edge module SK 4.7; GFMesstechnik). Tool-wear parameters are displayed in Table 1. Both the higher radius of the cutting edge and the notable radius delta of the dull knives indicate the degree of welladvanced abrasion. Additionally, the asymmetry toward the clearance surface (K < 1) and the lower degree of chipping verify the difference of knife qualities, which were included in this study. Alternatively to peripheral planing, a wide-belt sanding machine (Duplex-1100; Kündig GmbH, Gotha, Germany) was used to process wood surfaces before bonding. The two-step machinecontrolled sanding process combined a preleveling (abrasive paper: P60) with a finishing step (abrasive paper: P100).

Adhesive bonding

For face gluing, two important structural adhesive families with opposite material properties are considered for this study. Either a two-component MUF (Türmerleim GmbH, Ludwigshafen am Rhein, Germany) mixed with a hardener using a ratio of 2:1, or a one-component moisture-curing PUR (Jowat AG, Detmold, Germany), both of type one (EN 15425:2008, EN 301:2013; Deutsches Institut für Normung [DIN] 2008, 2013a), were used. All bonding processes were conducted according to the manufacturers' recommendations. For PUR bonding, acetylated and heat-treated lamellar surfaces were wetted (approximately 25 g m⁻²) before adhesive application. Both adhesives were applied manually on one side with the MUF, amounting to about 440 g m⁻², and the PUR, amounting to about 250 g m⁻². In total, the individual open and closed assembly time

Table 1.—Tool-wear parameters of sharp and dull planing knives determined with a three-dimensional metrology system.^a

Knife quality	Radius of cutting edge (µm)	Radius delta (µm)	K-factor	Chipping (µm)
Sharp	5.8 ± 3.0	6.8 ± 4.4	1.2 ± 0.6	2.0 ± 1.4
Dull	12.5 ± 5.5	15.0 ± 3.6	0.7 ± 0.2	1.7 ± 0.6

^a Values are arithmetic means ± standard deviations.

took approximately 15 minutes. The adhered boards were pressed in an automatically controlled hydraulic press (Gottfried Joos Maschinenfabrik GmbH & Co. KG, Pfalzgrafenweiler, Germany) for 8 hours at room temperature with a specific pressure of 1 MPa. Subsequently, the bonded assemblies were stored at 20°C and 65 percent RH for 4 weeks.

Analytical methods

Surficial appearance.—The morphology of the processed surfaces was visualized by a scanning electron microscope (SEM; EVO LS15, Carl Zeiss Microscopy Ltd., Cambridge, UK). Small specimens were cut out of lamella sections (10 by 6 by 6 mm³, longitudinal [L] by radial [R] by tangential [T]; Fig. 1) and were coated with a thin layer of carbon. Under high vacuum conditions a backscattered detector was used to magnify the surface at different scales on the basis of software (SmartSEM, Version 5.07, Service Pack 4; Carl Zeiss Microscopy).

A digital light microscope (VHX-5000, version 1.6.1.0, objective: VH-Z100R; Keyence Corporation, Osaka, Japan) was utilized to display the cell tissue close to the processed surfaces on cross-sectional planes. Specimens were cut out of lamella sections (10 by 20 by 10 mm³, L by R by T; Fig. 1) and the surfaces of the cross sections were prepared by using a microtome (Model 31 A 30; Sartorius AG, Göttingen, Germany). With a magnification of ×500, inspection areas (0.61 by 0.46 mm²) were placed either on earlywood or on latewood sections. Coaxial lighting in combination with a reduction of reflection of approximately 50 percent were used.

Surface roughness.—Roughness parameters were quantified to describe surface texture subsequent to mechanical processing. As displayed in Figure 1, the actual surface of lamellae was scanned with a confocal laser scanning microscope (LSM; microscope: VK-X110, control unit: VK-X100; Keyence Corporation), which was equipped with a red laser (658 nm) and a light source. With a magnification of ×10, single measurements focused on inspection areas of 1,350 by 1,012 μ m². On each specimen, 130 inspection areas were scanned and merged into a single data set. The morphological data sets were analyzed in accordance with normative requirements (EN ISO 25178-3:2012, EN ISO 25178-1:2016; DIN 2012b, 2016) on the basis of LSM analysis software (VK-H1XAD; Keyence Corporation). For this purpose, measuring ranges (10,000 by 10,000 μm^2) were positioned randomized on the merged surface area including equal proportions of earlywood and latewood sections. Subsequently, the extracted surface was fitted with a nominal shape by applying an automated linear correction of plane inclination (SF surface, F operator). Long-scale components were removed from the SF surface by utilizing an L filter (double Gaussian filter) with a nesting index of 2 mm (EN ISO 16610-1:2015; DIN 2015). Microroughness was not extracted from the data. On the basis of normative definitions (EN ISO 25178-2:2012; DIN 2012a), functional parameters, which are derived from the areal material ratio curve, were calculated to describe the spatial surface texture. Values of Spk (reduced peak height), Sk (core roughness depth), and Svk (reduced valley depth) were determined to yield descriptive height information (Fig. 2). Additionally, the corresponding proportions of the surface area were quantified.

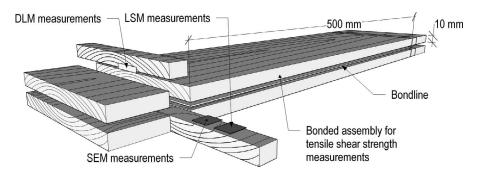


Figure 1.—Bonded assembly of lamellae and exemplary localization of surface measurements. DLM = digital light microscopy; LSM = laser scanning microscopy; SEM = scanning electron microscopy.

TSS of single lap joints.—TSS of MUF- and PUR-bonded specimens was defined in accordance with normative standards (EN 15425:2008, EN 302-1:2013; DIN 2008, 2013b). For each examined combination, 20 single lap-joint specimens were cut from bonded assemblies (Fig. 1). In contrast to normative requirements, lamella thickness of tensile shear test specimens was 10 mm instead of 5 mm (Fig. 3). Specimen geometry was adapted to prevent premature cohesive failure of the modified adherend material during testing. To consider aging effects, two conditioning sequences were applied as specified in standards (EN 15425:2008, EN 302-1:2013; DIN 2008, 2013b): A1 (standard state: 20°C and 65% RH) and A2 (wet state: submersion in water at room temperature for 4 days). Therefore, specimens were divided randomly into two groups before testing. In accordance with Hill (2006), the corresponding MC was determined gravimetrically (untreated samples: dry untreated wood basis; modified wood: dry modified wood basis). In a Zwick/Roell universal testing machine (10 kN load cell), specimens were stressed position controlled at an elongation rate of 1 mm min⁻¹ along the grain to failure. The TSS $(f_{v,t})$ was gained instrumented as a main result of the mechanical tests:

$$f_{v,t} = (F_{\text{max}}/A)$$

where $F_{\rm max}$ is the applied load at failure and A is the area of the shear plane. Moreover, the percent area of wood failure (wood failure percentage, WFP) on the bonding surface was determined optically.

Statistics

Numerical and graphical data analyses were implemented by R software (R Core Team 2017). A classical twofactorial analysis of variance (Tukey honest significant difference [HSD] test) was used to test homological groups for significant differences. Beforehand, normality was tested with a Shapiro-Wilk normality test (*P* value of 0.05) and homogeneity of variances was verified with a Levene test (*P* value of 0.05). For heteroscedastic distributions, an adapted

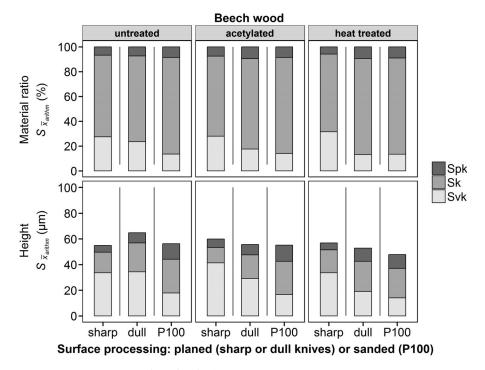


Figure 2.—Functional roughness parameters (Spk, Sk, Svk) of untreated, acetylated, and heat-treated beech surfaces subsequent to surface processing (sharp and dull planing, abrasive paper P100) measured by laser-scanning microscopy.

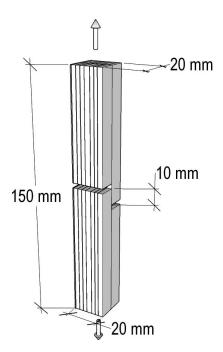


Figure 3.—Tensile shear test specimen based on EN 301-2:2013 (Deutsches Institut für Normung 2013a) considering a material-adapted specimen geometry.

Tukey HSD test was applied. In case of rejecting normality, the nonparametric Mann-Whitney U test was implemented.

Results and Discussion

Surface morphology

Surface planing with sharp knives.—Planing of untreated beech wood surfaces utilizing sharp knives opened the vessel and fibers, exposing lumens intensively (Fig. 4, 1a and 1c). They were largely free of cell fragments. In accordance with previous studies, mechanical disturbance of

surficial cell tissue was marginal (Knorz et al. 2014, Grüll et al. 2016). Evaluation of functional roughness parameters revealed that the mechanically exposed lumens are deep and account for approximately one-third of the total surface area (Fig. 2). Comparable surfaces were described to be plateaulike (de Moura et al. 2010). Only a few fractured elements of the cell wall could be detected.

Similar surface textures were observed on both acetylated and heat-treated beech wood (Fig. 5, 1a and 1c, and Fig. 6, 1a–1c). Since larger vessel cells are associated predominantly with earlywood sections, the material ratio of the surface valley depth (Svk) correlated strongly with the amount of earlywood or latewood tissue within the measurement ranges (Fig. 2). All sharply planed surfaces seemed to be characterized by a heterogeneous morphology because the height and the proportion of core roughness (Sk) is limited.

Surface planing with dull knives.—Deteriorated surface qualities are supposed to be induced by advanced tool wear (Gottlöber 2014b). Cutting edges of dull knives are characterized by varying geometries, which lead to irregular plane knock patterns (Kollmann 1955).

The examined cross-sections of untreated beech wood illustrated a wavy surface morphology (Fig. 4, 2b and 2c). Furthermore, surfaces exhibited deep valleys subsequent to surface processing (Fig. 4, 2a). The proportion of the total valley area, though, was decreased (Fig. 2). Lumens of cutopen vessels were partly filled with fractured cell walls and loose fibers. Hse (1968) and Follrich et al. (2010) confirmed that cell fracture and tissue deformation is more assigned to earlywood sections. Also in this study, cell wall fracture was especially more pronounced in earlywood tissue. As a result, both the height and the area of core roughness (Sk) and peaks (fuzziness, Spk) were increased. The observed wavy surface morphology was a result of tissue fracture, because dull planing knives cause higher cutting forces, which intensively stress the wood matrix, initializing failure (Singh et al. 2002).

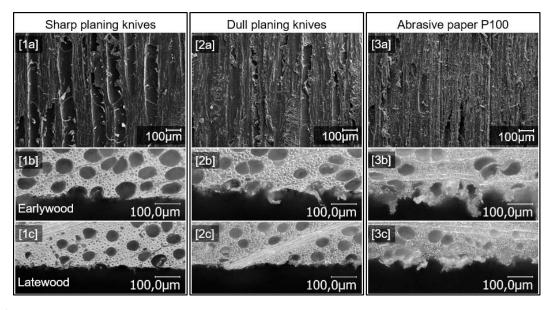


Figure 4.—Surfaces and cross sections of untreated beech wood in dependence on surface processing techniques: (1) sharp knives, (2) dull planing knives, (3) abrasive paper P100. (a) Scanning electron microscopy measurements, (b) digital light microscopy (DLM) measurements of earlywood, and (c) DLM measurements of latewood.

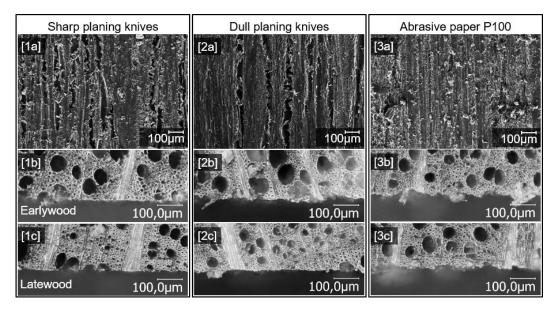


Figure 5.—Surfaces and cross sections of acetylated beech wood in dependence on surface processing techniques: (1) sharp knives, (2) dull planing knives, (3) abrasive paper P100. (a) Scanning electron microscopy measurements, (b) digital light microscopy (DLM) measurements of earlywood, and (c) DLM measurements of latewood.

In contrast, surface texture and surficial cell tissue of acetylated and heat-treated beech wood differentiated strongly from untreated beech wood after surface processing with dull planing knives (Fig. 5, 2 and Fig. 6, 2). Fuzziness and core roughness increased considerably, whereas the valley height and the proportional area decreased simultaneously (Fig. 2). Cell wall fragments of variable sizes were deposited within cut-open lumens. Induced by high modification temperatures and low equilibrium MCs, brittle tissue failure is more pronounced on modified wood (Rowell et al. 2009). As a result, high cutting forces led to intensive fracture of modified cell walls, which were spread over the surface. Thus, the influence of surface valleys on surface

morphology declined. Moreover, visualizations of crosssections depicted almost undisturbed surficial cell tissues (Fig. 5, 2b and 2c, and Fig. 6, 2b and 2c). Again, the lower EMC of modified wood in combination with grain orientation might be causal for the higher stiffness of the cell matrix, promoting sound tissue structures.

Sanding.—Abrasive planing or sanding of untreated beech wood surfaces produced structures that are influenced primarily by mechanical processing rather than by the anatomy of native cells (Kollmann 1955, Marra 1992, Cool and Hernández 2011). Surface visualizations indicated channellike structures parallel to machining direction (Fig. 4, 3a). However, no definitely opened cell lumens were

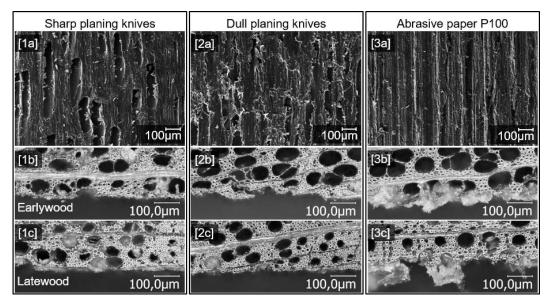


Figure 6.—Surfaces and cross sections of heat-treated beech wood in dependence on surface processing techniques: (1) sharp knives, (2) dull planing knives, (3) abrasive paper P100. (a) Scanning electron microscopy measurements, (b) digital light microscopy (DLM) measurements of earlywood, and (c) DLM measurements of latewood.

observable anymore. Roughness parameters confirmed the distinct reduction of valley depth and the proportional area of valleys (Fig. 2). Accumulated sander dust might have been causative, which could have been plasticized and solidified by frictional energy during sanding. Further cell wall material of small sizes was deposited on the entire surface area. Thus, core roughness and fuzziness increased remarkably. Cross-sectional planes displayed intensive disintegration of wooden tissue primarily within earlywood sections of untreated beech wood (Fig. 4, 3b and 3c).

Similar surface textures could be observed on acetylated and heat-treated wood (Fig. 5, 3a and Fig. 6, 3a). Depth and proportions of valleys were decreased, whereas core roughness and peak height increased (Fig. 2). Hence, surface areas were texturally homogenized. As opposed to untreated wood, only minor distortions of the surficial cell matrix were visible on cross sections of modified wood. The stabilization of wood tissue could also be attributed to lower EMCs and partly to higher densities of modified wood.

Evaluation of surface analysis.—Surface morphology is a major impact factor influencing relevant adhesion mechanisms (Habenicht 2008). Therefore, sensitive testing methodologies are necessary that deliver realistic numerical data describing surface textures. By LSM, the genuine wood surface is digitalized nondestructively, generating an extracted surface that is comparable with a surface envelope. Although individual structures, e.g., steep sides of cell walls, provoke imprecise laser scanning measurements because of the limited reflection of beams (Arnold et al. 1992), laser-based measurements of surface morphology represent a reproducible tool to differentiate process-induced physical surface characteristics (Funck et al. 1993).

Common roughness parameters, e.g., the arithmetic mean height (Ra or Sa, respectively), provide limited information about the actual surface texture. Conversely, functional roughness parameters represent expedient indicators to summarize the distribution of height, including information about relative proportions. In accordance with previous investigations, the results of the present study indicate the reduced valley depth (Svk) to be strongly correlated with anatomical features of wood, which can be superimposed by process-related cell wall fragments (Fujiwara et al. 2005, Gurau et al. 2005). Furthermore, the core roughness (Sk) is directly associated with surface processing and the reduced peak height (Spk) quantifies fuzziness. Microscopic visualizations of surfaces can successfully be utilized to verify functional roughness parameters. Cross-sectional planes of surficial cell tissue enable important supplementary information about the integrity of the boundary layer within the bonded assembly because mechanically weak boundary layers immediately limit the performance of the bonded assembly (Stehr and Johansson 2000).

TSS of bonded specimens

Testing methodology.—Generally, quality control of adhesively bonded joints can be based on criteria like TSS and WFP, as intended with the corresponding standards (EN 15425:2008, EN 301:2013; DIN 2008, 2013a). Regarding modified wood, both criteria have to be taken into account simultaneously to evaluate bonding performance adequately (Brandon et al. 2005). However, testing procedures and normative thresholds are basically valid for testing the bonding performance of untreated beech wood only, because changes in strength characteristics of alternative

or modified adherends are not considered. Hence, Konnerth et al. (2016) related bonding strength to specific material strength, for example. In this study, the geometry of specimens was adapted (Fig. 3). Therewith, the number of valid specimens that did not exhibit premature cohesive failure within the bulk wood during applied stresses was increased compared with the standard geometry as recommended by EN 302-1:2013 (DIN 2013b and unpublished results). As a result of thicker specimen geometries, TSS increases because of decreasing occurrences of local stress peaks (Crocombe and Ashcroft 2008, Stoeckel et al. 2013). Consequently, TSS presented in this study cannot be compared directly with previous publications, in which standard geometries were applied. Moreover, the differentiating MC (Table 2), density (Table 2) and, hence, corresponding elastomechanical properties between untreated and modified wood impede relative classifications. Therefore, the bonding performance was analyzed separately depending on wood treatment.

Bonding performance of untreated beech wood.—Regardless of surface-processing technique, MUF-bonded joints exhibited similar bonding strength of approximately 12 MPa and 100 percent WFP (Fig. 7). With up to 15 MPa, the bonding performance of PUR-bonded joints was slightly higher. Only sanded surfaces displayed lower PUR-bonding strength. Additionally, deviation of WFP was higher for sharply planed PUR bonds. As expected, bonding performance of both adhesive systems declined when exposed to wet testing conditions (Kläusler et al. 2014, Konnerth et al. 2016).

In the present study, the sharply planed surfaces were characterized by many cut-open lumens of vessel cells. Thus, the observed valley depth and valley proportion (Fig. 2; Svk) might have promoted adhesive penetration into the wood. Furthermore, only minor degrees of cell wall fragments were observed, which could block adhesive flow and penetration. Referring to the literature, higher bonding strength is related to sufficient adhesive penetration into the wood, because load transfer is optimized and mechanical entanglement contributes to adhesion forces (Kamke and Lee 2007, Cool and Hernández 2011). Consequently, high amounts of adhesive with sufficient cohesive strength are necessary. Moreover, bonding strength was benefiting from sound boundary layers (de Moura et al. 2010), which was indicated by intensive WFP within the bulk wood (Singh et al. 2002). Deteriorating bonding performances, especially of PUR joints in the wet state, could be attributed to damaged wood tissues in combination with adhesion failures (Kläusler et al. 2014). In comparison with the MUF, the viscosity of the utilized PUR adhesive system was higher.

Table 2.—Material properties of untreated, acetylated, and heat-treated beech wood.^a

	Density (g/cm ³)	MC (%)	
Modification		Standard	Wet
Untreated	0.70 ± 0.01	12.3 ± 0.4	72.3 ± 2.2
Acetylated	0.74 ± 0.03	5.5 ± 0.4	33.9 ± 2.2
Heat treated	0.72 ± 0.01	4.5 ± 0.1	43.0 ± 3.9

^a Values are arithmetic means ± standard deviations. Density was calculated with a moisture content (MC) at 20°C and 65 percent relative humidity (standard). Displayed MCs comply with the state of specimens at the time of tensile shear testing.

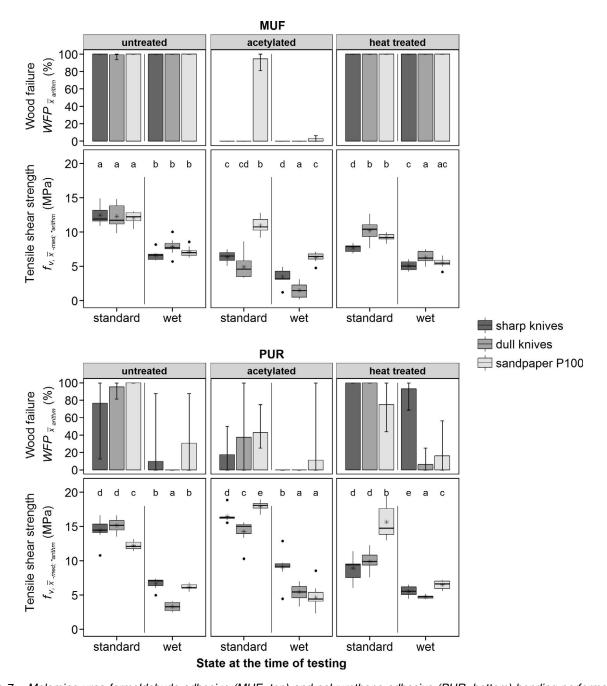


Figure 7.—Melamine-urea-formaldehyde adhesive (MUF, top) and polyurethane adhesive (PUR, bottom) bonding performance of untreated, acetylated, and heat-treated beech wood joints in relation to surface processing techniques (sharp and dull planing, abrasive paper P100) and the specimens' state at the time of testing (standard: storage at 20°C and 65% relative humidity [RH]; wet: 4 days of water submersion). Box and whisker plots indicate median (line), arithmetic mean (asterisk), 25 and 75 percent percentiles, minimum or maximum values within the 1.5× interquartile range; dots represent outliers. Bar plots display the arithmetic mean and the minimum and maximum values. Differentiating letters symbolize significant differences.

As a result, the adhesive's capacity to compensate substrate damage might have been limited.

Bonding performance of modified beech wood.—Both acetylation and heat treatment influence the bonding performance diversely (Sernek et al. 2008, Bongers et al. 2016). The TSS of acetylated MUF bonds was low (Fig. 7). Only sanded surfaces exhibited higher bonding strength accompanied with higher WFP. In contrast, high bonding strength of approximately 15 MPa was documented for PUR joints of acetylated wood. Again, the bonding strength of

both adhesive systems was reduced remarkably subsequent to water storage. Compared with sharply planed surfaces, MUF- and PUR-bonded joints of acetylated wood exhibited lower TSS when dull planing knives were applied. No wood failure was observed on the shear plane of MUF joints. As a result, adhesion of dully planed acetylated surfaces might have been deteriorated by loose cell wall fragments, which could not transfer any load between the adjacent adherends. In contrast, wood failure was verifiable on the shear plane of PUR joints. Besides chemical surface properties, soundness

and elastomechanical properties of the boundary layer are of great importance (Stehr and Östlund 2000, Singh et al. 2002). TSS of acetylated joints was clearly higher for PUR bonds, and surficial tissue failure might be causal to reductions of TSS of dully planed surfaces. However, the highest TSSs and WFPs were documented on sanded surfaces of acetylated beech wood, which is in line with previous studies (Hunt et al. 2007). It is assumed that the wetting and curing of water-based MUF adhesives is diminished because of hydrophobic surfaces of modified wood. Sander dust, as well as the intensified fuzziness, might accelerate adhesive gelling by water absorption. Accompanied by acidic surface conditions, adhesive spreading, wetting, penetration, and curing might have been optimized on acetylated wood by abrasive paper processing. It has been discussed before that intensified roughness might be associated with enhanced wetting of water-based systems (Hernández and Cool 2008b). Eventually, the potential adhesion area is increased with increasing roughness, which is supposed to positively influence the bonding strength (de Moura et al. 2010). Moreover, the area of adhesion might partly explain the very high TSS of PUR joints of acetylated wood subsequent to sanding (standard state). Similarly important seems to be the integrity of the boundary layer (Stehr and Johansson 2000, Stehr and Östlund 2000) and the flexibility of the PUR bond line. Instead of comprehensive load transfer from one adherend into the bulk of the other, PUR bond lines are elongated, compensating the development of stress peaks (Konnerth et al. 2006, Stoeckel et al. 2013).

TSS of MUF-bonded joints of heat-treated beech wood was increased when dull planing knives or abrasive paper, respectively, were utilized (Fig. 7). Although heat treatment directly influences the curing of adhesives (Kariz et al. 2013), no cohesive failure within the bond line could be observed. Instead, WFP was maximal in all cases. Thus, MUF-TSS predominantly was determined by the cohesive strength of the boundary layer. Roughness measurements have indicated strong reductions of valley depth as well as proportional valley area (Fig. 2). Simultaneously, core roughness and fuzziness were increased by both dull planing and sanding. Therefore, MUF bond line formation and adhesive penetration might have been homogenized, encapsulating cell wall fragments and loose fibers. Applied load was transferred deeper into the fairly sound boundary layer, increasing the MUF-TSS of the bonded assembly. However, in comparison with dull planing knives, excessive deposition of sander dust on the surface might inhibit adhesive penetration, limiting the MUF-TSS. In contrast, very high TSS was verified for PUR-bonded joints of heattreated and sanded beech wood, which also might be associated with optimized flow and penetration of the highly viscous PUR adhesive. Furthermore, the increasing core roughness and fuzziness might contribute to adhesion, because the adhesion area was increased strongly.

Evaluation and future work

The results support the hypothesis that surface processing directly influences the bonding performance of modified wood joints. The individual bonding performance, though, has to be related to specific adherend-dependent surface textures rather than be associated with general processing techniques, which is in line with previous studies focusing on untreated wood (Jokerst and Stewart 1976, Kläusler et al.

2014, Ammann et al. 2016). Intra- and intermolecular fracture of cell walls and deformation of the cell matrix define elastomechanical properties of the bonded assembly decisively (Stehr and Johansson 2000; Gindl et al. 2004a, 2004b).

Detailed examinations of the impact of processing techniques on chemical surface properties were not considered for this study. It is assumed, though, that process-related chemical changes influence the bonding properties diversely.

Further research would be needed to confirm the impact of increased roughness on wetting properties. Investigations of adhesive penetration and bond line formation could be expedient to distinguish between relevant adhesion mechanisms. Additionally, certain bonding performances, e.g., the combination of MUF and acetylated wood subsequent to sanding, could be further improved by adapting mixing ratios or assembly times, as it was documented before to be relevant for untreated beech wood (Schmidt et al. 2010).

Conclusions

MUF- and PUR-bonded joints of untreated, acetylated, and heat-treated beech wood were prepared to evaluate the TSS considering moisture-induced influences. The bonding surface was either planed (sharp or dull planing knives) or sanded (abrasive paper P100). Process-dependent surface textures were visualized microscopically and area-related functional roughness parameters were calculated. On the basis of the results, the following conclusions might be drawn

Surface processing techniques defined the surface morphology distinctively in dependence on the wood modification system. Cell wall fragmentation increased with declining knife quality. Sanding leveled the impact of wood anatomy on surface morphology.

Regarding surface processing techniques applied on modified wood, area-related functional roughness parameters were sensible indicators sufficiently describing the complex texture of specific surfaces. Process-induced changes of valley depth and the proportional valley area could be assigned to cut-open lumens, which were observable predominately on sharply planed surfaces. Conversely, the core roughness and the fuzziness increased with the utilization of dull planing knives and abrasive paper processing, respectively.

Moreover, the surficial cell tissue of modified wood exhibits a pronounced structural integrity subsequent to processing.

In contrast to untreated wood, the bonding performance of acetylated and heat-treated wood joints varied strongly in relation to processing techniques. Further impact factors in the form of adhesive system and moisture regime could be verified. MUF-bonded joints of acetylated wood exhibited a significantly higher TSS in the case of sanded surfaces, whereas sanding of heat-treated surfaces decreased the bonding strength of MUF-bonded joints. The highest TSS was observed on PUR bonds of acetylated and heat-treated joints under limited moisture exposition.

In conclusion, bonding performance in relation to surface texture has to be studied individually depending on processing parameters and adherend substrate, rather than deriving findings from previous studies.

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