Fachgebiet Holztechnologie

Investigation of structurally bonded ash (*Fraxinus excelsior* L.) as influenced by adhesive type and moisture

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Abstract

The utilization of ash wood (*Fraxinus excelsior* L.) in bonded load-bearing products requires in-depth knowledge of bond performance. The aim of this work was to investigate the gluability of ash wood for load-bearing purposes. Particular emphasis was placed on the suitability of different adhesive types for bonding of ash, and the influence of moisture changes on the performance of bonded ash.

The moisture-related durability of ash assemblies that were bonded with different adhesive types was evaluated in delamination tests. This investigation showed that adhesive types which are regarded to be equally well suited for structural bonding of softwoods perform considerably different in combination with ash wood. Phenol-resorcinol-formaldehyde (PRF) proved to be the most appropriate adhesive type to produce durable structural bonds with ash. The closed assembly time was identified as key bonding parameter. Prolonged closed assembly times produced increasing glueline thicknesses and higher moisture-related durability of ash bonds.

In addition, the influence of the bonding surface on the performance of ash bonds was investigated. Bonding surfaces were prepared by means of planing, face milling and sanding and differed primarily in the level of fibrillation and the degree of cell damage. An increasing level of fibrillation had a positive effect on the moisture-related durability of ash bonds for all tested adhesive types. Optimization effects varied with surfaces and adhesive types, and very likely depended on adhesive elasticity and the way of strain distribution in the bond in case of moisture change.

In order to better understand moisture-related bond durability the deformation behavior of ash assemblies was investigated. Main focus was put on moisture-induced shear strains in bonded ash. It could be shown that deformations in and adjacent to the glueline are greatly influenced by adhesive elasticity. In addition, the ability of adhesives to penetrate into cell walls most likely affected the deformation behavior. The investigation also revealed that the thickness of melamine-urea-formaldehyde (MUF) gluelines has an effect on moisture-induced shear strains in bonded ash. The results indicate higher interfacial stress for thin gluelines and a buffering effect of thicker gluelines. This assumingly explains the lower moisture-related durability of ash bonds with thin gluelines as determined in delamination tests.

The knowledge gained in this work can serve as a basis for a technical approval of structurally bonded elements made with ash wood. The results can also be used for new adhesive developments for hardwood bonding, or contribute to modeling of adhesively bonded wood assemblies.

Zusammenfassung

Die Verwendung von Eschenholz (*Fraxinus excelsior* L.) in geklebten, tragenden Bauteilen setzt ein umfassendes Wissen um die Leistungsfähigkeit von Verklebungen voraus. Das Ziel der vorliegenden Arbeit war daher, die Verklebung von Eschenholz für die Verwendung in tragenden Bauteilen zu untersuchen. Hierbei wurde einerseits die Eignung verschiedener Klebstofftypen für Eschenverklebungen bewertet. Andererseits wurde der Einfluss von Holzfeuchteänderungen auf das Leistungsverhalten von Eschenverklebungen untersucht.

Die Dauerhaftigkeit bei Feuchteänderungen wurde für Eschenholzbauteile, die mit verschiedenen Klebstofftypen hergestellten wurden, in Delaminierungsprüfungen beurteilt. Diese Untersuchung machte deutlich, dass sich Klebstofftypen, welche für die Verklebung tragender Bauteile aus Nadelholz gleichermaßen gut geeignet sind, in Verbindung mit Eschenholz stark unterschiedlich verhalten. Es zeigte sich, dass sich mit Phenol-Resorcinol-Formaldehyd (PRF) Klebstoff am ehesten dauerhafte Eschenholzverklebungen erzielen lassen. Die geschlossene Wartezeit erwies sich hierbei als ein entscheidender Verklebungsparameter. Eine Verlängerung der geschlossenen Wartezeit führte zu dickeren Klebfugen sowie zu einer höheren Dauerhaftigkeit von Eschenholzverklebungen bei Feuchteänderungen.

Darüber hinaus wurde der Einfluss der zu verklebenden Oberflächen auf das Leistungsverhalten von Eschenholzverklebungen untersucht. Die Oberflächen wurden mittels Hobeln, Stirnplanfräsen und Schleifen hergestellt und unterschieden sich hauptsächlich hinsichtlich der an der Oberfläche vorhandenen gelösten Fasern sowie im Ausmaß der Zellschädigung. Das Vorhandensein von Fasern an der Verklebungsoberfläche bewirkte eine erhöhte Dauerhaftigkeit der Eschenholzverklebungen in Verbindung mit allen untersuchten Klebstoffen. Inwieweit sich Verbesserungen erzielen lassen, variiert mit der jeweiligen Oberflächen-Klebstoff-Kombination und hängt offensichtlich mit der Elastizität der Klebstoffe und dem Deformationsverhalten von Verklebungen bei Holzfeuchteänderungen zusammen.

Um ein besseres Verständnis zur Dauerhaftigkeit von Verklebungen zu erlangen, wurde das Formänderungsverhalten von Eschenholzverklebungen bei Holzfeuchteänderung untersucht. Hierbei wurden hauptsächlich auftretende Scherverformungen betrachtet. Es konnte gezeigt werden, dass Verformungen in der Klebfuge sowie in angrenzenden Bereichen stark von der Steifigkeit der Klebstoffe abhängen. Zudem beeinflusst die Fähigkeit mancher Klebstoffe, in die Zellwände des Holzes eindringen zu können, das Verformungsverhalten der geklebten Verbindung. Darüber hinaus ließ diese Untersuchung erkennen, dass verschiedene Dicken von Melamin-Harnstoff-Formaldehyd (MUF) Klebfugen unterschiedliche Scherverformungen in Eschenholzverklebungen zur Folge haben. Die Untersuchungsergebnisse deuten darauf hin, dass bei dünnen Klebfugen höhere Spannungen im Holz-Klebstoff-Interface auftreten als bei dicken Klebfugen. Darin wird ein Erklärungsansatz für die in Delaminierungsprüfungen ermittelte geringere Dauerhaftigkeit von Eschenverklebungen mit geringen Klebfugendicken gesehen.

Die in dieser Arbeit erzielten Ergebnisse können als Grundlage für eine bauaufsichtliche Zulassung geklebter, tragender Elemente aus Eschenholz dienen. Darüber hinaus können die Erkenntnisse bei der Neuentwicklung von Klebstoffen für die Laubholzverklebung förderlich sein, oder zur Modellierung von Verklebungen beitragen.

List of papers

This dissertation is based on investigations that were published in the following three original articles:

I. Knorz M, Schmidt M, Torno S, van de Kuilen J-W (2014) Structural bonding of ash (*Fraxinus excelsior* L.): resistance to delamination and performance in shearing tests. European Journal of Wood and Wood Products 72(3):297-309.

II. Knorz M, Neuhaeuser E, Torno S, van de Kuilen J-W (2015) Influence of surface preparation methods on moisture-related performance of structural hardwood-adhesive bonds. International Journal of Adhesion and Adhesives 57:40-48.

III. Knorz M, Niemz P, van de Kuilen J-W (2015) Measurement of moisture-related strain in bonded ash depending on adhesive type and glueline thickness. Holzforschung DOI: 10.1515/hf-2014-0324.

The above listed articles are attached in the appendix. Hereinafter, reference to the original articles is made using the terms ‚paper I', ‚paper II' and ‚paper III'.

Contents

1 Introduction

1.1 Potential of ash wood for use in structural products

The efficient use of resources is a key challenge of our time. One important renewable resource that is provided by our forests is wood. The application of wood as a building material offers a valuable opportunity for a sustainable material utilization. Recently, forestry surveys have reported significant changes in central European forests, the most important being an increasing availability of hardwoods (e.g., Brändli 2010; Bundesministerium für Ernährung und Landwirtschaft (BMEL) 2014). Whereas the use of softwoods as structural timber is well established in Europe, large amounts of hardwoods are only used for energy production (Krackler et al. 2010; Weimar and Seintsch 2012; Hübner 2013). The use of some hardwoods as structural timber would however account for significant benefits, for example for a considerable enhancement of the load-bearing capacity of building products.

The good material properties of ash wood (*Fraxinus excelsior* L.) have been known for a long time for which reason ash wood used to be applied for instance in aircraft and automobile construction. Because of its high fracture toughness ash is still frequently used in tool handles or sports equipment. A rather new field where the use of ash is considered to be beneficial is the application in structural building products. This is because ash in particular provides outstanding strength properties that significantly exceed the properties of softwoods. The strength and stiffness properties have been determined both on small and defect-free specimens (Baumann 1922; Anonymous 1939; Kollmann 1941, 1951; Kühne 1951; Schwenke 1956; Oliver-Villanueva 1993; Sell 1997; DIN 68364:2003; Bonoli et al. 2005; Wagenführ 2007; Clauß et al. 2014; Niemz 2014; Niemz et al. 2014) and on ash timber with larger dimensions (Quer 1997; Frühwald and Schickhofer 2004; Glos and Torno 2008; van de Kuilen and Torno 2014) taking strength grading as a basic requirement for the use of timber in structural applications into account.

The utilization of sawn timber is limited by log dimensions with regard to length or cross-sections. The use in adhesively bonded wood products, for example in glued laminated timber (glulam) makes it possible to overcome dimensional limitations of sawn timber and at the same time offers substantial advantages such as improved dimensional stability, and enhanced strength and stiffness. The potential of ash wood to improve the load-bearing capacity of glulam and joints in timber structures was already identified by Gehri (1983). However, scientific investigations that facilitate the utilization of ash in glulam are still missing. This in particular is the case for the examination of the gluability of ash. A preliminary study with a limited number of specimens confirmed the good material properties of ash glulam (Frühwald et al. 2003) but also showed a poor performance of the ash bonds in durability evaluations. First experiences with ash glulam in structural applications have been made in Switzerland for a few years due to more innovation-minded building regulations. However, the durability of ash bonds has not been examined in-depth before application which may lead to delaminations in the long term.

1.2 Research gaps

A safe utilization of ash wood in structural bonded products requires in-depth knowledge of bond performance. This applies both to mechanical stability and bond durability that need to be guaranteed during the service life of structures. While adhesive bonding of load-bearing softwood products is well established, there is lack of knowledge with regard to performance of bonded hardwoods. This for instance applies to several adhesive types that are established for structural softwood bonding but may perform differently with hardwoods. In particular, it needs to be investigated if specific bonding parameters are required to obtain efficient ash bonds. To close this knowledge gap, the suitability of different adhesive types for bonding of ash should be investigated by evaluating the performance of bonded ash and its dependence on bonding parameters.

Moisture-related bond durability is a fundamental prerequisite for structural bonds and therefore has to be examined with bonded ash. Bond durability is normally being evaluated in accelerated aging tests such as the delamination test according to EN 302-2 (2013) in Europe or ASTM D2559 (2012) in North America. This test method is of particular importance and well established to evaluate the suitability of adhesives for softwood glulam. It is the only method within relevant adhesive approval tests where stresses are generated by the wood itself as a result of water impregnation and subsequent rapid drying of test specimens. Therefore, physical and mechanical properties of the bonded wood species have major impact on the test outcome. Based on the high strength and stiffness values of ash wood, significantly higher stresses in the bonds can be anticipated when compared to softwood. As a consequence, the delamination test is assumed to be a highly critical test for bonded ash. Furthermore, the delamination test is of major importance in practical terms. When evaluating the suitability of an adhesive that is already approved for bonding of softwoods in Europe, the resistance to delamination is the only additional requirement that needs to be verified when this adhesive is intended to be used for bonding of hardwood species, i.e., in the present research with ash.

Even though the delamination test according to EN 302-2 (2013) is regarded a valuable assessment method for bond durability and it therefore is compulsory for adhesive approval tests in Europe, it does not allow for a clear prediction of the service life of a wood-adhesive bond when climate conditions of applications are unknown. For this reason, there is a general need for a better understanding of the moisture-related durability of wood-adhesive bonds. In particular, there is a lack of knowledge how the frequency of wood moisture changes and swelling and shrinkage processes in practice influence bond durability and how wood moisture changes can be related to results from accelerated aging tests. For a better predictability of the long-term behavior of wood-adhesive bonds, deformations and stresses that occur with changing wood moisture content (MC) both in the wood and in the glueline should be examined. As deformations and stresses vary with wood properties, consideration in this research should be given to the deformation behavior of bonded ash assemblies.

1.3 Objectives

The main objective of this thesis is to examine the gluability of ash timber for structural purposes. The research aims at an improved understanding of the moisture-related behavior and performance of ash bonds when bonded with different adhesive types[1]. The main goal is approached in three experimentally oriented investigations with individual research objectives which are addressed in three peer-reviewed papers:

Paper I – Structural bonding of ash (*Fraxinus excelsior* L.): resistance to delamination and performance in shearing tests

Objectives of this investigation are the following:

- Evaluation of the suitability of different adhesive types that are commercially available and established for structural bonding of softwoods (phenol-resorcinol-formaldehyde (PRF), melamine-urea-formaldehyde (MUF), one-component polyurethane (1C-PUR) and emulsion polymer isocyanates (EPI)) for structural bonding of ash. Both the short-term bond performance in shearing tests and the bond durability in delamination tests are to be examined.
- Determination of the influence of various closed assembly times as important bonding parameter on bond formation and performance.

Paper II – Influence of surface preparation methods on moisture-related bond performance of structural hardwood-adhesive bonds

Objectives of this investigation are the following:

- Examination of the influence of the surface preparation methods peripheral planing, sanding and face milling on the properties of the ash bonding surfaces.
- Evaluation of bond performance of different adhesives (PRF, MUF, 1C-PUR and EPI) as a function of surface preparation methods with emphasis being placed on tensile shear tests after exposure to moisture change and resistance to delamination.

Paper III – Measurement of moisture-related strain in bonded ash depending on adhesive type and glueline thickness

Objectives of this investigation are the following:

- Examination of the moisture-induced deformation behavior of bonded ash with respect to adhesive types (PRF, MUF, 1C-PUR and EPI).
- Analysis of strain distribution due to moisture change in MUF-bonded ash as a function of glueline thickness in an appropriate range between 0.01 mm and 0.2 mm.
- Development of a better comprehension about the distribution of moisture-induced strain in dependence of adhesive penetration.

[1] In this thesis, the term ‘adhesive type’ is used with respect to the chemical system (PRF, MUF, 1C-PUR or EPI). It does not refer to the classification into adhesive type I or II according to adhesive classification standards EN 301 (2013), EN 15425 (2008) or EN 16254 (2013).

2 Background

2.1 Ash wood (*Fraxinus excelsior* L.)

Wood can be characterized as a three-dimensional network of conducting, storing and strengthening cells. The composition and arrangement of the cell types greatly varies for softwood and hardwood, and moreover for individual wood species. For a complete anatomical characterization of wood, the transverse, radial and tangential plane have to be considered. In Figure 1, the three main planes are illustrated for ash (*Fraxinus excelsior* L.).

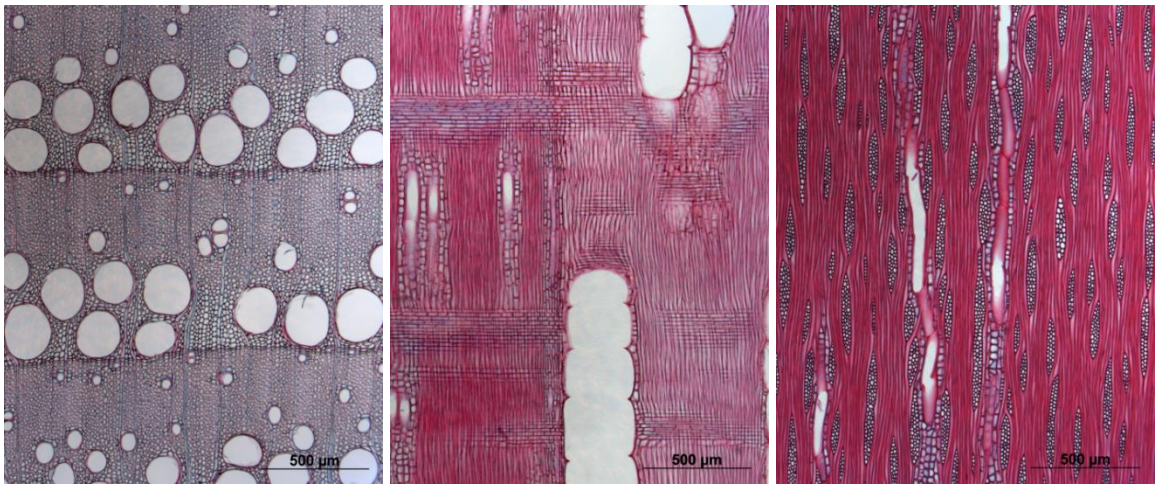


Figure 1 Microscopic images of ash wood in transverse (left), radial (center) and tangential section (right)

Ash is a ring-porous hardwood species showing circularly arranged earlywood (EW) vessels with diameters up to 350 µm (Anonymous 1939). Latewood (LW) vessels are significantly smaller with an average diameter of 50 µm (Grosser 2003) and a diffuse distribution. The vessels are connected to neighboring cells via many small, up to 5 µm wide bordered pits (Kollmann 1941). Vessel lumens in ash are frequently blocked by tylosis. The strengthening tissue of ash consists of fibers with small lumens. Fiber cell walls are thin in EW and up to 10 µm thick in LW. Pits in fiber walls are less numerous and smaller (< 3 µm) than in vessels (Kollmann 1941). Storing cells in ash can be found as longitudinal parenchyma surrounding LW vessels and in terms of frequent, small and homogeneous wood rays.

A comprehensive description of the structure of ash wood can be found in Kollmann (1941). The concise anatomical characterization of ash wood given in Table 1 is based on data from Wagenführ (2007).

Table 1 Anatomical characterization of ash (Wagenführ 2007)

		Measure and dimension (µm)		Percentage (%)
Vessels		Lumen diameter:	60…120…350 (EW); 15…50…130 (LW)	4.7…12.1…21.3
Fibers		Lumen diameter:	5.3…15.0…24.2	50.5…62.4…72.4
		Cell wall thickness[a)]:	1.5…3.6…5.1	
		Length:	150…1600	
Paren-chyma	Ray	Height:	90…290..500 (≈10 cells)	13.9…14.9…16
		Width:	23..27..60 (1..3..5 cells)	
	Longitu-dinal			5.7…10.6…15.1

a) Values correspond to two cell walls, i.e., a cell wall thickness value of 3.6 µm comprises two cell walls with approx. 1.8 µm thickness each

Ash is a wood species that can develop black heartwood. The occurrence of black heart in ash is related to the age of the tree: while only 6 % trees at age 30 showed black heart, the occurrence increased to 45 % at age 60 and to 100 % at age 100 (Kerr 1998). The formation of black heart in ash is most likely a physiological process (Bosshard 1967). The chromophore heartwood substances can be found in small droplets (size: 5-7 µm) in lumens of parenchyma and ray cells and are considered to develop from storage substances in these cells (Bosshard 1953). The wood anatomy in ash heartwood, i.e., the cell shapes and walls, however remains unaltered. Furthermore, both the mechanical properties (Kollmann 1941; Kühne 1954) and shrinkage and swelling behavior (Trendelenburg 1939) of white and discolored ash are similar.

A concise chemical characterization of ash wood is given in Table 2. When comparing chemical properties of ash to those of other wood species, differences can be found in particular with regard to water soluble extractives, pH and buffering capacity (Schmidt et al. 2012). These properties are considered to possibly influence bond performance, in particular in combination with water based, acid catalyzed adhesives.

Table 2 Chemical characterization of ash; values in brackets are coefficients of variation (%)

Property		Value	Reference
Cellulose	(%)	44.2…46.8	(1)
Lignin	(%)	21.1…30.4	(1)
Polyoses	(%)	36.0	(2)
Pentosan	(%)	22.6…26.7	(1)
Extractives (cold water)	(%)	White: 3.92 (41.3); coloured: 2.63 (24.7)	(3)
pH (surface)	(-)	White: 5.48 (2.4); coloured: 5.22 (3.6)	(3)
Buffering capacity	(mmol/100 g dried wood)	White: 5.75 (18.4) coloured: 4.71 (15.7)	(3)

References: (1) Kollmann (1941) (2) Fengel and Wegener (1989) (3) Schmidt et al. (2012)

Physical and mechanical properties of ash have been determined in several investigations and are presented for example in Baumann (1922), Anonymous (1939), Kollmann (1941), Kollmann (1951), Kühne (1951), Schwenke (1956), Oliver-Villanueva (1993), Sell (1997), DIN 68364 (2003), Bonoli et al. (2005), Wagenführ (2007), Clauß et al. (2014), Niemz (2014) and Niemz et al. (2014). A summary of density and shrinkage properties of ash is given in Table 3.

Table 3 Density and shrinkage values of ash

Property			Value	Reference
Density	ρ_0	(kg/m^3)	410…650…820 372…569…680 (EW) 673…753…866 (LW)	(1)
	ρ_{15}	(kg/m^3)	450…690...860	(2)
Maximum shrinkage	β_{max}	(%)	5.0 (R); 8.0 (T); 0.2 (L)	(2)
Differential shrinkage	β_d	(%/%)	0.17…0.21 (R); 0.27…0.38 (T)	(3)

References: (1) Kollmann (1941) (2) Kollmann (1951) (3) Sell (1997)

The range of density values as well as some strength properties (Table 4) reflects the natural variability of wood. Properties can vary significantly not only within one wood species depending for example on the growth region and condition but also within one tree depending on the position in the trunk. Differences in density between EW and LW reflect the differentiated anatomy between these areas. The comparison of densities of important wood species in central Europe reveals that values for ash and beech (*Fagus sylvatica* L., $\rho_{15,mean}$ = 720 kg/m³) are similar, whereas the density for spruce (*Picea abies* L. Karst., $\rho_{15,mean}$ = 470 kg/m³) is distinctly lower (Kollmann 1951).

Wood is an anisotropic material and therefore, several properties need to be described as a function of one of the principal directions. For example, shrinkage varies

significantly with the main directions radial (R), tangential (T) and longitudinal (L) of the wood (Table 3). Shrinkage values of ash lie between those of spruce and beech.

Strength and stiffness properties likewise need to be specified in dependence of the fiber direction (strength, modulus of elasticity) or the plane (shear modulus). Table 4 contains an overview of mechanical values determined on small, clear ash specimens as summarized by Kollmann (1951) and presented in two current studies (Clauß et al. 2014; Niemz et al. 2014).

Table 4 Mechanical properties of ash; values in brackets are coefficients of variation (%)

Mechanical property			Value		Reference
Bending strength	f_m	(MPa)	56.9…118…206;	95 (5)	(1); (2)
Tensile strength	$f_{t,0}$	(MPa)	69…162…287;	130 (13)	(1); (2)
	$f_{t,90}$	(MPa)	6.9;	R: 12.5 (8); T: 8.3 (5.7)	(1); (2)
Compressive strength	$f_{c,0}$	(MPa)	22.6…51.0…78.5;	43 (3)	(1); (2)
	$f_{c,90}$	(MPa)	R: 10.5 (4.4); T: 10.0 (3.6)		(2)
Shear strength	f_v	(MPa)	8.8…12.6…14.3		(1)
Modulus of elasticity	E_L	(MPa)	4300…13100…17800[a)] 12400 (10)[a)]; 12200 (13.8)[b)]; 9221 (10)[c)]		(1) (2)
	E_R	(MPa)	1510 (16.8)[b)]; 1368 (7)[c)]		(2)
	E_T	(MPa)	759 (7.7)[b)]; 643 (6)[c)]		(2)
Shear modulus	G_{LR}	(MPa)	1468 (4.7)		(3)
	G_{LT}	(MPa)	1234 (16.5)		(3)
	G_{RT}	(MPa)	302 (6.2)		(3)

References: (1) Kollmann (1951) (2) Niemz et al. (2014) (3) Clauß et al. (2014)

For comparability reasons, values from Kollmann (1951) were converted from kg/cm² to MPa.
Mechanical properties relate to an average MC of about 12 %.
Modulus of elasticity values were obtained in a) bending, b) tension, or c) compression tests.

The utilization of ash in structural applications requires strength grading of the sawn wood to predict the load-bearing capacity. An investigation by Glos and Torno (2008) on ash timber created the basis for the assignment of the visual grade “LS10 & better” (DIN 4074-5:2008) to the strength class D40 (EN 338:2009) in EN 1912 (2013). This corresponds for instance to an increase in characteristic bending strength of more than 30 % when compared to the highest visual grade (C30) that can be produced with spruce. In addition, the strength classes in EN 338 (2009) specify further characteristic material properties (e.g., strength perpendicular to the grain, modulus of elasticity). For example, an increased compressive strength perpendicular to the grain of ash timber (D40: 5.5 MPa; C30: 2.7 MPa) allows for an enhancement of structures when forces have to be transferred between building elements.

2.2 Principles of wood bonding

2.2.1 Structure of wood-adhesive bonds

Adhesive bonds create a link between two adherends by means of an adhesive layer. The stability of an adhesive bond is determined by internal, cohesive forces in the adherends and the adhesive layer together with adhesion forces that develop in the adhesive-adherend interfaces (Figure 2a).

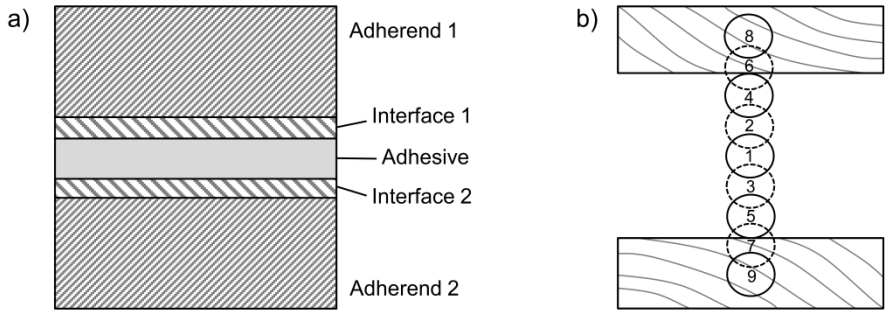


Figure 2 **a) General composition of an adhesive bond (Habenicht 2009, p. 315, Figure 6.1, terms translated by the author of this thesis) and b) interpretation of a wood-adhesive bond as a chain with nine individual links according to Marra (1992, p. 37, Figure 3-1)**

For the composition of wood-adhesive bonds, Marra (1992) presented a differentiated model that interprets a bond as a chain with nine links (Figure 2b). In addition to 'bulk wood' (8, 9), 'bulk adhesive' (1) and the wood-adhesive interfaces (4, 5), the model defines wood (6, 7) and adhesive (2, 3) interphase regions [terminology of the links as proposed by Frihart (2009)]. The adhesive interphase ranges from the wood-adhesive interface to the point where unaffected bulk adhesive properties are obtained. The material properties in the adhesive interphase can for instance be influenced by the ability of wood to absorb adhesive components or by the wood pH value and buffering potential (Marra 1992). The 'wood interphase' is defined as a region where both wood and adhesive are present. It begins at the wood surface and ends with the maximum adhesive penetration depth. The definition of a wood-adhesive bond as a chain implies that the performance of a bond is determined by its weakest link. In wood bonding, a general principle is that cohesion in the adhesive and adhesion between adhesive and wood should exceed the cohesive strength of the wood, i.e., wood is supposed to be the weakest link in the bond model according to Marra (1992).

2.2.2 Adhesion in wood bonding

Adhesion is assumed to result from a complex combination of mechanical, physical and chemical processes. This is reflected by several theories that have been proposed to explain the phenomenon of adhesion. However, up to now no individual theory or combination of theories has been developed that sufficiently explains adhesion (e.g., Bischof and Possart 1986; Pizzi 1994; River 2003; Zeppenfeld and Grunwald 2005; Habenicht 2009). A summary of adhesion theories is given in Bischof and Possart (1986) (Figure 3).

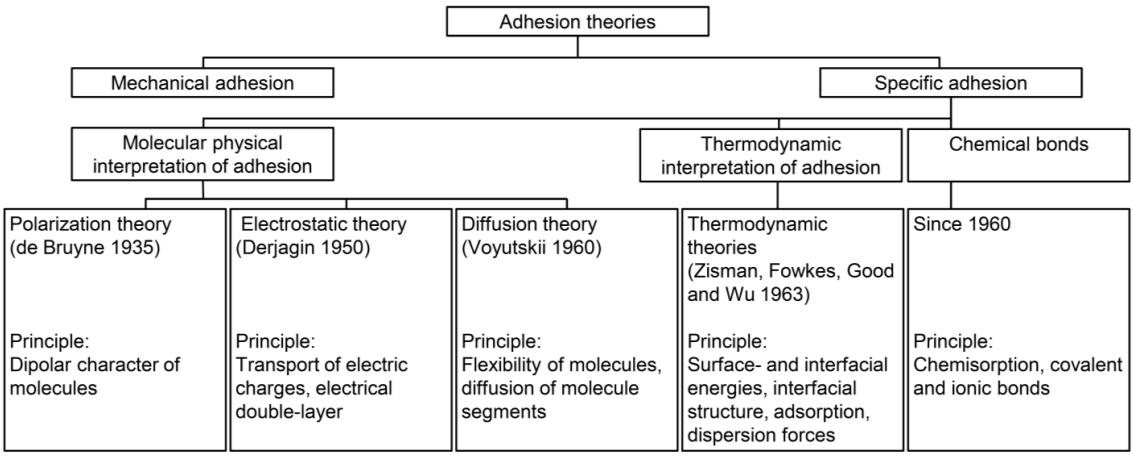


Figure 3 Adhesion theories and principles (Bischof and Possart 1986, p. 27, Figure 3.5, translated by the author of this thesis)

Adhesion depends on the bonding material. The adhesion theories and mechanisms that are considered relevant for wood bonding are presented in the following:

Mechanical adhesion is a rather traditional approach to adhesion and particularly applies to porous materials such as wood. It is based on the idea that a liquid adhesive can penetrate into bonding surface cavities and pores and provide an anchoring effect after hardening. Although mechanical interlocking can explain only a small part of observed adhesion effects (Bischof and Possart 1986) it is considered to contribute to some extent to wood-adhesive bond strength.

Consequently, theories that explain adhesion in more detail and are based on chemical, thermodynamic or physical principles have been suggested. These have been summarized as specific adhesion (Bischof and Possart 1986; Habenicht 2009) and include the following theories and mechanisms:

> The polarization theory was for the first time proposed by de Bruyne in the 1930ies and interprets adhesion as a physical interaction between polar molecules (Bischof and Possart 1986). This theory is however limited to permanent polar molecules and cannot explain adhesion to non-polar substances. The subdivision of physical intermolecular forces (van der Waals forces) into forces between permanent dipoles (Keesom), between permanent and induced dipoles (Debye) and between momentarily induced dipoles (London dispersion forces) allows for a more detailed interpretation of adhesion.

Thermodynamic approaches to adhesion (also referred to as adsorption or wetting), for example, take polar and disperse surface energy fractions of substrate and fluid into account when interpreting adhesion on basis of the contact angle that develops between the two condensed phases. Although wetting is a widely accepted theory to explain adhesion, it involves important drawbacks. For example, the wetting theory does not fulfill the basic requirement of reversibility for thermodynamic analyses. Also, it does not take the surface roughness into account that shows significant impact when measuring on wood surfaces. A particularly strong bond between dipoles is the hydrogen bond which develops between hydrogen and two neighboring atoms that provide a large gradient in electronegativity to hydrogen (e.g., oxygen). Hydrogen bonds in adhesion have also been interpreted as acid-base interactions where one molecule provides the bonding electron pair (proton donator) instead of two bonding partners sharing an electron pair (Habenicht 2009). In general, bonds that are based on the physical interactions between molecules are considered to be of significant importance in wood bonding (Pizzi 1994).

The diffusion theory is based on the assumption that molecules or segments of molecules can interact by interdiffusion across the adhesive-substrate interface (Voyutskii 1963). For the bonding of wood, this theory seems to be restricted to the amorphous wood components where adhesive solvents can produce increased molecule flexibility and facilitate wood-adhesive interactions. In addition, it has been shown for specific thermosetting adhesives (e.g., MUF, PRF) that low-molecular adhesive components can penetrate into the wood cell-wall and create an interpenetrating network beyond the wood-adhesive interface (e.g., Gindl et al. 2002; Konnerth and Gindl 2006; Konnerth et al. 2008). The formation of an interpenetrating network is assumed to enhance strength and durability of wood-adhesive bonds.

Further adhesion theories such as chemical bonds and electrostatic forces are considered to be of minor importance for wood bonding. While formation of covalent bonds under practical bonding conditions is hardly possible, the electrostatic theory has only been verified for a particular material combination (metal-elastomer) and therefore is assumed not to be applicable for wood-adhesive bonding (Pizzi 1994).

The weak boundary layer (WBL) theory was developed by Bikerman in the 1960ies. According to Bikerman (1967), interfacial adhesion failure can be interpreted as cohesive failure in a WBL, i.e., a layer with significantly lower strength than the cohesive strengths of the adhesive or adherend. This means that the WBL theory does not explain an adhesion phenomenon but rather a lack of adhesion. A WBL can for example originate from insufficient wetting, an impurity of the bonding substrate or a deactivation of the bonding surface due to environmental influences (Bikerman 1967). For wood bonding, Stehr and Johansson (2000) distinguished between weak boundary layers of chemical (CWBL) and mechanical origin (MWBL). While a CWBL is described as a weakness on molecular level caused by migration of wood extractives to the surface, a MWBL is considered a weakness on particle level generated during surface preparation.

2.3 Structural wood bonding

2.3.1 Adhesive types and properties

Several adhesive types are considered suitable for adhesive bonding in load-bearing timber products. For surface gluing and finger joints in structural softwood timber, primary chemical systems are PRF, MUF, 1C-PUR and EPI. Habenicht (2009) classified adhesives by their chemical basis and curing mechanism. According to these classifications, PRF, MUF, 1C-PUR and EPI adhesives used in this thesis belong to organic chemistry and show a chemical hardening reaction (polycondensation or polyaddition reaction). All adhesive systems are cold-setting and create thermosetting networks. The suitability of PRF, MUF, 1C-PUR and EPI for bonding of ash wood is examined within this thesis; the adhesive systems can be characterized as follows:

PRF adhesives can be characterized as modified phenol-formaldehyde (PF) systems. The addition of resorcinol to PF has a catalytic effect and enables a cold-setting curing reaction. PRF adhesives are water based two-component systems that harden in a polycondensation reaction and produce a dark brown glueline. They are well known for their high resistance against exposure to moisture or varying climates (e.g., Dinwoodie 1983; Pizzi 2003). As a consequence, PRF-bonded structural timber has frequently been used in outdoor applications.

MUF adhesives result from a modification of urea-formaldehyde (UF) adhesives. The incorporation of melamine in the resin leads to an increased resistance to hydrolysis of the cured adhesive compared to UF. MUF adhesives are water based two-component systems that generate an insoluble whitish glueline in a polycondensation reaction. The curing reaction requires an acidic environment which is provided by addition of the hardener component with pH values as low as 1-2. MUF adhesives are widely used for structural solid wood bonding and accounted for more than 50 % in the European glulam production in 2005 (Mack 2006).

1C-PUR adhesives are one-component systems containing prepolymers with reactive isocyanate groups. The polyaddition reaction of 1C-PUR takes place with participation of moisture, for which reason wood adherends have to provide a minimum MC (Kägi et al. 2006). The curing reaction produces CO_2 which can lead to foam formation in the glueline. 1C-PUR adhesives are free of formaldehyde and solvents and their solid content amounts to 100 %. The production of 1C-PUR bonded structural timber is currently of increasing importance with cross-laminated timber almost exclusively being bonded with 1C-PUR (Lehringer and Gabriel 2013).

EPI adhesives are two-component systems composed of water based emulsion and isocyanate as a cross-linking agent. The curing reaction combines physical processes of the emulsion component with chemical reactions of isocyanate with molecules providing active hydrogen (e.g., alcohols, water, carboxylic acids) (Grøstad and Pedersen 2010). As with 1C-PUR, CO_2 develops during the reaction of isocyanates with water and causes foaming of the adhesive. EPI produces a light colored glueline and is free of formaldehyde.

In general, the adhesives presented above can be referred to as synthetic materials with isotropic mechanical properties. Knowledge of these properties is important for a

fundamental understanding of load transfer in wood-adhesive bonds and thus, for a good performance of a bonded product. Strength and elasticity of cured adhesives depend on a number of factors, for example on the chemical nature of the adhesive, on molecular weight and polymerization degree as well as on shape and orientation of the molecules (Scheikl 2002). Material properties of commercial PRF, MUF, 1C-PUR and EPI adhesives have been determined in several investigations (Table 5). A comprehensive summary of mechanical properties of wood adhesives is given in Stöckel et al. (2013).

Table 5 Material properties (mean values of modulus of elasticity (MoE) and tensile strength f_t) of PRF, MUF, 1C-PUR and EPI, determined by means of nanoindentation (NI) in bond lines (BL) and adhesive films (AF) as well as in tensile tests (TT) on AF

Method	Material property	PRF	MUF	1C-PUR	EPI	Reference
NI (BL)	MoE (GPa)	5.8–7.8[a]	7.6–8.9[a]	1.5–3.0[a]	ca. 3.8[a]	(1)–(5)
NI (AF)	MoE (GPa)	5.2[a]	8.6[a]	ca. 2.4[a]	-	(3)
TT (AF)	MoE (GPa)	3.3–3.6	3.1–7.0	0.33–1.1	-	(1), (6)–(8)
	f_t (MPa)	29.5–33.8	26.7-39.8	11.5–26.6	-	(1), (8)

References:
(1) Clauß et al. (2011a)
(2) Follrich et al. (2010)
(3) Konnerth et al. (2006b)
(4) Konnerth et al. (2007b)
(5) Konnerth and Gindl (2008)
(6) Konnerth et al. (2007a)
(7) Gindl and Müller (2006)
(8) Kläusler et al. (2013)

a) Mean MoEs in references (2), (3) and (5) are approximate values which were taken from diagrams.

Note: All investigations were performed at ambient temperature. Test specimens were conditioned at 65 % relative humidity in investigations (1), (2), (4) and (8); conditioning is however not precisely specified in investigations (3), (5), (6) and (7). Due to moisture dependent characteristics of adhesives the comparability of material properties therefore may be limited.

As can be seen in Table 5 the modulus of elasticity (MoE) significantly depends on the test method. However, a clear trend can be observed towards a higher stiffness of MUF and PRF compared to 1C-PUR and EPI, although the available data for EPI is limited to NI investigations. The use of commercial adhesives with different formulations possibly explains the variability of MoE for some adhesive types. For example, originally rigid MUF adhesives can be modified towards a more elastic system by adding Polyvinylacetate (PVAc).

The influence of moisture on material properties of PRF, MUF and 1C-PUR adhesives was investigated by Konnerth et al. (2010) by means of NI and by Kläusler et al. (2013) in TT. Both studies showed significantly reduced MoEs of adhesive films at high MC. After immersion of adhesive films into water, Konnerth et al. (2010) found the most significant decrease of MoE for PRF (-90 %), MoE reductions for MUF and 1C-PUR were -60 % and -40 %, respectively. These results are explained with a softening effect caused by the water that is absorbed by the adhesive films, the quantity of which significantly varies with the adhesive chemistry as determined by Wimmer et al. (2013). The research by Kläusler et al. (2013) showed the most significant loss in MoE for MUF

(-76 %) when comparing values obtained after specimen conditioning at 5 % and 95 % RH. The decrease for two 1C-PURs and PRF was between -31 % and -47 %.

Similar characteristics could be observed in strength tests for MUF and 1C-PUR with decreasing strength at increasing humidity. The strength of PRF was independent from moisture influence (Kläusler et al. 2013). After redrying, the adhesive samples regained their initial performance in terms of MoE and strength to a large extent, for some adhesives the original properties were even outreached. For EPI, no data regarding moisture dependent elasticity or strength is available in literature.

2.3.2 Requirements for adhesives

During service life, wood-adhesive bonds in timber structures have to tolerate various loading conditions that comprise mechanical, physical, chemical and biological loads. Mechanical loads comprise the load of the structure together with varying loads caused by wind, snow or working loads. In addition, the exposure to surface weathering and radiation (ultraviolet, infrared) as well as changing temperature and humidity can result in considerable stresses. In practice, these effects often occur simultaneously, interfere with each other and cause a highly complex load situation that can lead to a chemical and physical deterioration of the wood-adhesive bond (Habenicht 2009; Clauß 2011). To minimize the risk of bond degradation, wood adhesives for structural purposes must have thermosetting properties and provide permanent cross-linking and water resistance.

For bonding of lamellas and finger joints, four chemical systems are primarily being used at present (MPA BAU TU München 2014; MPA Stuttgart 2014a, b):

- PRF
- MUF
- 1C-PUR
- EPI

To be approved for structural wood bonding in Europe, adhesives have to meet substantial performance requirements that are specified in standards depending on adhesive chemistry:

- EN 301 (2013) for PRF and MUF
- EN 15425 (2008) for 1C-PUR
- EN 16254 (2013) for EPI

The corresponding test methods give an indication of the importance of moisture-related behavior of wood-adhesive bonds. All test methods, including the evaluation of bond strength (e.g., EN 302-1:2013; EN 302-3:2013; EN 302-4:2013), creep behaviour (e.g., EN 15416-2:2007) and resistance to delamination (EN 302-2:2013), involve water storage or temperature and humidity cycling before or during testing.

EN 301 (2013), EN 15425 (2008) and EN 16254 (2013) also allow for a classification into adhesive type I and II. This classification is related to the expected moisture exposure of the bonded product. While adhesive type I may be used in service classes 1 – 3 according to DIN EN 1995-1-1 (2010), i.e. in interior and exterior applications, adhesive type II is restricted to interior applications (service class 1). In Germany, only adhesive

type I may currently be used for structural wood-adhesive bonds. This is a consequence of the collapse of the ice rink in Bad Reichenhall in 2006 which led to a raise of safety requirements in Germany. The collapse was caused by a combination of influencing factors, with important ones being the adhesive and bond degradation induced by moisture (Anonymous 2006; Winter and Kreuzinger 2008).

2.4 Bond durability

2.4.1 Evaluation of bond durability

Wood-adhesive bonds have to withstand complex load and exposure conditions during service life that may lead to bond alteration and often also to bond deterioration over time. Bond durability is considered as the resistance of an adhesively bonded joint against deterioration caused by the main influencing factors heat, moisture and stress (Dinwoodie 1983). In addition, radiation and surface weathering may be considered as parameters that influence bonds.

A wide range of approaches to evaluate bond durability has been developed. For example, long-term experiments under actual service conditions can be used. In such investigations, specimens are stored under outdoor, sheltered outdoor or indoor conditions, sometimes in combination with mechanical loads for several years. After that the bond performance is determined and compared to the initial performance. Long-term investigations on structural wood adhesives have for example been performed by Egner and Kolb (1966) and Raknes (1983, 1997). Although long-term investigations provide valuable and reliable information about bond durability, these tests are inefficient for the development of new adhesives due to their long duration.

To predict the durability of wood-adhesive bonds within shorter time spans, accelerated aging approaches have been developed. Test methods frequently include varying temperature and moisture to induce swelling and shrinking stresses that possibly lead to bond degradation (Dunky and Niemz 2002). In this context, the moisture-related durability of wood-adhesive bonds is of particular importance. This can also be seen from the high number of test standards that involve moisture variations (Frihart 2009). In fact, all performance tests within an adhesive approval procedure for structural bonding (section 2.3.2) involve water soaking, boiling or temperature and humidity cycling.

Dinwoodie (1983) classified durability tests according to their exposure and load situation into single exposure, cyclic exposure and mechanical tests. In single exposure tests, specimens are subjected to one exposure condition (e.g., soaking or boiling in water or storage at high temperature or humidity), usually followed by a strength test. Tensile shear (EN 302-1:2004) and block shear tests (EN 392:1995) used within this thesis can be classified as single exposure tests. The information obtained from single exposure tests is however considered limited, as varying environmental conditions are not taken into account. Cyclic exposure tests are used to simulate effects from varying exposure situations and involve methods such as vacuum and pressure soaking with subsequent drying, temperature and humidity cycling or UV-radiation of specimens. An important cyclic exposure test to determine bond durability of structural adhesives is the

delamination test according to EN 302-2 (2013). Mechanical tests comprise fracture mechanic and creep investigations.

The greatest challenge of accelerated aging tests is to predict the service life of a bonded product. Although several approaches have been made to correlate test results of accelerated aging and natural weathering, correlations in many cases only apply to a specific bonded product or adhesive, or are only effective for a specific climate zone (Dinwoodie 1983; Dunky and Niemz 2002).

In the following, the shear and delamination tests used within this thesis and their relevance with respect to bond durability are presented.

The block shear test (EN 392:1995, recently incorporated in the glulam standard EN 14080:2013) is frequently used for a rapid assessment of bond integrity. The determination of shear strength in compressive action and wood failure percentage within this test usually serve quality assessment purposes within factory production control. The test specimens are stored at room temperature prior to testing; no exposure to alternating or elevated temperature or humidity is involved.

The tensile shear test according to EN 302-1 (2013) is very common for determination of performance characteristics of structural adhesives in Europe. The standard specifies a variety of treatments to be applied prior to testing. Treatments are soaking of specimens in cold or boiling water with or without subsequent re-drying or exposure to elevated temperature. Shear results (strength and wood failure) serve as a basis for classification and for stability evaluation of adhesives at specified treatments. In research studies, the tensile shear test is frequently used to compare bond performance of adhesives or varying bonding parameters (e.g., Müller et al. 2005; Konnerth et al. 2006a; Clauß et al. 2011b; Brandmair et al. 2012).

The delamination test is a rapid method to predict bond durability of laminated timber under exterior service. It was first developed by Truax and Selbo (1948) who correlated the amount of delamination in laminated timber sections after long-term exterior exposure (4 years) with delamination values of specimens that had been treated in a vacuum-pressure, soaking-drying cyclic procedure. The idea of the test is that induced stresses in the bond are similar to those which result from exterior exposure. This test method allows for an evaluation of bond durability in 21 days by using extreme moisture changes to generate swelling and shrinkage of wood and as a result, stresses in the bonds. If exceeding bond strength, these stresses can initiate bond failure. Further developments at the Forest Products Laboratory, Madison, Wisconsin led to a reduced test duration of only three days (Selbo 1964). The findings from these investigations were integrated in a standardized evaluation process (ASTM D2559) of adhesives for laminated timber for exterior exposure (River et al. 1991). Today, delamination tests are used both to evaluate the suitability of adhesives for structurally laminated wood (ASTM D2559:2012; EN 302-2:2013) and in factory production control (EN 14080:2013). While the delamination test according to ASTM D2559 (2012) is intended for evaluation of adhesives for exterior service only, the European standards comprise test methods both for interior and exterior use. The test methods vary for example with respect to growth ring alignment, lamella thickness and drying conditions (temperature and humidity). Although bond durability from natural weathering has been correlated with

accelerated aging results by Truax and Selbo (1948), a limitation is that the correlation is valid for only one climate zone as well as specific wood species and specific adhesives. Other climate conditions or wood species may lead to different results.

The correlation between results from block shear tests and resistance to delamination is however not conclusively clarified. On the one hand, Zeppenfeld and Grunwald (2005) assume a positive correlation between both methods. A research by Steiger et al. (2014) showed comparable results for both methods by trend but not for all individual cases. In contrast, examinations with hardwoods gave contradictory results (Aicher and Reinhardt 2007) or no evidence for a correlation between shear and delamination test results (Truax and Selbo 1948; Ohnesorge et al. 2010; Schmidt et al. 2010).

2.4.2 Strains and stresses in bonds due to moisture change

Strains and stresses in laminated timber occur with changing wood MC and temperature. They result for instance from different swelling or shrinkage coefficients due to the anisotropy of wood or from moisture gradients in the cross-section or between lamellas. On microscale, stresses are already induced during the bonding process when the adhesive solvent causes swelling of the surface layers and after that, polymerization leads to adhesive shrinkage (Frihart and Wescott 2008; Hass et al. 2012). At the beginning of service life, stresses occur when bonding lamellas with different moisture content (Niemz et al. 2005) or during initial adjustment of moisture to environmental conditions. However, the frequent variations of wood MC due to temperature and humidity changes during service life are assumed to have the greatest impact on bond durability (Frihart 2005). Under outdoor conditions, radiation and surface weathering may be additional factors of influence. The resulting swelling and shrinkage stresses are of a fluctuating character and can initiate bond failure (Truax and Selbo 1948), with starting points being local discontinuities (e.g., knots, grain deviations or growth ring angle differences) or abrupt material property changes (River 2003).

In delamination tests, severe stresses on the bonds are deliberately generated in sections obtained from small laminated timber beams to evaluate bond durability within short time. First, vacuum-pressure soaking causes maximum swelling of the specimens and leads to a MC that depends on the wood species but can be as high as 150 % for spruce (Schmidt and Knorz 2010). Although bond fractures are rarely observed after soaking, wood swelling induces severe stresses (e.g., shear) that are considered to at least weaken the bond (Frihart 2009). The subsequent drying procedure produces a significant moisture gradient between the quick-drying end-grain faces and the fully swollen core and, as a consequence, high tensile forces perpendicular to the glueline (Figure 4). The maximum stresses are generated at the end-grain faces. Delamination occurs where these stresses exceed bond strength (Marra 1992).

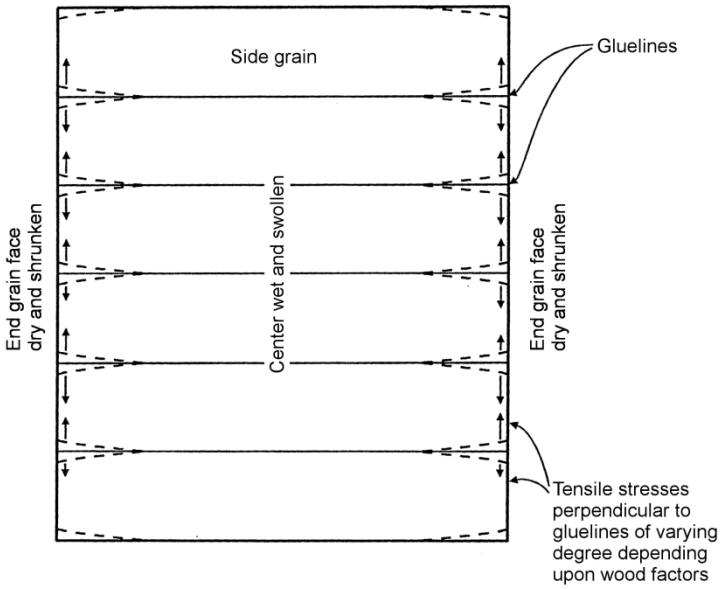


Figure 4 Tensile stresses perpendicular to the gluelines in delamination specimen (lateral view) that result from the drying procedure in delamination tests (Marra 1992, p. 394, Figure 11‑8)

Both EN 302-2 (2013) and ASTM D2559 (2012) use flat-sawn lamellas, the layup specification in the bonded members however varies. ASTM D2559 (2012) specifies an alternating growth ring orientation of the lamellas and therefore, mainly tensile stresses on the gluelines can be expected. In contrast, EN 302-2 (2013) defines a consistent growth ring alignment that assumingly leads to a combination of tensile stresses perpendicular to the grain and shear stress due to the restrained cupping during drying of the lamellas. Although the make-up of the laminated test specimens is close to reality one restriction of the delamination test is that the wood is given no opportunity to stress relax during the fast drying procedure.

In general, there is a need to scientifically understand moisture-related durability. For this reason, a model was developed (Frihart 2005; Frihart and Wescott 2008; Frihart 2009) that relates bond durability to strain in the wood-adhesive bond generated by moisture increase. The model discusses strain distribution as a function of two adhesive groups: in-situ polymerized (e.g., PRF, MUF) and pre-polymerized (e.g., 1C-PUR, EPI) adhesives. In-situ polymerized adhesives are able to infiltrate the cell wall and develop a rigid network during hardening. Adhesive penetration into cell walls changes their mechanical properties and reduces the degree of swelling (Gindl and Gupta 2002; Lukowsky 2002; Gindl et al. 2004). This stabilizes the cell walls in the interphase and reduces the stress at the interface in case of moisture change. Stress is then primarily distributed in the wood (Frihart 2009). In contrast, pre-polymerized adhesives consist of polymers with higher molecular weight that cannot infiltrate and stabilize cell walls. These adhesives however provide a higher flexibility due to limited cross-linking so that interfacial strain can be dissipated in the glueline rather than in the wood (Frihart and Wescott 2008; Frihart 2009). In Figure 5, differences between clear and adhesive-penetrated cell walls in case of moisture increase are illustrated.

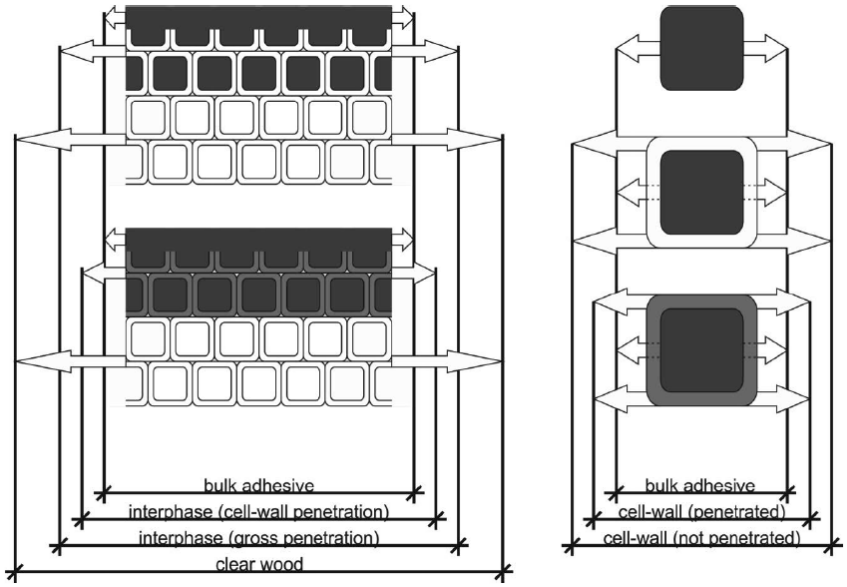


Figure 5 Response of wood-adhesive bonds with clear and adhesive-penetrated cell walls in case of moisture increase; arrows show the degree of swelling with differences between bond components being an indication for stress occurrences (Hass 2012, p. 36, Figure 14)

The above described model emphasizes the importance of the interphase to distribute stresses in case of moisture changes. In general, a homogeneous stress distribution with limited local stress concentrations is considered to contribute to higher moisture-related bond durability. Adhesive penetrated cell walls show the advantage that mechanical and physical properties are between bulk wood and adhesive and therefore, provide a buffering action between the individual bulk materials. On the other hand, stresses may be more evenly distributed in bonds with pre-polymerized adhesives because of the higher elasticity when compared for the same glueline thickness.

2.4.3 Influencing factors on moisture-related performance of wood-adhesive bonds

Bond formation and bond performance depend on a large number of parameters. Marra (1992) described the strength and durability performance of a glued product in the 'equation of performance' (Equation 1):

$$\text{Glued Product performance} = \underset{(\approx 483\ \text{MPa})}{70000\ \text{psi}} \pm \sum \underset{\text{(A)}}{\text{Adhesive Composition Factors}} \pm \sum \underset{\text{(B)}}{\text{Wood Property Factors}} \pm \sum \underset{\text{(C)}}{\text{Wood Preparation Factors}} \pm \sum \underset{\text{(D)}}{\text{Adhesive Application Factors}} \pm \sum \underset{\text{(E)}}{\text{Wood Geometry Factors}} \pm \sum \underset{\text{(F)}}{\text{Product Service Factors}} \quad (1)$$

Marra (1992) differentiates between bond formation which is mainly affected by groups (A) to (D) and bond performance which primarily depends on (F). Wood geometry factors (E) affect both bond formation and performance. However, a good bond performance always requires the formation of an intact bond. In the following, major influences on bond performance as related to the factor groups A-E that are relevant within this thesis are presented.

Adhesive composition (A)

An important factor for bond durability is the type of adhesive. First investigations showed that adhesives that perform well with softwoods may vary in their performance with hardwoods. This is due to the fact that structural bonding of hardwoods was of minor importance in the last decades and adhesives were mainly developed for softwoods. In delamination tests with beech, MUF-bonded specimens showed a significantly better performance than 1C-PUR bonds (Aicher and Reinhardt 2007; Schmidt et al. 2010). Furthermore, Stapf et al. (2007) found distinct differences between adhesive types when examining the resistance to delamination of oak bonds. The most promising adhesive in combination with oak was PRF; EPI and 1C-PUR bonds showed lower resistance to delamination. Bonded ash was examined in shear tests by Brandmair et al. (2012). Higher bond strength and wood failure was determined with PRF than with 1C-PUR, in particular after soaking of the specimens with water.

The lower resistance to delamination of 1C-PUR bonded hardwood is probably attributed to the characteristic that hydrogen bonds between adhesive and functional groups in wood can be disrupted by water molecules (Clauß 2011). Although this is potentially a reversible effect, permanent damage may occur in presence of water.

Wood property (B) and wood geometry (E)

Bond performance is significantly influenced by the bonded wood species. In general, it is regarded more difficult to achieve a good bond performance with hardwood than with softwood (Marra 1992). This is because hardwoods usually have a higher density than softwoods [ash: $\rho_{15,mean}$ = 690 kg/m³, spruce: $\rho_{15,mean}$ = 470 kg/m³ (Kollmann 1951)]. The increased density is associated with reduced porosity and thicker cell walls. This influences adhesive penetration in lumens and cell walls (Frihart 2008) and possibly produces weaker bonds. In addition, high density is often connected with increased shrinkage and swelling as well as higher strength and stiffness. In case of moisture changes, these wood properties account for significantly higher stresses in hardwood bonds and increase the danger of bond separations. The higher strength of hardwoods also implies that the strength difference between wood and wood-adhesive bond is lower. This means that a bond failure is more likely to happen with hardwoods than with softwoods. Several studies have proven that in particular a high resistance to delamination is more difficult to achieve with hardwoods than with softwoods (Frühwald et al. 2003; Aicher and Reinhardt 2007; Stapf et al. 2007; Schmidt et al. 2010).

Moreover, the different anatomy between softwoods, ring-porous and diffuse-porous hardwoods may influence adhesive penetration and thus have an impact on bond performance. Further properties that are important for bonding are for instance extractives content, pH-value, buffering capacity and water absorption. These were also found to vary significantly with wood species in several investigations (e.g., Boehme and Hora 1996; Niemz et al. 2012; Schmidt et al. 2012).

Furthermore, the performance of wood-adhesive bonds in laminated timber significantly depends on the dimension of the laminates (Truax and Selbo 1948). A research by Aicher and Reinhardt (2007) with beech wood showed higher delamination values with increasing specimen width. In addition, the thickness of the laminates was found to influence bond durability (Ohnesorge et al. 2008; Ohnesorge et al. 2010). Besides, the

growth ring alignment in the laminates has an effect on the resistance to delamination (Marra 1992). For example, an investigation by Ohnesorge et al. (2008) with beech wood showed higher delamination values with flat-sawn lamellas than with quarter-sawn wood. Ohnesorge et al. (2008) explained these results with the higher dimensional stability of quarter-sawn lamellas which leads to lower stresses in the bond at moisture change.

Wood preparation (C)

Another key element for bond durability is the bonding surface. For instance, the wetting and, as a consequence, the intensity of wood-adhesive interactions and the formation of the interface are affected by the surface. Moreover, the porosity and the moisture content of the bonding surface influence adhesive penetration and thus have an impact on stress transfer in the bond (Marra 1992).

The structure of the bonding surface depends both on the wood anatomy and the quality of the surfacing method. The machining process greatly influences the roughness of the surface and determines if the substrate is accessible for wetting and penetration. In addition, the surface preparation determines if damaged cells in the surface and subsurface layers occur. In this case, damaged cells often act as a MWBL and thus, weaken the bond (Stehr and Johansson 2000).

Peripheral planing, sanding and face milling are established methods to generate bonding surfaces. Bonding surfaces that are prepared with the most common method peripheral planing greatly vary with the condition of planing knives. While sharp knives produce surfaces with sound and open cells that are accessible for penetration (de Moura and Hernández 2005; Cool and Hernández 2011a) the use of dull knives can lead to significant cell damage (Singh et al. 2002; Kläusler et al. 2014b). Accordingly, bond performance varies with the quality of planed surfaces (Jokerst and Stewart 1976; Kläusler et al. 2014b). Sanding frequently produces surfaces with crushed cells (Murmanis et al. 1986; de Moura and Hernandez 2006; Hernández and Cool 2008) and torn-out fibrils (de Moura and Hernández 2005; Cool and Hernández 2011b). While fibrillation is considered to improve bond performance by strengthening the glueline (Stehr and Johansson 2000), damaged cells can both promote bond quality by avoiding excessive penetration (de Moura and Hernández 2005) and have negative impact by inhibiting penetration (Murmanis et al. 1983). In general, the quality of the bond performance highly depends on the grit size that has been used for surface preparation by sanding (Jokerst and Stewart 1976; Kläusler et al. 2014b). Face milling generates bonding surfaces that are characterized by low cell damage and cell wall fibrillation due to the cutting action perpendicular to the grain. The bond performance with face milled surfaces was found to be equal or better with black spruce (*Picea mariana* [Mill.] B.S.P.) by Cool and Hernández (2011a) and with beech (*Fagus sylvatica* L.) by Kläusler et al. (2014b) when compared to peripheral planing and sanding.

An opportunity to increase bond durability is the modification of bonding surfaces by application of primers such as hydroxy-methylated resorcinol (HMR) or *N,N*-dimethyl-formamide (DMF). HMR primer is assumed to stabilize amorphous regions in the wood (Sun and Frazier 2005) by penetrating into the cell walls of the bonding substrate (Gardner et al. 2005). This reduces the shrinking and swelling behavior as well as mechanical stresses due to moisture changes and as a consequence, contributes to

increased bond durability. Increased resistance to delamination after HMR treatment could be shown with various softwoods (Vick et al. 1996; Vick 1997; Vick and Okkonen 2000) and hardwoods (López-Suevos and Richter 2009; Ohnesorge et al. 2010). DMF primer is assumed to produce a pre-swollen condition in the bonding surface (Kläusler et al. 2014a). In case of moisture increase, the swelling and moisture-induced stresses are reduced. In tensile shear tests, the DMF treatment led to an enhancement of both bond strength and wood failure (Kläusler et al. 2014a).

Adhesive application (D)

Bonding is a time sensitive process and therefore, open and closed assembly times have significant influence on bond formation and performance. In particular, the closed assembly time, i.e. the time between assembling the adhesive covered lamellas and application of pressure, proved to affect the resistance to delamination. In investigations with beech, longer closed assembly times showed to increase the resistance to delamination of MUF bonds, in combination with 1C-PUR however no clear trend could be observed (Aicher and Reinhardt 2007; Schmidt et al. 2010).

The closed assembly time is a determining parameter for glueline thickness and formation of the interphase. A prolongation of the closed assembly time leads to an increased viscosity. At the time of pressure application, the adhesive viscosity determines penetration depth and glueline thickness. In addition, waste of adhesive can be avoided with longer assembly times since squeeze-out is prevented.

3 Material and methods

3.1 Ash wood and bonding surface preparation

Investigations were carried out with ash wood (*Fraxinus excelsior* L.) originating from southern Bavaria, Germany. Before bonding, the sawn ash boards were stored at 20°C temperature (*T*) and 65 % relative humidity (RH) (20/65) and at 20°C and 95 % RH (20/95), respectively, for several weeks. Moisture content (MC) was determined following EN 13183-1 (2002). Density was evaluated according to ISO 3131 (1975) and adjusted to 12 % MC (ρ_{12}). Mean values and standard deviations (sd) were calculated both for MC and ρ_{12}. Ash wood used within the investigations is characterized in Table 6. As differences in bond durability between white and discolored ash were shown to be insignificant in preliminary investigations both white and discolored ash were used.

Table 6 Characteristics of ash wood used in investigations

Paper	$\rho_{12,mean}$ ($\rho_{12,sd}$)	Conditioning		MC_{mean} (MC_{sd})	Timber: L x W x T		Growth ring alignment[a)]
		T	RH		Sawn timber	After preparation for bonding	
	(kg/m^3)	(°C)	(%)	(%)	(mm^3)	(mm^3)	
I	661 (60)	20	65	11.4 (1.4)	550 x 180 x 40	500 x 160 x 30	Tangential
II	633 (75)	20	65	10.8 (1.2)	1050 x 170 x 40	500 x 160 x 30[b)] 320 x 140 x 5[c)]	Tangential[b)] 30°-90°[c)]
III	618 (46)	20	95	19.7 (0.6)	550 x 185 x 40	80 x 180 x 30	Tangential

a) Growth ring alignment in timber cross-section with width as reference surface
b) Delamination test
c) Shear test, roughness measurements and microscopy

The investigations of the influence of surfacing methods on bond quality involved peripheral planing, sanding and face milling. Machines and parameters used for surface preparation in paper II are shown in Table 7. Bonding surfaces in paper I and III were prepared by peripheral planing only with machine and parameters as specified in Table 7, column 2. In a pretest, planed surfaces were plasma treated. This test did not lead to an improved bond performance and plasma treatment was therefore not further pursued.

Table 7 Surfacing machines and parameters (paper II)

	Planing	Sanding	Face milling
Surfacing machine	Otto Martin “T43”	Kuendig “MAGIQ”	Ledinek „Rotoles 400 D“
Characterization of blades / sanding belt	HSS, freshly sharpened	80 grit, new sanding belt	HSS, freshly sharpened
Number of cutting edges *z*	4	n/a	48
Cutting speed v_c (m/s)	32.7	17	80
Feed speed v_f (m/min)	6	7	10
Feed f_z (mm)	0.3	n/a	0.07
Cutting depth (mm)	2	0.5	2.5
Rake angle	34°	n/a	Axial: 10°, Radial: 15°

3.2 Adhesives and bonding parameters

Five adhesives were used within the investigations. They reflect the range of chemical systems primarily used for production of structural laminations (PRF, MUF, PUR[2] and EPI). The adhesives fulfill European standard requirements (EN 301:2006; EN 15425:2008), or have a general technical approval which both allows for bonding of load-bearing timber products made of certain softwoods as specified in relevant standards (e.g., for glulam in EN 14080:2013). Given the importance of MUF systems, two MUF adhesives were included in the initial performance evaluation (paper I) with two mixing ratios for one MUF adhesive. The investigations in paper II and III were made with the best performing MUF adhesive and mixing ratio from paper I.

While PUR adhesive consists of one component, PRF, MUF and EPI adhesive systems are composed of two components that were mixed directly before application on the adherend. The time between surface preparation and bonding was at maximum 6 h. Adhesives were spread one-sided with a spatula. Both the open assembly time (< 5 min) and the pressure (1.2 MPa) were kept constant for all investigations. An overview of adhesives and bonding parameters that vary with the adhesive is shown in Table 8.

Table 8 Adhesives and bonding parameters

Adhe-sive	Mixing ratio resin/hardener (parts by weight)	Adhesive spread (g/m^2)	Closed assembly time (min)	Press time[b)] (h)	Solid content[c)] (%)	Paper
PRF	100/20	450	15, 30, 60, 80, 100	6	55.8	I
			60			II
		n/a[a)]	80	12		III
MUF-1	100/60	450	30, 60, 90, 120	12	not determined	I
MUF-2	100/20	450	30, 60, 90	12	not determined	I
	100/50	450	30, 50, 70	6	50.2	I
			70			II
		n/a[a)]	30 (0.01mm), 70 (0.1/0.2 mm)	12		III
PUR	n/a	200	20, 50	24	100	I
		140	20	2.5		II
		n/a[a)]	5	12		III
EPI	100/15	300	10, 25	2.5	66.4	I
			25			II
		n/a[a)]	10	12		III

a) Adhesive spread could not be quantified as it was determined by glueline thickness (0.1 mm/0.2 mm)
b) Press times were defined in consultation with adhesive manufacturers
c) Solid content for PUR as declared in the manufacturers' technical data sheet, values for PRF, MUF2 and EPI were determined following EN 827 (2005)

[2] Hereinafter, the abbreviation PUR is used for the 1C-PUR adhesive. This also corresponds to the abbreviation used in papers I, II and III.

3.3 Test specimens

For investigations in paper I and II, bonded members were prepared following EN 302-2 (2004). Figure 6a shows a bonded member and the cutting pattern as used in paper I to obtain delamination specimens (Figure 6c), slices for microscopy, and shear specimens (Figure 6b). In addition, solid timber was tested in block shear tests to determine reference values (paper I). For the investigation of bonding surfaces (paper II), four delamination test specimens were cut from each bonded member, two of which were tested in different delamination test procedures each. Tensile shear specimens (Figure 6d) were prepared within this investigation according to EN 302-1 (2004). The preparation of test specimens with the glueline thicknesses 0.01 mm (GL0.01), 0.1 mm (GL0.1) and 0.2 mm (GL 0.2) for strain measurements (paper III) is shown in (Figure 6e).

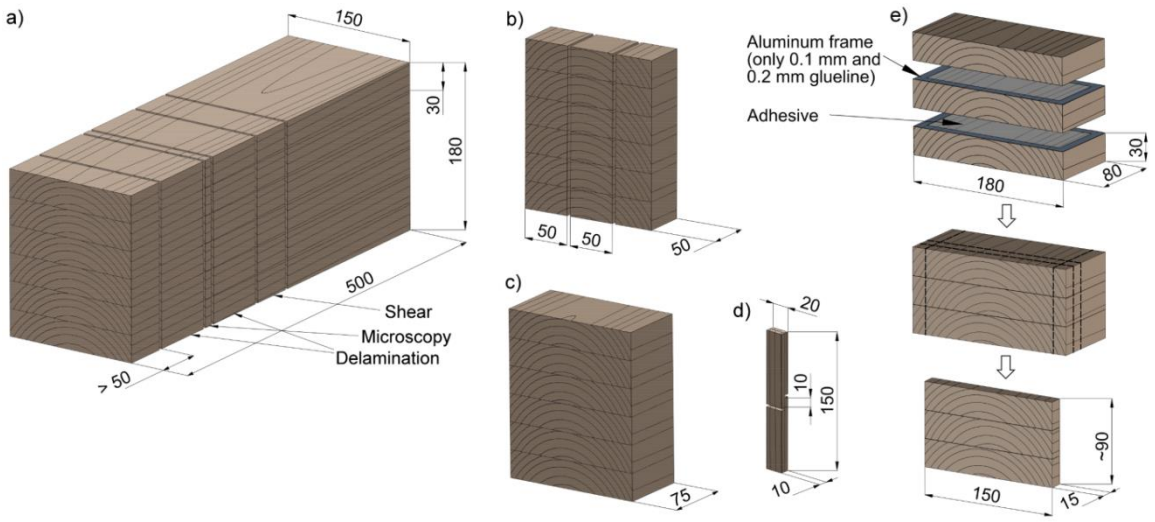


Figure 6 Overview of test specimens: a) Bonded member and cutting of test specimens, b) shear specimens, c) delamination specimen, d) tensile shear specimen, e) preparation scheme and specimen for strain measurement

For reflected light microscopy, pre-cut sections from the bonded members (Figure 6a, paper I) and strain measurement specimens (Figure 6e, paper III) were used. To obtain test specimens for transmitted light microscopy, small blocks were removed from the sections cut from the bonded members (Figure 6a, paper I) or from the tensile shear specimens (Figure 6d, paper II). From these blocks, microtome slices with a thickness of approximately 20 µm were taken from the end grain surfaces and dyed with safranin or methylene blue depending on the adhesive to analyze adhesive penetration. For surface examination with scanning electron microscopy (paper II), blocks with 20 mm length, 10 mm width and 5 mm thickness were cut and coated with gold. For this evaluation as well as for surface roughness measurements, pieces from the same batch as for preparation of tensile shear specimens were used.

3.4 Shear tests

Shear tests have been performed according to EN 392 (1995) in compression loading (paper I) and following EN 302-1 (2004) in tensile loading (paper II). All specimens were tested parallel to the grain with the glueline being the shear plane. Block shear tests were carried out with a universal testing machine (DOLI Elektronik GmbH, Munich, Germany) on specimens that had been conditioned at 20/65 which corresponds to treatment A1 according to EN 302-1 (2004). The displacement rate was specified with 3 mm/min. Shear tests in tensile loading were performed after half of the specimens each had been subjected to treatment A1 and A4 (Table 9), respectively, according to EN 302-1 (2004).

Table 9 Treatments A1 and A4 according to EN 302-1 (2004)

Name of treatment	Treatment specification
A1	Conditioning at 20/65 and testing in dry state
A4	6 h cooking, 2 h storage in water and testing in wet state

Treatment A1 involves conditioning at 20/65 and testing in dry state, whereas treatment A4 contains 6 h cooking, 2 h storage in water and testing in wet state. Tensile shear tests were carried out with a universal testing machine (TesT GmbH, Erkrath, Germany). Displacement rates were chosen to meet the time to failure of 60 ± 30 s as specified in the standard.

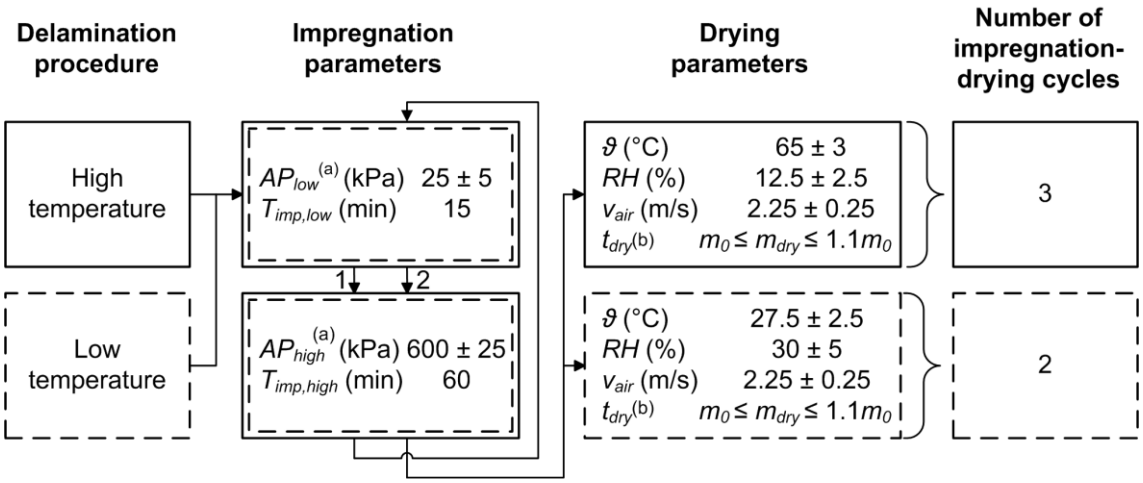
For shear tests both in compression and tensile loading, the shear strengths f_v were calculated according to Equation 2:

$$f_v = \frac{F_{\max}}{A} \quad \text{(MPa)}, \tag{2}$$

where F_{max} is the load at failure and A is the glueline area between laminates. Furthermore, the percentage of wood failure (*WF*) was visually estimated to the nearest 5 %.

3.5 Delamination test

The resistance to delamination was determined according to EN 302-2 (2004) in accelerated exposure testing. EN 302-2 (2004) differentiates between high temperature (HT, for adhesive type I) and low temperature (LT, for adhesive type II) test procedures. While parameters for specimen impregnation with water are in agreement for both test procedures, the drying parameters and the number of cycles vary between HT and LT (Figure 7). Bond performance in paper I was evaluated with test procedure HT. The bonding quality of different surfaces (paper II) was analyzed both by means of LT and HT. The tests were performed with a drying plant ULWA-E (Ulrich Lübbert Warenhandel GmbH & Co. KG, Henstedt-Ulzburg, Germany).



a) Absolute pressure
b) The drying period is finished when the mass of the test specimen m_{dry} reaches a value between m_0 (specimen mass before test) and 1.1 times m_0

Figure 7 Delamination test parameters according to EN 302-2 (2004)

Immediately after the test, the lengths of the glueline openings were determined on the end grain surfaces. The total delamination of a test specimen D_{SP} (Equation 3) and delamination values for every individual glueline D_{GL} (Equation 4) were calculated:

$$D_{SP} = \frac{\sum l_{Delam,SP}}{\sum l_{GL,SP}} \cdot 100 \quad (\%), \tag{3}$$

$$D_{GL} = \frac{\sum l_{Delam,GL}}{2 \cdot l_{GL}} \cdot 100 \quad (\%), \tag{4}$$

where l_{Delam} is the length of a glueline opening and l_{GL} is the length of a glueline on one end grain surface. Delaminated gluelines were then closer examined by opening them with a chisel.

3.6 Microscopy

For examination of wood-adhesive bonds (paper I, II and III) and bonding surfaces (paper II), reflected light microscopy, transmitted light microscopy and scanning electron microscopy were applied. For reflected light microscopy, a Leica MZ FLIII microscope with imaging software Application Suite 4.1.0 (Leica Microsysteme Vertriebs GmbH, Wetzlar, Germany) was used. With this method, glueline thicknesses (paper I) and adhesive penetration depth (paper III) of bonded ash were determined in cross-sectional view. Transmitted light microscopy was done with a Zeiss Axiophot microscope with imaging software Axio Vision 4.8.3.0 (Carl Zeiss Microscopy GmbH, Jena, Germany). This method allows for a more detailed analysis of bonds by investigating thin slices taken from cross-sections that have been stained with safranin or methylene blue, thus providing a clearer differentiation between adhesive and wood substrate. It was used for qualitative evaluation of adhesive penetration and glueline formation (paper I) as well as for examination of the bonding surface in cross-sectional view and its influence on adhesive penetration (paper II). Scanning electron microscopy (SEM) was carried out with a Zeiss EVO 40 XVP microscope with Smart SEM V05.04.03.00 software (Carl

Zeiss Microscopy GmbH, Jena, Germany). The structure of the bonding surfaces as influenced by surface preparation methods was investigated with this method.

3.7 Surface roughness

Within the examination of bonding surfaces (paper II) a Taylor-Hobson Form Talysurf Series 2 profilometer with Ultra 4.1.7 software (Taylor Hobson, Leicester, England) was applied. Surfaces were scanned across the grain on 48 mm long measuring sections at 0.5 mm/s using a stylus tip (90° cone angle, 2 µm tip radius). From the obtained surface profiles, the roughness parameters *Ra* (arithmetical mean deviation), *Rp* (maximum peak height) and *Rv* (maximum valley depth) following ISO 4287 (1997) as well as *Rk* (core roughness depth), *Rpk* (reduced peak height) and *Rvk* (reduced valley depth) based on the material ratio curve according to ISO 13565-2 (1996) were calculated.

3.8 Strain measurement

Strain in bonded ash specimens after climate change from 20/95 to 20/40 was determined using digital image correlation (paper III). For this purpose, a stochastic speckle pattern consisting of dark speckles on a white base layer was first applied on the end grain surfaces of the specimens. The experimental setup was assembled from a specifically designed specimen mount, cold light lamps and two cameras with their optical axis (z) being orientated normal to the specimen surface (Figure 8, left). Alignment pins were used to guarantee position preservation of the specimens in x- and y-direction. Monochrome images were taken from underneath the specimens at 20/95 to document the initial situation and after 1 h, 2 h, 3 h, 4 h, 6 h, 8 h, 24 h, 72 h and 144 h after a sudden climate change to 20/40.

The two cameras recorded overall view images with an actual size of 160.6 mm x 160.6 mm and detailed views with 23 mm x 23 mm image size. With a resolution of 2048 pixel x 2048 pixel this corresponds to pixel sizes of 78.4 µm x 78.4 µm for overall view images and 11.2 µm x 11.2 µm for detailed view images. Detailed view images were located at the end of one bond line of the specimens where significant strain was anticipated. Before processing the images, the area of interest (AOI, Figure 8a and c), the *subset* value (21, size of pixel array for displacement analysis) and the *step* value (overview: 3, detail: 1, grid space) were defined. The images were cross-correlated by means of the software VIC 2D (Correlated Solutions, Inc., Columbia, SC, USA). From the image processing, displacements *u* (in x-direction) and *v* (in y-direction) were obtained so that strains ε_{xx} ,ε_{yy}, and ε_{xy} (Figure 8b and d) could be calculated.

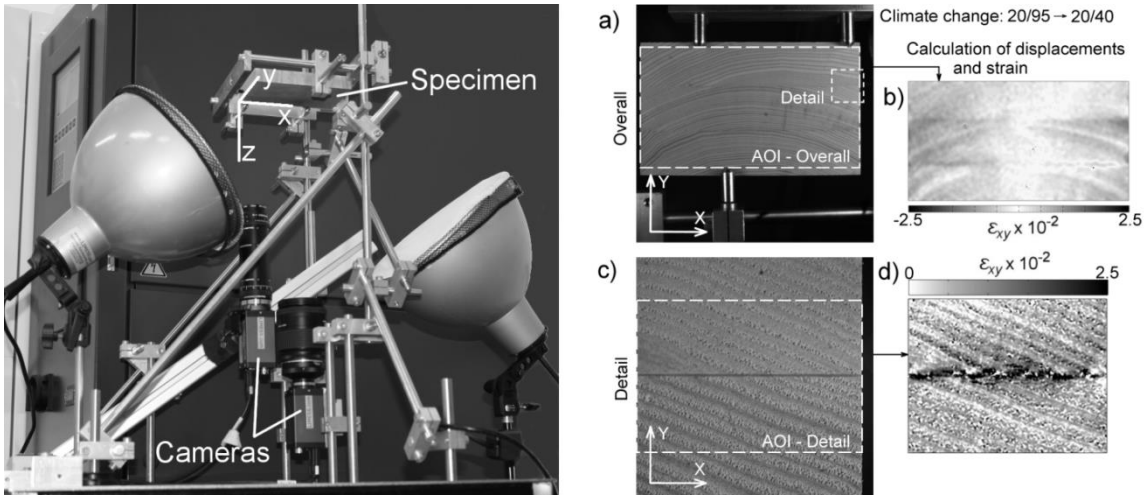


Figure 8 **Left: Test setup with specimen mount, two cameras and cold-light sources (paper III, p. 3, Figure 2). Right: OV (a) and detailed view (c) of the specimens with definition of AOIs together with strain distributions after conditioning at 20/40 (b, d) (paper III, p. 3, Figure 3)**

3.9 Data analysis

Results from paper I, II and III were analyzed with statistics software SPSS (IBM Corp., Armonk, NY, USA). A level of significance of $\alpha = 0.05$ was selected for all statistical analyses. Variance analysis was used for comparison of parametric data. Depending on variance homogeneity and sample size the post hoc tests Tamhane, Scheffe and Dunnet-C were applied. Non-parametric data (e.g., *WF*) was compared by means of the Kruskal-Wallis test with Mann-Whitney U post hoc test.

For closer investigation of moisture-induced strain (paper III), MATLAB (The Mathworks, Inc., Natick, MA, USA) was used to retrieve data from arrays obtained after cross-correlation.

4 Results and discussion

4.1 Bond formation as influenced by bonding parameters and surfaces

The selection of closed assembly time as key bonding parameter had a strong impact on glueline formation and adhesive penetration (paper I). Short closed assembly times led to deep penetration in EW vessels together with adhesive squeeze-out when applying pressure and resulted in thin gluelines with values below 20 µm (Figure 9, left). These observations can be explained with the adhesive viscosity at the time of pressure application. According to Kamke and Lee (2007) gross penetration occurs primarily with low viscosity adhesives into cell lumens that show little resistance against hydrodynamic flow of adhesive. This is the case for EW lumens in ash with their large diameter and easy accessibility in longitudinal direction. However, this also results in penetration into areas remote from the glueline which possibly does not contribute to bond performance (Hass et al. 2012). In contrast, increasing closed assembly times led to reduced EW penetration and thicker gluelines (Figure 9, center).

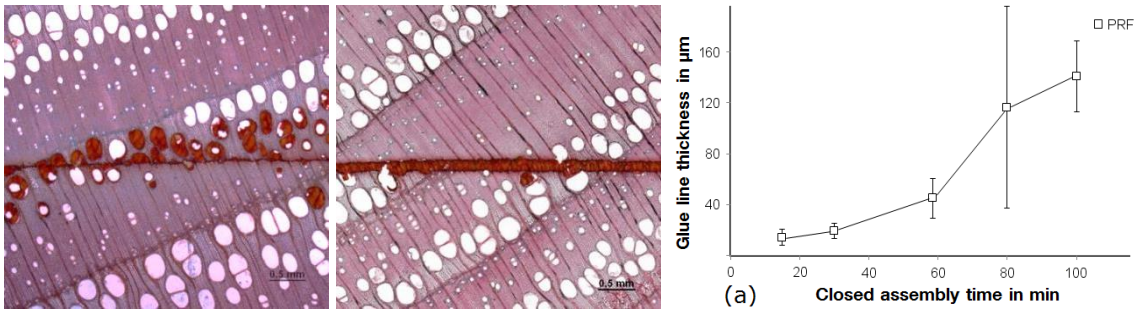


Figure 9 **Microscopic images of PRF bonded ash with closed assembly times of 30 minutes (left) and 80 minutes (center) (paper I, p. 305, Figure 4); glueline thicknesses (mean ± sd) as a function of closed assembly time (right) for PRF-bonded ash (paper I, p. 305, Figure 5a)**

Adhesive penetration in the low porosity fibers was observed only in cell layers adjacent to the glueline and seemed not to be affected by closed assembly time. The generally low penetration in fibers is not surprising since fibers in ash wood hardly provide pathways that facilitate adhesive penetration. Instead, the small lumen diameter and the low number of pits together with a low pit diameter make it difficult for the adhesive to access deeper cell layers. However, penetration into fibers is assumed to be of high importance for bond performance as they account for the major part of the adhesion surface.

It is possible that the choice of closed assembly time also affects cell wall penetration of in-situ polymerized adhesives and thus, influences bond durability for these adhesives (Frihart 2009). However, the evaluation of cell wall penetration was not possible with transmitted light microscopy within the present investigation and for ash there is no knowledge available for cell wall penetration as a function of closed assembly time or viscosity so far.

The effect of closed assembly time on glueline thickness was systematically examined by means of glueline thickness measurements. The increase of glueline thickness with longer closed assembly time was considerable for PRF, MUF-1 and MUF-2-100/50. For these adhesives, the glueline thickness approximated maximum values with increasing closed assembly time (Figure 9, right). The maximum values are very likely determined by the adhesive spread and the solid contents of the adhesives. Within the range of processing parameters as recommended by the adhesives manufacturers the increase of glueline thickness was less pronounced for EPI, PUR and MUF-2-100/20.

In paper II, three surface preparation methods have been investigated and were found to greatly influence adhesive penetration. The structure of the bonding surfaces differed considerably with respect damage of wood cells and fibrillation as shown in SEM micrographs (Figure 10a, c and e). Moreover, differences between surfaces with regard to smoothness and adhesive penetration could be observed after bonding in cross-sectional view by means of transmitted light micrographs (Figure 10b, d and f).

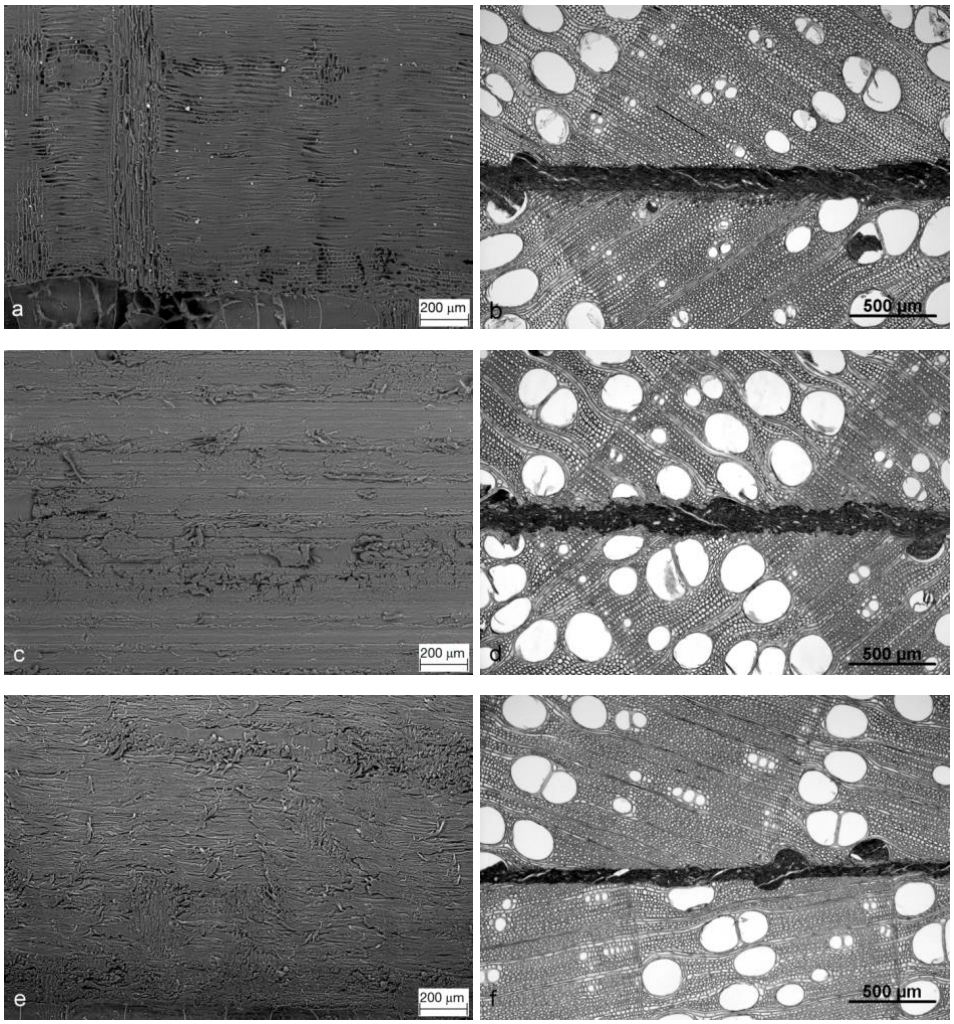


Figure 10 Planed (a, b), sanded (c, d) and face milled (e, f) surfaces as examined in topview by means of SEM images (a, c and e) and in cross-sectional view by means of transmitted light micrographs (b, d and f) after being bonded with MUF adhesive (paper II, p. 43, Figure 2)

After planing, anatomic details of ash such as vessels, fibers and parenchyma cells were easy to identify in SEM images. Cells were cut open and undamaged and, in addition, fibrillation was noticed to be very low (Figure 10a). These observations are in agreement with the roughness measurements which showed a plateau-like surface with low *Rp* and *Rpk* values (Table 10). Also, high *Rv* and *Rvk* values were determined which can be attributed to valleys caused by EW vessels of ash. In contrast, face milled surfaces showed slight cell damage and a significant level of fibrillation with fibrils being deflected in cutting direction (Figure 10e). This fibrillation is likely to improve bond performance (Stehr and Johansson 2000) and could also be detected in roughness measurements in terms of increased *Rp* and *Rpk* values. The highest degree of cell damage was found for sanded surfaces. While some anatomic components such as fibers and rays were no longer identifiable (Figure 10c) vessels were often still intact but frequently filled with abrasive dust. When comparing the surfaces, sanding produced a more uneven surface than planing and face milling. This was associated with increased *Ra* and *Rk* values and most likely, with a larger surface for adhesion.

Table 10 Roughness of planed, sanded and face milled ash surfaces (paper II, p. 44, Table 3)

Roughness parameters	Planing		Sanding		Face milling	
			$\bar{x}$ (µm) (sd (µm))			
Ra (arithmetical mean deviation)	11.8 (3.2)	A[a)]	16.2 (1.6)	B	13.1 (4.0)	A
Rp (maximum peak height)	20.1 (4.0)	A	57.6 (13.9)	B	40.6 (7.8)	C
Rv (maximum valley depth)	107.3 (13.7)	A	85.6 (13.8)	B	103.5 (23.5)	A
Rk (core roughness depth)	9.9 (2.6)	A	47.2 (4.4)	B	19.2 (3.8)	C
Rpk (reduced peak height)	4.5 (2.0)	A	18.1 (5.8)	B	15.7 (4.0)	B
Rvk (reduced valley depth)	57.3 (14.0)	A	37.3 (8.8)	B	64.1 (18.9)	A

a) Roughness values were compared by means of variance analysis and Scheffe post-hoc test using a 0.05 significance level. Different letters for surfacing methods indicate statistically significant differences in roughness.

Adhesive penetration in planed surfaces was generally between 0 and 2 cells (fibers) and therefore relatively low, although cells were cut open and thus showed good accessibility. In individual cases, penetration of up to five cell layers was found (Figure 10b). However, this was likely to be attributed to slope of grain and in addition, occurred on the adherend where adhesive was applied. Subsurface damage for planed surfaces could not be observed. This conforms to findings from de Moura and Hernandez (2006) and Murmanis et al. (1983) obtained with the wood species sugar maple (*Acer saccharum* Marsh.), Yellow-poplar (*Liriodendron tulipifera* L.) and Douglas-fir (*Pseudotsuga menziesii* (Mirb.). Face milled surfaces likewise exhibited no subsurface damage. However, the topmost cell layer showed deformed cells and detached fibrils which possibly inhibited adhesive penetration (Figure 10f). Sanding produced significant damage both in surface and subsurface cell layers (Figure 10d). It is likely that crushed cells generated a strong barrier for adhesive penetration. This layer of crushed cells possibly acts as a MWBL and thus, affects bond performance. On the other hand, sanding also produced a highly irregular surface with a large interface area which may improve bond performance.

4.2 Shear tests

The shear tests according to EN 392 (1995) exhibited little difference between shear strengths of solid ash ($f_{v,mean}$ = 14.3 MPa) and bonded ash, with mean values ranging from 13.2 MPa for PUR to 14.6 MPa for PRF and MUF-2, 100/20 (Table 11, paper I). The statistical analysis confirmed that the shear strengths for solid ash and for bonded ash samples are statistically similar. However, when comparing the bonded samples only, statistics revealed a difference between PUR bonded ash and specimens prepared with PRF, MUF-1 and MUF-2, 100/20. Furthermore, the statistical investigation of strength values as a function of closed assembly time within adhesive samples showed that this bonding parameter does not lead to different shear strengths. When comparing shear results from the present investigation with values from literature, slightly higher shear strengths can be found in paper I. For example, Brandmair et al. (2012) determined a mean shear strength of 13.3 MPa for solid ash, values between 9.1 MPa and 10.7 MPa for ash bonded with three PUR adhesives and a mean shear strength of 10.9 MPa for PRF bonded ash. Reasons for differences between studies are likely different specimen shapes, the testing device that may affect the outcome of the shear test (Steiger et al. 2010) or the natural variation of the tested wood.

The evaluation of shear planes showed high mean *WF* for EPI (90 %), PRF (96 %) and the three MUF-bonded samples (88 % - 99 %). Only the PUR bonded sample exhibited a somewhat lower mean *WF* of 63 %; the difference to the other adhesive samples was found to be statistically significant. The results are in agreement with findings from Brandmair et al. (2012) who also determined significantly lower *WF* with PUR than with PRF in combination with ash. In addition, a generally lower *WF* for PUR was also determined by Clauß (2011) when comparing PUR with MUF and PRF bonds. Clauß (2011) attributed the lower wood failure to higher deformation energy absorption and ductile failure of PUR.

Table 11 Shear strength f_v of solid and bonded ash together with *WF* as determined in a block shear test according to EN 392 (1995); specimens conditioned at 20/65 (paper I, p. 304, Table 3, units were adapted from N/mm² to MPa)

Specimen	N	Shear strength f_v (MPa)					*WF* (%)
		Mean	sd	Min	Max	CoV (%)	Mean
Solid ash timber (reference)	35	14.3	2.3	8.3	18.0	15.9	-
PRF	80	14.6	1.7	9.9	19.0	11.6	96
MUF-1	80	14.5	1.6	7.1	17.7	10.9	89
MUF-2, 100/20	30	14.6	1.4	11.8	17.3	9.6	99
MUF-2, 100/50	50	14.4	2.3	6.4	17.6	15.8	88
PUR	20	13.2	1.0	11.2	15.4	7.8	63
EPI	40	13.7	2.1	9.4	17.8	15.3	90

In paper II, the influence of surface preparation methods on the bond performance with PRF, MUF, PUR and EPI was examined in tensile shear tests. The results of the tensile shear tests are presented in Table 12. The comparison of compressive (paper I) and tensile shear results (planed surface, treatment A1, paper II) shows high strength values and *WF* for PRF-bonded ash. Although the mean tensile shear strengths of the MUF- and EPI-bonded sample were lower than the compressive shear strength, the comparably high *WF* in both tests indicates that differences in shear strength were caused by different strengths of the wood. Moreover, in both investigations the *WF* of PUR-bonded ash was significantly lower than with other adhesives. This leads to the overall conclusion that the results of both investigations are in good agreement.

Table 12 Shear results $f_{v,t}$ and *WF* for ash bonds as influenced by surfacing method, adhesive and treatment according to EN 302-1 (2004) (paper II, p. 45, Table 4)

Treat-ment	Surface	Adhesive	Shear strength $f_{v,t,mean}$ (MPa) (CoV (%))			WF_{mean} (%)		
A1	Planing	PRF	14.53 (9.0)	A[a)]	a[b)]	89	A[a)]	ab[b)]
		MUF	11.10 (18.7)	B	a	92	A	b
		PUR	11.84 (10.2)	B	a	33	B	c
		EPI	11.76 (21.0)	B	ab	88	A	a
	Sanding	PRF	12.61 (13.9)	A	b	87	AB	b
		MUF	11.16 (15.9)	BC	a	93	A	b
		PUR	12.27 (15.6)	AB	a	62	C	b
		EPI	10.85 (11.3)	C	b	85	BC	a
	Face milling	PRF	13.60 (13.0)	A	ab	94	AB	a
		MUF	11.52 (10.6)	B	a	98	A	a
		PUR	12.74 (11.7)	AB	a	94	A	a
		EPI	12.74 (21.0)	AB	a	89	B	a
A4	Planing	PRF	7.18 (24.9)	A	a	90	A	a
		MUF	4.99 (11.5)	B	c	12	B	b
		PUR	5.61 (12.7)	B	a	5	C	c
		EPI	5.46 (22.1)	B	a	16	BC	b
	Sanding	PRF	7.01 (11.4)	A	a	92	A	a
		MUF	6.29 (20.2)	AB	a	50	B	a
		PUR	5.98 (13.2)	B	a	40	B	a
		EPI	5.15 (20.4)	C	a	24	C	a
	Face milling	PRF	7.34 (23.2)	A	a	96	A	a
		MUF	5.52 (11.6)	B	b	64	B	a
		PUR	5.86 (15.3)	B	a	29	C	b
		EPI	5.62 (17.1)	B	a	37	C	a

Shear strength values were compared by means of variance analysis and Scheffe post-hoc test. *WF* values did not follow a normal distribution for which reason the non-parametric Kruskal-Wallis test and Mann-Whitney U post-hoc test were used. The level of significance was set at 0.05 for all statistical tests.

a) Uppercase letters were used for comparison of adhesive performance within one treatment and surfacing group.

b) Lowercase letters were used for comparison of surfacing methods within one treatment and adhesive group.

Different letters indicate statistically significant differences in shear strength or *WF*.

The tensile shear tests according to EN 302-1 (2004) as documented in paper II showed good performance of ash bonds when tested in dry condition (treatment A1). Differences between adhesives and surfaces however could be observed after treatment A4. For example, the reference value[3] of $f_{v,t,mean} \geq 10$ MPa after treatment A1 as specified in relevant standards (EN 15425:2008; EN 301:2013; EN 16254:2013) for bonds made with beech (*Fagus sylvatica* L.) was reached by all four adhesives in combination with all three surfacing methods. For PRF, MUF and EPI, the high strength values in combination with high mean *WF* values (≥ 85 %) suggest a good quality of the ash bonds. Only PUR bonded ash showed varying *WF* with the different bonding surfaces (WF_{planed} = 33 %, $WF_{face\ milled}$ = 94 %), thus indicating a special importance of fibrillation on the bonding surface for PUR bonds. In contrast, the reference shear strength of $f_{v,t,mean} \geq 6$ MPa after treatment A4 was only obtained with PRF-bonds and with sanded surfaces bonded with MUF. The good bond quality of PRF-bonds for all surfaces after A4 is supported by high mean *WFs* ≥ 90 %, and reflects the well-known high moisture resistance of this adhesive type (e.g., Dinwoodie 1983; Frihart 2009). Ash bonds made with MUF, EPI and PUR on the contrary showed a more significant decrease in shear strength and, in particular, in *WF* which indicates significant weakening of the bonds by the A4 moisture treatment. The high adhesion failure for these adhesives strongly depended on the surface preparation method, with the lowest *WF* being determined for planed surfaces. Sanding and face milling produced statistically significant improvements in terms of *WF* which may be due to a larger surface available for adhesion or an enhanced contact between wood and adhesive caused by fibrillation.

No correlation between surface roughness values and tensile shear results could be found, most likely because subsurface damage as determined with microscopy on sanded surfaces could not be quantified in terms of roughness parameters. However, when comparing planed and face milled surfaces with limited subsurface damage, *Rk* and *Rpk* values which represent the processing roughness and fibrillation (Westkämper and Schadoffsky 1995) may be useful to characterize surfaces. For example, face milled surfaces showed increased *Rk* and *Rpk* values (Table 10) and, at the same time, a tendency towards higher shear strength and *WF* for all four adhesives (Table 12).

Overall, it can be concluded from the tensile shear tests that differences between surfaces and adhesives after A1 were minor (except for *WF* of PUR bonds). Moisture treatment A4 however revealed significant differences between adhesives and surface preparation methods, respectively, in particular for the *WF* results. According to the results of the tensile shear test, the surface preparation method, which is most promising to produce moisture resistant bonds, seems to be face milling.

[3] Minimum strength values given in EN 15425 (2008), EN 301 (2013) and EN 16254 (2013) are only applicable for bonds being prepared with beech wood (*Fagus sylvatica* L.). However, for the present investigation with ash, values specified for beech were taken as reference values.

4.3 Delamination test

The resistance to delamination of bonded ash (paper I) was significantly different between adhesives. In addition, the closed assembly time very clearly affected the delamination behavior (Figure 11). The highest resistance to delamination was found with PRF adhesive with mean values between 38.3 % (15 min) and 4.2 % (100 min). These results are in agreement with the shear test results where the highest resistance against moisture exposure was also determined with PRF adhesive. Delamination values for the MUF-bonded samples were on average higher than for PRF and strongly varied between the MUF-1 and MUF-2 as well as between mixing ratios for MUF-2 (Figure 11b). The lowest resistance to delamination was determined with PUR- and EPI-bonded ash. One reason for differences between MUF adhesives is very likely variations between adhesive formulations. In addition, different resin-hardener ratios imply different polyvinylacetate (PVAc) and formaldehyde percentages. This may affect wetting behavior of the adhesive (Scheikl and Dunky 1996) and the development of adhesion forces as well as the material properties of the cured adhesive.

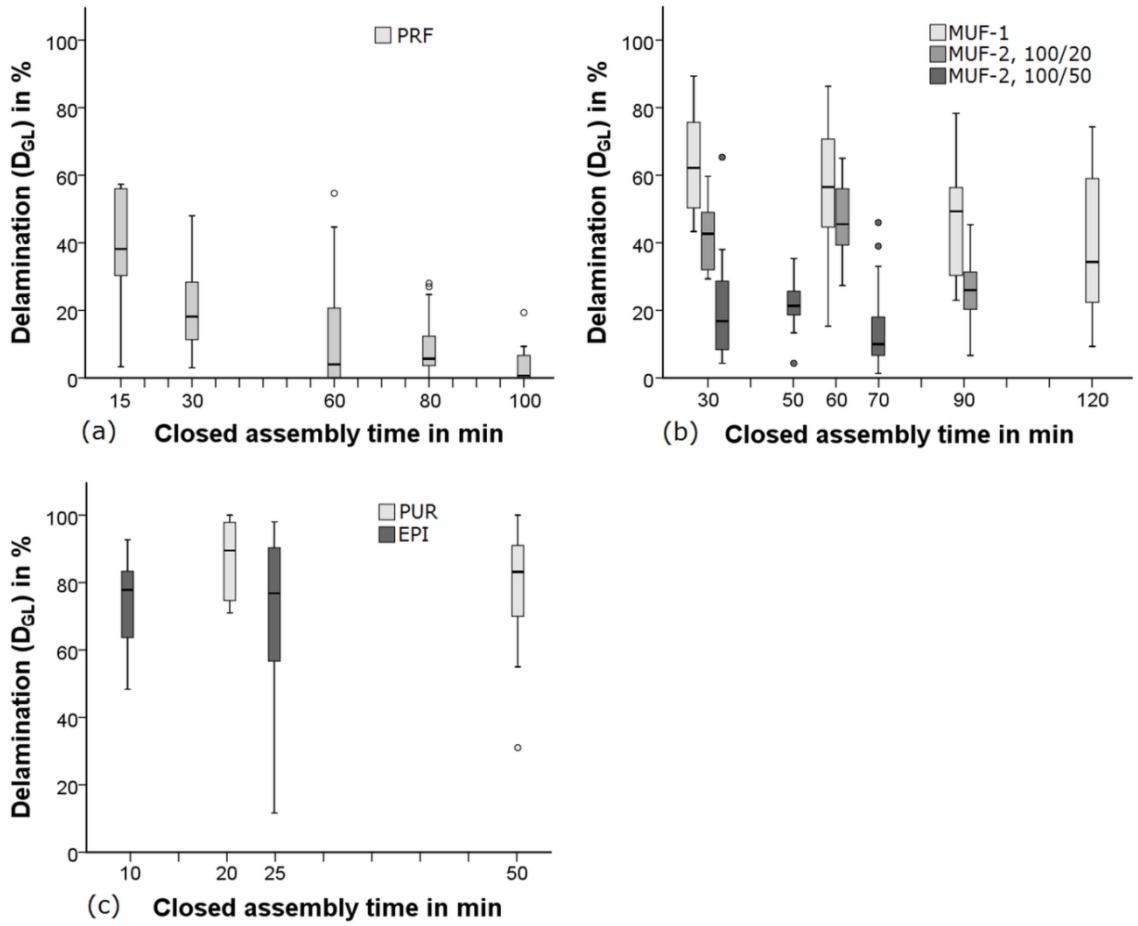


Figure 11 Resistance to delamination (D_{GL}) for ash bonded with PRF (a), MUF (b), and PUR and EPI (c) as a function of closed assembly time (paper I, p. 303, Figure 2)

Boxplot elements: box: values between the 25th and 75th percentile (interquartile range), horizontal line: median, whiskers: lowest/highest values, circles: outliers beyond 1.5 times the interquartile range

For PRF and MUF, the effect of closed assembly time on resistance to delamination was evident. Although only two closed assembly times were tested with PUR and EPI, a trend towards lower delamination values with longer closed assembly time was observed. The results with MUF confirm the findings from Schmidt et al. (2010) who determined higher resistance to delamination with longer closed assembly times for MUF-bonded beech. Delamination values with ash were however significantly higher than with beech. Reasons for the higher delamination values of bonded ash may be differences between ash and beech in anatomy or wood chemistry. However, the investigation of causes would require more research.

After the tests, the delaminated gluelines were opened with a chisel to assess glueline openings not only on the end-grain surfaces of the specimens but also at the bonding surface. The evaluation showed frequent adhesion failure in the wood-adhesive interface predominantly in the LW areas of the bonding surface and reflects the low penetration of adhesive in these areas (Figure 9). This failure pattern was observed for all adhesives.

It was realized that both the glueline thickness and the delamination behavior depend on the closed assembly time. For this reason, the glueline thickness and the resistance to delamination were analyzed in relation to one another and the resistance to delamination was found to be higher with increasing glueline thickness as shown in Figure 12 for PRF-bonded ash. This trend was observed for all adhesives.

Also, the increasing glueline thickness was associated with lower penetration in EW vessels (Figure 9). Although penetration behavior of adhesives is considered to be important for bond performance (Kamke and Lee 2007), the results indicate that adhesive penetration in EW vessels of ash is insignificant for the delamination resistance. Instead, the glueline thickness and associated with this the altered conditions for the stress distribution in case of moisture change are most likely more important. An improved penetration of adhesive into fibers, in particular in LW areas, would possibly contribute to an enhanced resistance to delamination. However, the anatomic characteristics of fibers (small lumens, low number and small diameter of pits) obviously prevent deeper adhesive penetration.

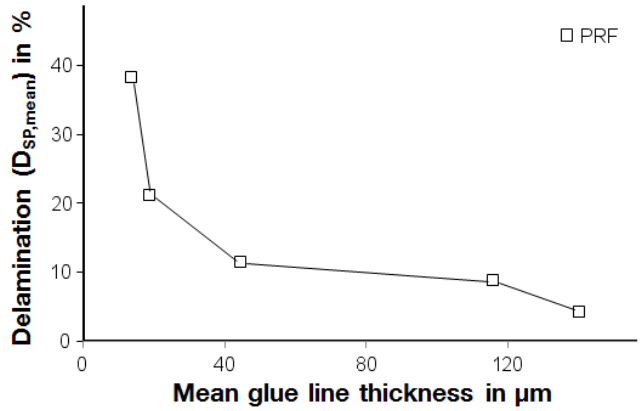


Figure 12 Resistance to delamination ($D_{SP,mean}$) of PRF-bonded ash as a function of glueline thickness (paper I, p. 306, Figure 6a)

The results from block shear and delamination tests obtained in paper I were compared to each other. As no correlation between shear strength or *WF* and delamination values was found, the shear test is evaluated to be of limited value for the assessment of bond durability. Instead, the shear test seems only suitable to identify local defects such as insufficient adhesive application. These results are in agreement with previous studies (Ohnesorge et al. 2010; Schmidt et al. 2010) on beech wood.

Normative requirements for resistance to delamination are given in dependence of the adhesive. The evaluation of delamination values in paper I and II was for instance made for PRF and MUF on basis of EN 301 (2006). This standard specifies maximum delamination values of 5 % for adhesive type I in the HT procedure and 10 % for adhesive type II in the LT procedure irrespective of the wood species. These values shall not be exceeded by any specimen. Taking the 5% limit value as pass/fail criterion for type I adhesives into account, no combination of adhesive, closed assembly time and surfacing method fulfilled the standard requirement in combination with ash wood. The latest edition of the standard (EN 301:2013) incorporated recent developments and specified separate, higher delamination limit values for oak (adhesive type I: 8 %; adhesive type II: 12 %) although no scientific basis had been established for such an adjustment. However, taking the 8 % for adhesive type I as a basis also for ash, the PRF-bonds with 100 min closed assembly time ($D_{SP,max}$ = 7,1 %, paper I) as well as sanded ($D_{SP,max}$ = 6.8 %, paper II) and face milled surfaces bonded with PRF ($D_{SP,max}$ = 6.1 %, paper II) would meet the requirements. As can be expected because of less severe test parameters and the lower number of cycles, the delamination was lower after the LT test procedure. The 12 %-requirement for adhesive type II as shown in paper II would be fulfilled by all three surfaces bonded with PRF (Planing: $D_{SP,max}$ = 4.4 %; Sanding: $D_{SP,max}$ = 2.9 %, Face milling: $D_{SP,max}$ = 4.7 %), the MUF-bonded sanded surface ($D_{SP,max}$ = 4.5 %) as well as EPI-bonded ash after face milling ($D_{SP,max}$ = 11.6 %).

The differences between adhesive types in delamination resistance are similar in paper I and in paper II for planed surfaces. However, significant differences in delamination behavior were determined in paper II depending on the surfacing method (Figure 13). Although planing is the surfacing method that is most frequently used in industry, planed surfaces showed on average the lowest resistance to delamination. Except for EPI-bonded ash, sanded surfaces provided a higher resistance to delamination than planed surfaces. On the one hand, this may be due to an increased surface available for adhesion. On the other hand, sanded surfaces showed a layer of damaged cells in subsurface areas which may have acted as a buffer zone at moisture change and contributed to a more homogeneous strain distribution. Face milled surfaces produced a higher resistance to delamination with all four adhesives than planing. As planed and face milled surfaces primarily differ in the level of fibrillation, it is very likely that fibrils strengthen the bond as suggested by Stehr and Johansson (2000).

While PRF and MUF showed the best performance with sanded surfaces (Figure 13), EPI- and PUR-bonded ash provided the highest resistance to delamination with face milled surfaces. This leads to the conclusion that the buffering effect of damaged cells with sanded surfaces as well as the fortifying effect of fibrils with face milled surfaces depends on the adhesive type. In this context, the classification into in-situ polymerized (PRF, MUF) and pre-polymerized (PUR, EPI) adhesives according to Frihart (2009)

helps to interpret differences in strain distribution with adhesive type and, as a consequence, the delamination results. For example, PRF and MUF penetrate into cell walls before hardening and develop a rigid interphase and adhesive layers. In case of moisture change, deformations primarily occur beyond the interphase zone in the bulk wood. Accordingly, the subsurface layer of damaged cells with sanded surfaces gains importance for strain distribution and leads to higher resistance to delamination as high shear stresses may be avoided. In addition, the fortifying effect of fibrils may be less effective due to the rigidity of the cured PRF and MUF adhesive than for pre-polymerized adhesives. In contrast, PUR and EPI are not able to diffuse into cell walls because of their larger molecule size and produce gluelines with high flexibility. Due to the adhesive elasticity, stress as a result of moisture change is mainly distributed in the glueline. As a consequence, the bond strengthening due to fibrils with face milled surfaces may be more pronounced with PUR and EPI and thus, increase the resistance to delamination for these adhesives. Further improvements in delamination resistance of PUR bonds may be realized when the limited adhesion under wet condition as investigated by Kläusler (2014) can be enhanced.

When evaluating the delamination results with regard to surface roughness, similar trends as for shear results could be found. When comparing planed and face milled surfaces, a trend towards higher resistance to delamination with increased *Rk* and *Rpk* values was observed. However, with sanded surfaces the roughness was of limited value as subsurface damage could not be evaluated. Nevertheless, sanded surfaces led to high resistance to delamination with PRF and MUF for which reason it was concluded that subsurface damage positively influences bond durability with these adhesives, possibly because of less stress peaks.

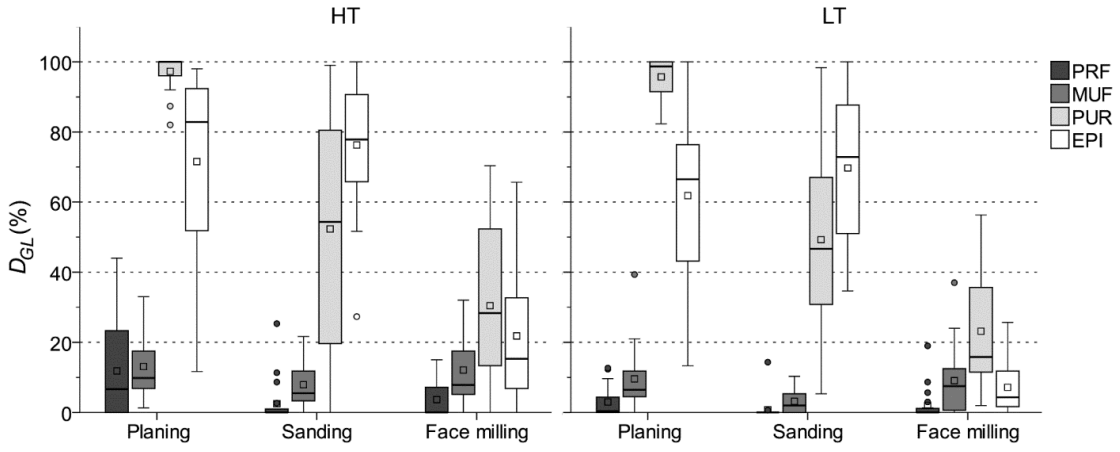


Figure 13 Resistance to delamination (D_{GL}) of bonded ash depending on surfacing method, adhesive and test procedure according to EN 302-2 (2004) (paper II, p. 47, Figure 4). Boxplot elements are explained in Figure 11. In addition, Figure 13 contains squares which represent the arithmetic mean.

4.4 Strain measurements

The deformation behavior of bonded ash assemblies at moisture change was investigated in paper III. Strains were analyzed after the specimens reached equilibrium moisture content (EMC) at 20/40 and are presented in the following. For the evaluation of results, emphasis is placed on shear strain ε_{xy} as the shearing load was expected to be the main load in the bond because of the specimen design.

The analysis of the overall view data (Figure 8a) showed a homogeneous strain distribution in x-direction (ε_{xx}, Figure 14a). In the y-direction strain maps (ε_{yy}, Figure 14b), the positions of annual growth rings and the gluelines were visible. The shear strain distribution ε_{xy} is displayed in Figure 14c. In the center area of the specimens, no or very little shear strain as characterized by a white or greyish zone was found. Towards the specimen sides in x-direction, the shear strain increased both in the wood and, in particular, in the glueline area.

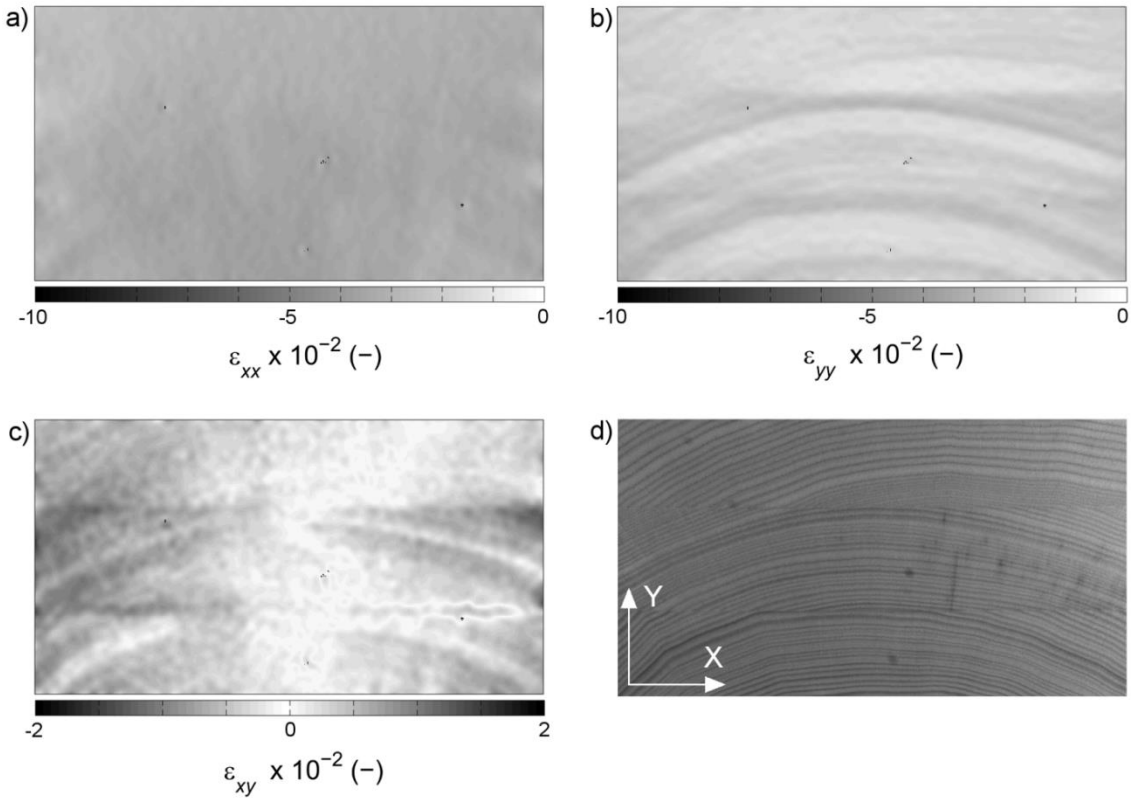


Figure 14 Overall view strain maps ε_{xx} (a), ε_{yy} (b) and ε_{xy} (c) for an EPI-bonded sample together with an illustration of the test specimen surface before test (d) (paper III, p. 5, Figure 5)

The analysis of ε_{xx} and ε_{yy} data obtained from detailed view (Figure 8c) corresponded well with the overall view results. Furthermore, ε_{xx} and ε_{yy} values for the wood surface (with gluelines excluded) were converted into tangential and radial strain (ε_t and ε_r) by means of coordinate transformation. ε_t and ε_r as derived from experimental data showed good agreement with shrinkage values from literature in tangential and radial direction.

The shear strains in glueline areas were analyzed only in detailed view due to the higher resolution of the data. As can be seen in Figure 15, the ε_{xy} values depended on adhesive type and glueline thickness. Although mean values showed a considerable range between 0.65 x 10^{-3} for PUR and 9.3 x 10^{-3} for MUF GL0.01, the evaluation by means of variance analysis exhibited no statistical difference between samples. For comparison, the shear strain was analyzed for the wood surfaces exhibiting a mean value of 6.03 x 10^{-3}. Except for PRF, which showed a similar strain level as wood, strain values for GL0.1 with PUR, MUF and EPI and MUF GL0.2 were significantly lower than for wood. Moreover, the comparison of glueline thicknesses of MUF bonds showed significantly higher shear strain with GL0.01 than with GL0.1 and GL0.2, thus indicating a significant influence of the GL.

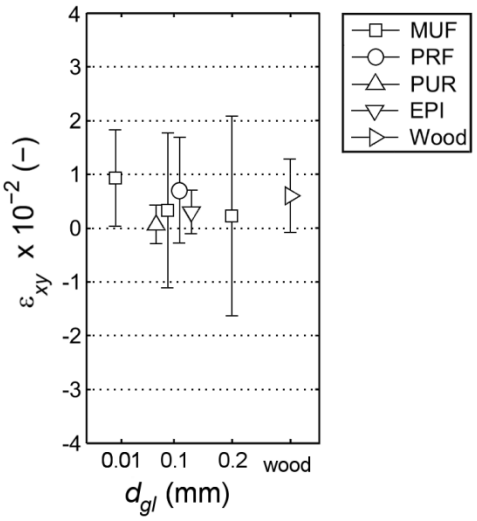


Figure 15 Shear strains ε_{xy} (mean with error bars showing the sd) in the gluelines in dependence on adhesive and glueline thickness and in wood (paper III, p. 6, Figure 6)

For MUF and PRF bonds, the sd of ε_{xy} was distinctly higher than for EPI or PUR (Figure 15). This may on the one hand be attributed to the occurrence of local strain maxima because of the high stiffness of MUF and PRF. Such localized strains were also determined by Serrano and Enquist (2005) for brittle PRF when investigating strain in shear specimens with different adhesives. A further reason for the increased sd of MUF GL0.1 and GL0.2 bonds are probably cracks that were found in the gluelines, possibly due to restrained shrinkage of the adhesive layer. This finding is considered to be in agreement with results from Hass et al. (2012) who reported about cracks in UF gluelines although the cracks were less numerous in the MUF bonds.

In Figure 16, ε_{xy} strain distributions that were calculated as average values across the detailed view AOI are displayed. As PRF and MUF as well as PUR and EPI showed similar deformation characteristics, only shear strain distributions of PUR and MUF as well as MUF with different glueline thicknesses are presented in Figure 16. As can be seen from Figure 16, the strain distribution in the wood showed stable values. When approaching the glueline, shear strain in some cases changed significantly which leads to the conclusion that the bond highly affects the shear strain distribution.

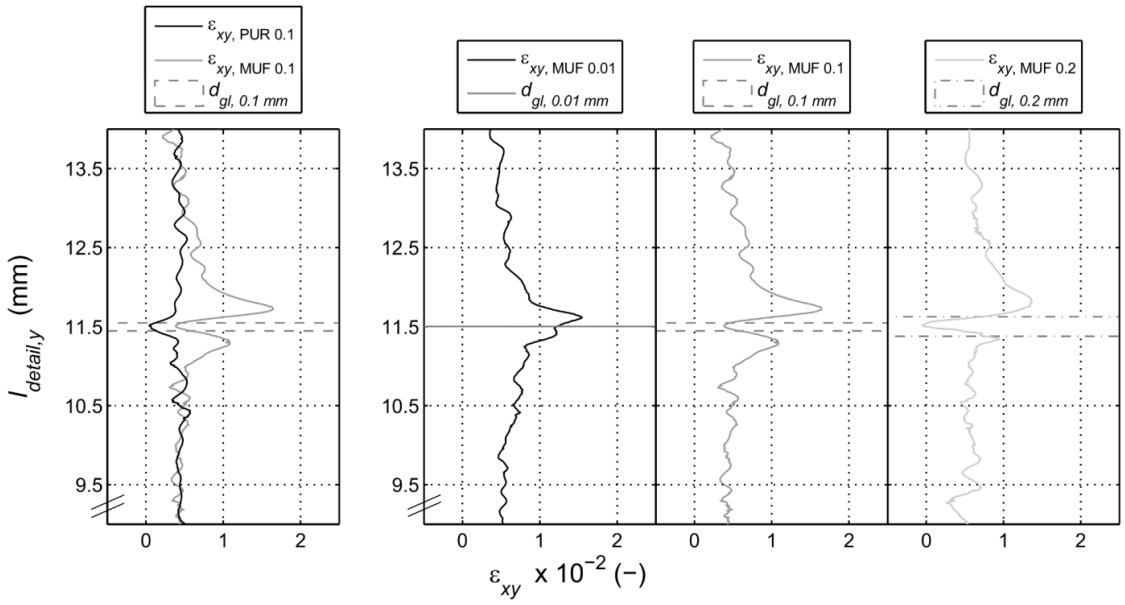


Figure 16 Left: Shear strain distributions ε_{xy} with 0.1 mm glueline for MUF and PUR. Right: Distributions for ε_{xy} in MUF-bonded joints with 0.01 mm, 0.1 mm and 0.2 mm glueline thicknesses (d_{gl}) (paper III, p. 7, Figure 7)

The comparison of ε_{xy} strain distributions for PUR and MUF with 0.1 mm glueline thickness shows significant differences (Figure 16, left). While shear strain for PUR decreases in close proximity to the glueline, strain for MUF starts to increase in some distance to the glueline and develops high values close to the glueline. Therefore, the strain decrease in the glueline is more pronounced for MUF, although the lower value in glueline center however was found for PUR. This behavior is most likely attributed to different elasticities and penetration characteristics of adhesives as also described by Frihart (2009). For example, strains as a result of moisture change are mainly distributed in the wood for in-situ polymerized adhesives such as MUF as their stiffness is significantly higher than that of ash wood in radial or tangential direction. In contrast, the flexibility of PUR (Table 5) which is similar or lower than ash wood prevents high strains and as a consequence, the occurrence of stress concentrations.

The distributions with MUF GL0.01, GL0.1, and GL0.2 show a steady increase in shear strain when approaching the glueline (Figure 16, right). While the strain transition across the glueline appears to be direct with GL0.01, a significant drop of ε_{xy} close to the glueline with very low shear strain in the glueline center can be observed both for GL0.1

and GL0.2. The strain decrease with GL0.2 moreover seems to be more pronounced than with GL0.1 and, in addition, the minimum value in GL0.2 is lower than in GL0.1. As a consequence, it can be assumed for the MUF bonds that the thicker the glueline the higher the strain reduction in GL center. These differences in strain transition between glueline thicknesses most likely correspond to varying stress levels in the wood-adhesive interface. Although stresses are difficult to quantify for example due to relaxation or plastic deformation, these findings possibly explain the correlation between glueline thickness and moisture-related durability as determined in paper I.

For a better understanding of strain distribution in case of moisture change, the distances $s_{def,1}$ and $s_{def,2}$ on both sides of the glueline were determined within which the strain level deviated from the strain in the wood (Figure 17). These areas were considered to be affected by the bond. The s_{def} values were then analyzed by calculating mean values and sd (Figure 18).

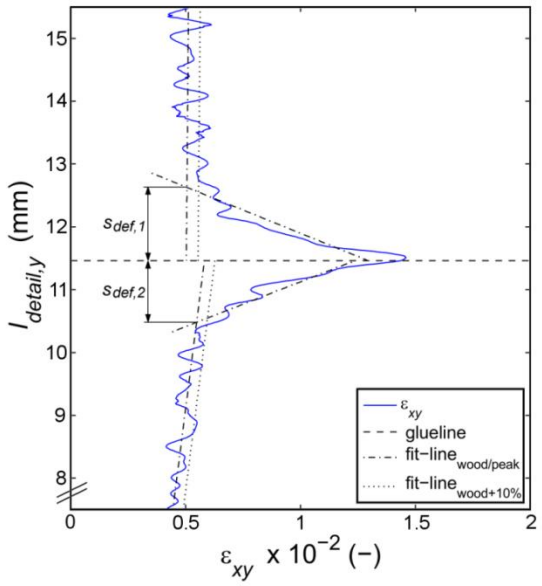


Figure 17 Determination of the distances $s_{def,1}$ and $s_{def,2}$ where increased strain occurred adjacent to the glueline shown for a MUF GL0.01 specimen (paper III, p. 8, Figure 8)

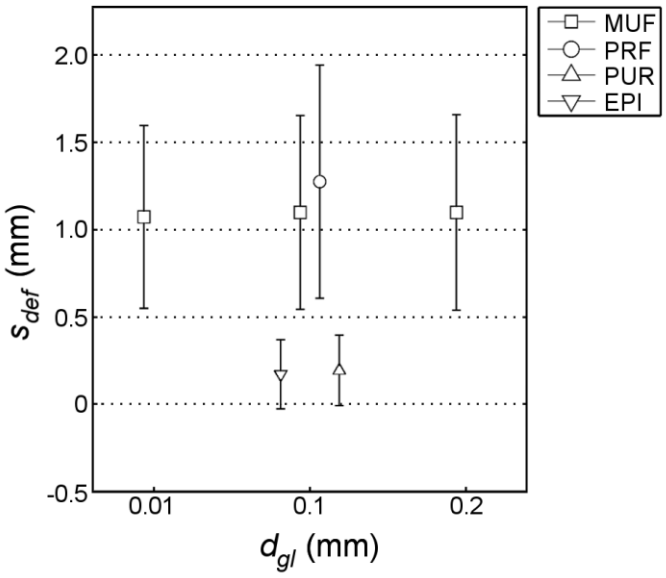


Figure 18 ***s_{def}*** **(mean with error bars showing the sd) for different adhesives and glueline thicknesses** ***d_{gl}*** **(paper III, p. 9, Figure 9)**

The evaluation of GL0.1 data showed that adhesives can be divided into two groups: PRF ($s_{def,mean}$ = 1.28 mm) and MUF ($s_{def,mean}$ = 1.1 mm) were statistically similar just as well as PUR ($s_{def,mean}$ = 0.19 mm) and EPI ($s_{def,mean}$ = 0.17 mm). These results correspond to previous findings and can be interpreted with different mechanical properties of adhesives. While strains in PRF- and MUF-bonded ash are primarily distributed in the wood due to the high rigidity of the adhesive, only little strain can be found in PUR- and EPI-bonded wood.

As strain distributions can be significantly influenced by adhesive penetration (Gindl et al. 2005), this was further analyzed. While penetration in fibers was very little for all adhesives, the maximum penetration depth in ash EW vessels varied. For GL0.1, the maximum penetration depth ranged from 132 µm (MUF) to 841 µm (PRF), while EPI and PUR showed penetration depths of 360 µm and 353 µm. The highest penetration depth for MUF was found with GL0.01 with 706 µm and is probably attributed to the shorter closed assembly time used for preparation of the GL 0.01 specimens. Despite of a clear variation in penetration depth with MUF and PRF, no significant differences between s_{def} values were determined for these adhesives. This leads to the conclusion that the strain distribution is independent from the penetration in EW vessels. In addition, $s_{def,mean}$ exceeded the penetration depths, i.e., the deformation reaches deeper into the wood than what can be explained by penetration of rigid adhesives. In contrast, penetration for PUR and EPI exceeds s_{def} values. These results suggest that the penetration in EW has little influence on strain distribution for PUR and EPI. In addition, the strain transition is probably smoother due to similar elasticities of wood and adhesives.

5 Synthesis

The investigations presented in this thesis cover the following aspects with regard to gluability of ash wood for load-bearing purposes: the evaluation of the suitability of adhesives for structural bonding of ash, the optimization of bond performance in ash assemblies by means of different bonding parameters and bonding surfaces as well as the achievement of a better understanding of the effects of moisture change in adhesively bonded ash.

It could be shown that the performance evaluation of adhesively bonded ash by means of the block shear test according to EN 392 (1995) gives limited information. This is due to the fact that this test method is carried out with test specimens after climate conditioning at 20/65. Frequent moisture changes that usually have to be tolerated by structurally bonded wood products in applications are not taken into consideration. From the generally high shear strength and wood failure it may appear that well-performing wood-adhesive bonds can be produced with ash wood. However, this test method was not suitable to identify differences in performance between adhesive types or bonding parameters as could be determined after moisture changes. As a consequence, the block shear test is considered insufficient to evaluate the suitability of adhesives for long term applications.

In general, it proved to be difficult to produce ash bonds with high moisture-related durability. Adhesive types that are regarded to be equally well suited for structural bonding of softwood showed considerably different behavior in combination with ash wood when subjected to severe moisture changes in delamination tests. The adhesive type that seems most appropriate to produce durable structural bonds with ash is PRF. MUF, EPI and PUR are less suitable to withstand stresses caused by moisture changes in ash assemblies (with planed bonding surfaces). The closed assembly time was identified as a key bonding parameter. A prolongation of closed assembly time leads to an increased glueline thickness, and thicker gluelines are associated with higher moisture-related durability of ash bonds. Furthermore, a prolonged closed assembly time prevents excessive EW penetration and adhesive squeeze-out and thus, adhesive can be used more efficiently. In addition, it could be shown that a thick glueline causes higher benefit with regard to moisture-related durability of bonded ash than deep adhesive penetration in EW.

The surfacing methods planing, sanding and face milling produce clearly distinctive bonding surfaces. The surfaces primarily differ in the level of fibrillation and the degree of cell damage and can be described by specific roughness values. The different surface characteristics strongly influence the performance of ash bonds. Differences in performance between surfaces and adhesive types can however hardly be observed in tensile shear tests (EN 302-1:2004) on dry test specimens which confirms conclusions regarding the limited significance of the block shear test. In contrast, boiling treatment of specimens prior to tensile shear tests reveals differences between surfaces and adhesive types, in particular with regard to wood failure. These differences are shown to be even more pronounced in delamination tests.

In general, face milled surfaces are associated with good bond performance, whereas planed surfaces which are most commonly used in glulam industry provide the lowest bond performance. The level of fibrillation can be described well by means of roughness values and can be considered an indicator for bond performance with planed and face milled surfaces. Increasing fibrillation enhances the moisture-related durability of ash bonds for all adhesive types. This optimization effect is more pronounced with PUR and EPI than with MUF and PRF and can be explained with a reinforcement of the glueline by fibrils together with the distribution of moisture-induced strain predominantly in the glueline due to a lower elasticity of PUR and EPI gluelines. The validity of roughness values is limited when crushed cells on the surface as well as subsurface damages as observed with sanding occur. These damages affect the bond performance, effects are however inhomogeneous for adhesive types: It could be shown that MUF and PRF provide a more pronounced optimization effect with sanded than with face milled surfaces than PUR and EPI. This can be attributed to the strain distribution mainly in the wood due to the high rigidity of MUF and PRF gluelines.

One limitation of the investigation with different bonding surfaces is that results refer to a momentary condition of surface preparation. As surfacing tools, i.e., blades and sanding belt, are subject to continuous wear the characteristics of bonding surfaces change with the condition of the tool. A question that could be approached in future research investigations is which the optimum level of fibrillation would be and how this level of fibrillation can be produced. Furthermore, to better predict bond performance with sanded surfaces a comprehensive characterization of surfaces that, in addition to the visible surface, also allows for assessing the condition of wood within a depth of a few cell layers from the surface would be required.

The resistance to delamination (EN 302-2:2013) of ash assemblies is the decisive property for the practical applicability of ash glulam in Germany with regard to building regulations. During this thesis, adjustments have been made in standardization works in terms of delamination boundary values for bonding of oak. Taking these adjusted boundary values as a basis also for ash bondings, a re-evaluation of the delamination results shows that PRF adhesive in combination with specific bonding parameters fulfills requirements for adhesive type I (EN 301:2013) and thus, a technical approval of ash glulam could be achieved. In addition, the results would basically allow for indoor application of ash glulam when using sanded surfaces in combination with MUF or face milled surfaces with EPI in European countries, where the use of adhesive type II is permitted.

It has to be taken into account that the validity of the results is limited to the particular adhesive products and only trends can be derived for the chemical group of adhesives. This is due to the fact that - despite of belonging to the same chemical group – formulations of commercially available adhesive vary between manufacturers and therefore may provide different moisture-related durability of ash bonds.

The deformation behavior of bonded ash assemblies caused by moisture change was investigated. It could be shown that deformation in and adjacent to the glueline is greatly influenced by the adhesive type, i.e., by the elasticity and stiffness of the adhesive

layers. The classification of adhesives into the two groups pre-polymerized (PUR, EPI) and in-situ polymerized (PRF, MUF) as suggested in a theoretical model by Frihart (2009) can be confirmed with regard to deformation behavior of wood-adhesive bonds in ash assemblies. The low stiffness of PUR and EPI together with nonexistent cell wall penetration leads to a generally low shear strain level. The difference in shear strain between glueline and the adjacent ash wood is small which suggests low shear stress in the wood-adhesive interface. In contrast, PRF and MUF penetrate into cell walls and when hardening, the adhesives stabilize cell walls and produce highly rigid gluelines. This leads to shear strain being mainly distributed in the wood and to significant differences between strain in the glueline and the adjoining wood. Due to the considerable variation of strain values it can be assumed that higher shear stresses develop in wood-adhesive bonds with PRF and MUF. Moreover, it could be shown that gross penetration of all adhesives in ash EW is insignificant for shear strain distribution and therefore, is assumed to have little impact on moisture-related bond durability.

The tests also reveal that the shear strain distribution of MUF-bonded ash varies with the glueline thickness. Shear strain with thin gluelines is directly transferred across the glueline and shows a high level of interfacial strain. In contrast, thicker gluelines show low strain values in the glueline center due to the high adhesive stiffness with strain already decreasing in the wood adjoining the glueline. This leads to lower interfacial strain in the wood-adhesive interface. The results indicate higher interfacial stress for thin gluelines and a buffering effect of thicker gluelines. Also, this can explain the lower moisture-related durability of ash bonds with thin gluelines as determined in delamination tests.

It has to be taken into account that one limitation of the investigation of deformation behavior is the resolution of the images and the size of the photographed surface, respectively. In future research studies, smaller pixel sizes would therefore allow for more detailed analyses for example of load transfer in thin gluelines, starting points of bond failure or the influence of EW and LW properties on stress occurrences. Also, a closer analysis of bonds would help for a further improvement of understanding of bond durability, e.g., with regard to stress transfer with different surfacing methods or the occurrence of stress concentrations.

The investigations generally help for a better understanding of load transfer in wood-adhesive bonds. Although shear strain distributions suggest that PUR and EPI are more suitable for ash bonding it has to be taken into account that the moisture resistance of these adhesives is lower than for PRF and MUF as could be shown in delamination tests. Future research subjects therefore should include how the resistance of PUR and EPI bonds against moisture change can be influenced, for example by means of a primer application. The gained knowledge could also be used for new adhesive developments for hardwood bonding, e.g., by adjusting material properties of PRF and MUF towards higher elasticity to avoid stress concentrations. In addition, the findings could contribute to modeling of adhesively bonded wood assemblies.

The practical implementation of ash glulam has not yet taken place in Germany. However, it is considered worth the effort to further develop ash glulam including the quality of adhesively bonded joints. The use of ash glulam would allow for developing

new fields of application due to its enhanced strength and stiffness properties. Also, this would promote a sustainable use of the natural resource ash wood.

6 Publications

6.1 Structural bonding of ash (Fraxinus excelsior L.): resistance to delamination and performance in shearing tests

“The utilization of ash (*Fraxinus excelsior* L.) allows for a significant enhancement of the load-bearing capacity in structural laminated products. However, such applications fundamentally require high-strength and durable bonds between lamellas and in finger joints. Therefore, the aim of the present survey was to evaluate ash bondings in terms of resistance to delamination (EN 302-2) and shear performance (EN 392). Investigations were performed with five adhesives: phenol-resorcinol-formaldehyde (PRF), melamine-urea-formaldehyde (MUF-1, MUF-2), polyurethane (PUR), emulsion polymer isocyanate (EPI) and varying closed assembly times as key bonding parameter. For all tested adhesives and closed assembly times, the shear test showed high wood failure percentages and bond strength values that compare to solid ash. In contrast, for resistance to delamination, significant differences were found between the adhesives as well as between closed assembly times, with improving resistance to delamination for increasing closed assembly times. The best performance was determined for the PRF-adhesive and long closed assembly times. However, standard requirements for resistance to delamination could not be met by any of the adhesives. The resistance to delamination showed no correlation with the shear performance for any of the adhesives. Microscopic examination of the bonded joints revealed that both the penetration behavior of the adhesives and glueline thicknesses clearly correlated with the closed assembly time.” (paper I, abstract)

For this publication, the first author conducted major parts of the laboratory work, in some cases supported by technical staff and students. In addition, the first author carried out all calculations and analyses and was responsible for composing and writing of the paper. The test design was prepared together with M. Schmidt. All co-authors contributed in terms of discussions and language to the paper.

6.2 Influence of surface preparation methods on moisture-related performance of structural hardwood-adhesive bonds

“The aim of this study was to investigate the influence of three surfacing methods (peripheral planing, sanding and face milling) on the moisture-related performance of bonded ash assemblies (*Fraxinus excelsior* L.). The different surfaces were tested in combination with four adhesives: phenol-resorcinol-formaldehyde (PRF), melamine-urea-formaldehyde (MUF), polyurethane (PUR) and emulsion polymer isocyanate (EPI). For evaluation, the surface roughness was measured and surfaces and bonds were examined by means of scanning electron microscopy (SEM) and transmitted light microscopy, respectively. To analyze bond performance, tensile shear tests were carried out as per EN 302-1 and the resistance to delamination was determined according to EN 302-2. Microscopy and roughness measurements showed significant differences between the bonding surfaces, notably with regard to cell damage and the level of fibrillation. The surface texture had significant impact on shear and delamination results. While shear tests showed good bond performance when tested in dry condition, moisture treatment revealed differences between surfaces, in particular with regard to wood failure. Based on shear results, the most appropriate surfacing method to produce moisture-resistant bonds appeared to be face milling together with PRF. Delamination results varied strongly with surfacing method and adhesive types. PRF and MUF showed highest resistance to delamination with sanded surfaces, possibly because damaged cells helped to dissipate strain. PUR and EPI provided lower moisture-related durability. For these adhesives, best results were obtained with face milled surfaces, probably because of a more homogenous strain dissipation in the glueline caused by fibrillation.” (paper II, abstract)

For this paper, the test design was developed by the first author together with S. Torno. The preparation of test specimens and the testing was done by the first author with support from E. Neuhaeuser. The data analyses as well as the composition and writing of the article was done by the first author. All co-authors contributed by discussions and language to the article.

6.3 Measurement of moisture-related strain in bonded ash depending on adhesive type and glueline thickness

“Structural wood-adhesive bonds (WAB) have to be durable while subjected to considerable stresses caused by mechanical loads and moisture content changes. To better understand the moisture-related durability of WABs, knowledge is important of how moisture changes generate strain in the bond. In this paper, strain on end-grain surfaces of bonded ash specimens was analyzed by means of digital image correlation. Strains were generated by wood shrinkage, and the evaluation was focused on shear strain (SStr). The bond lines were studied depending on the adhesive type – phenol resorcinol formaldehyde (PRF), melamine urea formaldehyde (MUF), polyurethane (PUR), and emulsion polymer isocyanates (EPI). Moreover, three different glueline (GL) thicknesses of MUF were taken into consideration. Comparing the adhesive types, SStr distributions (SStrD) were strongly influenced by adhesive elasticity. MUF and PRF bonds were quite rigid and were associated with pronounced strain amplitudes in and close to the GL together with strain dissipation reaching deep in the wood. PUR and EPI adhesives were more elastic and therefore allowed for smoother strain transition showing less distinct strain peaks. GL thickness had significant impact on SStrD. A high strain level and direct strain transition between adherends was found for the 0.01 mm GL, whereas a pronounced strain decrease was observed in the 0.1 and 0.2 mm GLs. This indicates different stress levels in the wood-adhesive interface dependent on GL thickness.” (paper III, abstract)

For this publication, the test design was developed by the first author in close cooperation with Prof. Dr.-Ing. P. Niemz, ETH Zürich. The preparation of the test specimens and the testing was done by the first author. In addition, the first author analyzed the data and composed and wrote the paper. The co-authors contributed to the content of the paper by scientific advice, discussions and language.

6.4 Further publications and conference contributions

Peer reviewed journal papers:

Schmidt M, Knorz M, Wilmes B (2010) A novel method for monitoring real-time curing behaviour. Wood Sci. Technol. 44(3):407-420.

Schmidt M, Thonnissen A, Knorz M, Windeisen E, Wegener G (2012) Relevant wood characteristics for gluing beech and ash with regard to discoloration. Eur. J. Wood Wood Prod. 70(1-3):319-325.

Peer reviewed conference papers:

Torno S, Knorz M, van de Kuilen J-W (2013) Supply of beech lamellas for the production of glued laminated timber. 4th International Scientific Conference on Hardwood Processing, Florence.

Knorz M, Torno S, van de Kuilen J-W (2013) Bonding quality of ash (Fraxinus excelsior L.) for use in glued laminated timber. 4th International Scientific Conference on Hardwood Processing, Florence.

Conference papers:

Schmidt M, Knorz M (2010) Gluing of European beech (*Fagus sylvatica* L.) and Douglas fir (*Pseudotsuga Menziesii* Mirb.) for load bearing timber structures. 11th World Conference on Timber Engineering, Trentino.

Schmidt M, Knorz M, van de Kuilen J-W (2010) Gluing of european hardwoods for load bearing timber structures. Conference of Processing Technologies for the Forest and Biobased Products Industries, Kuchl.

Knorz M, van de Kuilen J-W (2012) Development of a high-capacity engineered product – LVL made of European beech (*Fagus sylvatica* L.). 12th World Conference on Timber Engineering, Auckland.

Jiang Y, Schaffrath J, Knorz M, Winter S (2013) Bonding of various wood species – Studies about their applicability in glued laminated timber. RILEM Conference – Materials and joints in timber structures, Stuttgart.

Jiang Y, Schaffrath J, Knorz M, Winter S, van de Kuilen J-W (2014) Applicability of various wood species in glued laminated timber - Parameter study on delamination resistance and shear strength. 13th World Conference on Timber Engineering, Quebec.

Other publications:

Knorz M (2012) Verklebung von Buchen- und Eschenholz für tragende Holzbauteile. Gülzower Fachgespräche - Stoffliche Nutzung von Laubholz, Fachagentur Nachwachsende Rohstoffe e.V. (FNR) (ed.), Würzburg.

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8 Appended papers

Paper I

Structural bonding of ash (Fraxinus excelsior L.): resistance to delamination and performance in shearing tests

Markus Knorz, Michael Schmidt, Stefan Torno and Jan-Willem van de Kuilen (2014)

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Structural bonding of ash (*Fraxinus excelsior* L.): resistance to delamination and performance in shearing tests

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Abstract The utilization of ash (*Fraxinus excelsior* L.) allows for a significant enhancement of the load-bearing capacity in structural laminated products. However, such applications fundamentally require high-strength and durable bonds between lamellas and in finger joints. Therefore, the aim of the present survey was to evaluate ash bondings in terms of resistance to delamination (EN 302-2) and shear performance (EN 392). Investigations were performed with five adhesives: phenol-resorcinol-formaldehyde (PRF), melamine-urea-formaldehyde (MUF-1, MUF-2), polyurethane (PUR), emulsion polymer isocyanate (EPI) and varying closed assembly times as key bonding parameter. For all tested adhesives and closed assembly times, the shear test showed high wood failure percentages and bond strength values that compare to solid ash. In contrast, for resistance to delamination, significant differences were found between the adhesives as well as between closed assembly times, with improving resistance to delamination for increasing closed assembly times. The best performance was determined for the PRF-adhesive and long closed assembly times. However, standard requirements for resistance to delamination could not be met by any of the adhesives. The resistance to delamination showed no correlation with the shear performance for any of the adhesives. Microscopic examination of the bonded joints revealed that both the penetration behavior of the adhesives and glueline thicknesses clearly correlated with the closed assembly time.

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Verklebungen aus Eschenholz (*Fraxinus excelsior* L.) für tragende Bauteile - Ergebnisse aus Delaminierungs- und Scherversuchen

Zusammenfassung Mit der Verwendung von Eschenholz (*Fraxinus excelsior* L.) kann die Tragfähigkeit geklebter Holzbauteile deutlich erhöht werden. Eine grundlegende Voraussetzung ist jedoch, dass zwischen Lamellen aus Eschenholz sowie in Keilzinkenverbindungen hochfeste und dauerhafte Verklebungen erzielt werden. Das Ziel der vorliegenden Untersuchung war daher, Eschen holzverklebungen mit Hilfe der Delaminierungs- (EN 302-2) und der Scherprüfung (EN 392) zu bewerten. Das Eschenholz wurde mit fünf verschiedenen Klebstoffen (Phenol-Resorcin-Formaldehyd (PRF), Melamin-Harnstoff-Formaldehyd (MUF-1, MUF-2), Polyurethan (PUR), Emulsion-Polymer-Isocyanat (EPI)) verklebt. Zudem wurden bei der Verklebung unterschiedliche geschlossene Wartezeiten berücksichtigt. In der Scherprüfung wurden mit allen getesteten Klebstoffen und geschlossenen Wartezeiten generell hohe Holzbruchanteile sowie Scherfestigkeiten erzielt, die an die Festigkeit des Holzes heranreichten. Im Gegensatz dazu zeigten sich in der Delaminierungsprüfung stark unterschiedliche Ergebnisse in Abhängigkeit des verwendeten Klebstoffs und der geschlossenen Wartezeit. So konnte für alle Klebstoffe mit zunehmender geschlossener Wartezeit eine höhere Delaminierungsbeständigkeit nachgewiesen werden, wobei sich der PRF-Klebstoff in Kombination mit langen

geschlossenen Wartezeiten am leistungsfähigsten erwies. Trotzdem konnten die normativen Anforderungen an die Delaminierungsbeständigkeit von Eschenholzverklebungen von keinem der verwendeten Klebstoffe erreicht werden. Für keinen der verwendeten Klebstoffe konnte zudem ein Zusammenhang zwischen den Ergebnissen der Delaminierungs- und der Scherversuche festgestellt werden. In mikroskopischen Untersuchungen der Klebfugen wurden dagegen Zusammenhänge zwischen der geschlossenen Wartezeit und dem Eindringverhalten der Klebstoffe sowie den erhaltenen Klebfugendicken festgestellt.

1 Introduction

1.1 Ash wood for structural applications

In Europe, the utilization of hardwoods for load-bearing timber structures is considered to have significant future potential. This evaluation is based on both forestry surveys (e.g., Polley et al. 2009, Krackler et al. 2011) and outstanding mechanical properties of some hardwoods. In this context, European ash (*Fraxinus excelsior* L.) is regarded as an important hardwood species with strength and stiffness characteristics that significantly exceed the mechanical properties of softwood. Therefore, the use of ash will lead to an enhancement of the load-bearing capacity of structural wood products.

The valuable mechanical properties of ash have been known for a long time (e.g., Kollmann 1941, 1951; Schwenke 1956). However, this knowledge is based on tests with clear specimens and therefore, the validity for structural timber is limited. Investigations on ash timber in large dimensions, which are fundamental for utilization as structural elements, have been carried out by Quer (1997), Frühwald and Schickhofer (2004) and Glos and Torno (2008). However, due to various reasons (e.g., limited lengths of sawn timber due to warping of stems) the application spectrum of solid ash timber is limited. The use of ash in products such as glulam offers however a valuable alternative, provided that gluability and glueline integrity can be ensured. So far, the utilization of ash in structural laminated products has hardly been studied scientifically. In the course of a research study by Frühwald et al. (2003), the high potential of ash glulam could be confirmed, although the quantity of specimens was very limited. Individual experiences with ash glulam have been gained in Switzerland within the last years as more liberal building regulations allow for the application of ash glulam (Bogusch 2012). However, the specific bonding characteristics of ash have not been examined in detail yet. In particular, there is a lack of knowledge of the long-term behavior of ash bondings when glued with different adhesive types and varying bonding parameters.

1.2 Bonding of hardwood and bond durability

In general, the utilization of timber in structural laminated products requires a secure and durable bond. While there is extensive experience as well as a number of adhesive systems available for structural bonding of softwoods, the knowledge concerning bonding of hardwoods in load-bearing products is limited. A good bond performance is generally considered more difficult to achieve with hardwoods of higher density (Marra 1992). In this context, ash with a mean oven-dry density of 650 kg/m^3 (Kollmann 1941) belongs to the group of medium-density hardwoods. According to Marra (1992), medium-density wood features lower porosity than low-density wood because of thicker cell walls and as a consequence, less penetration of adhesive may cause weaker bonds. In addition, high density is usually associated with enhanced strength and stiffness characteristics as well as increased shrinkage and swelling and therefore, the stresses in bonds induced by moisture changes may be significantly higher (Marra 1992). Furthermore, Truax and Selbo (1956) state that stress in the glueline generated by swelling or shrinkage can initiate bond failure and hence is related to bond durability. For this reason, bond durability is usually determined with accelerated aging tests that use the properties of the tested wood species to induce stresses on the gluelines by means of severe and rapid moisture changes. The research by Truax and Selbo (1956) formed the basis for standardization and requirements for delamination. Since then, the application of these tests has also been extended to other types of adhesive and differing wood species. At present, the commonly used standards for the evaluation of structural bond durability are ASTM D2559 (2004), EN 302-2 (2004) and EN 391 (2001). In Europe, two established standards describe the delamination test with varying parameters (e.g., specimen size, drying temperature), for adhesive approval tests (EN 302-2) and for factory production control (EN 391). The respective requirements to be met by the specimens are specified, for example, for phenolic and aminoplastic adhesives in EN 301 (2006) and, for instance, for glulam, in EN 386 (2001). The standards define test parameters which mainly depend on the adhesive type (type I for exposure to high temperatures or outdoor conditions, type II for indoor applications) and on the service class according to EN 1995-1-1 (2010) in which a product will be used.

1.3 Research on structural bonding of hardwood in Central Europe

Due to its importance in forestry, scientific activities in central Europe in the last years have primarily been focused on bonding of beech (*Fagus sylvatica* L.) (e.g.,

Aicher and Reinhardt 2007; Schmidt et al. 2010). On the one hand, comparability between ash and beech exists with regard to density values and enhanced strength and stiffness characteristics. In contrast, differences can be found in swelling and shrinkage characteristics (Kollmann 1951). Also, the anatomy of ash and beech differs significantly from each other. Ash with its ring porous structure provides a more inhomogeneous surface for bonding than the diffuse porous beech wood. In addition, water absorption and pH-value, wood characteristics that proved to be relevant for bonding, were found to be different (Schmidt et al. 2012). As a result, the bond performance obtained with beech may give an indication of ash bondings, but cannot be directly transferred.

The bond durability of beech bondings was determined in delamination tests and found to be influenced by the specimen width (Aicher and Reinhardt 2007), the lamella thickness (Ohnesorge et al. 2010) and the alignment of the annual rings in the lamellas (Ohnesorge et al. 2008). However, the closed assembly time, i.e., the time period between assembly of the lamellas after glue spread and application of pressure, was identified as a key bonding parameter required to achieve high bond durability (Ohnesorge et al. 2010; Schmidt et al. 2010). Thus, Schmidt et al. (2010) showed that short closed assembly times in combination with increased pressure cause very high penetration and very low glueline thicknesses, respectively, and result in poor bond durability. On the other hand, extended closed assembly times led to good glueline formation and enhanced bond performance with two melamine-urea-formaldehyde (MUF) adhesives. In general, a prolongation of the closed assembly time is associated with an increase of adhesive viscosity. Hence, the adhesive viscosity at the moment of pressure application and the pressure as the driving force for hydrodynamic flow are the determining factors for penetration (Brady and Kamke 1988) and glueline thickness. In addition, applying pressure after short closed assembly times and with low adhesive viscosity, respectively, leads to considerable adhesive squeeze-out and contributes to a low glueline thickness. Consequently, an adjusted viscosity obtained by means of increased closed assembly time was assumed to be a requirement for good durability of medium-density wood bondings by Schmidt et al. (2010).

In contrast, the shear test according to EN 392 (1995), which is performed under dry conditions, showed little differences between the various bonding parameters (closed assembly time) with regard to shear strength and wood failure percentage (WF) in the studies described above. These results support the theory that the bond durability is mainly related to induced stresses in the glueline caused by swelling or shrinkage of the wood.

A further decisive factor for bond performance of medium-density hardwoods is the adhesive system. It was found that adhesive families of comparable bond performance for softwood can significantly vary in their bonding quality for hardwood. For example, the bond performance of several polyurethane (PUR) adhesives and one phenol-resorcinol-formaldehyde (PRF) adhesive with ash wood was examined by means of shear tests by Brandmair et al. (2012). The authors observed higher bond strengths and WFs for ash bondings with PRF than with PUR, in particular after water immersion. Furthermore, higher bond durability with beech was determined by means of delamination tests for MUF bondings compared to PUR bondings (Aicher and Reinhardt 2007; Schmidt et al. 2010).

1.4 Objectives

In this research work, the gluability of ash for structural products was examined for various adhesive systems and differing closed assembly times. For evaluation of the bond performance, two established methods were applied: (i) a delamination test to provide insight into the bond durability in accelerated aging and (ii) a shear test for evaluation of the short-term bond performance. Moreover, the gluelines were investigated with respect to bond formation and penetration behavior by means of transmitted light microscopy. The glueline thicknesses were measured using reflected light microscopy.

2 Materials and methods

2.1 Wood

The ash wood used in this survey originated from a forest area approx. 50 km south of Munich in Germany. The ash logs were cut into boards with a thickness of 40 mm and kiln dried. From the sawn wood, lamellas with a length of 550 mm and a width of 180 mm were obtained. The lamellas were stored at standard climate (20 °C and 65 % relative humidity) for several weeks. All lamellas showed annual growth rings with tangential orientation.

Prior to bonding of the delamination specimens, clear specimens were sawn from the lamellas for the determination of moisture content (MC) according to EN 13183-1 (2002) and density following DIN 52182 (1976). The mean value and standard deviation were 11.4 ± 1.4 % for the MC and 661 ± 60 kg/m^3 for the density of the lamellas in relation to 12 % MC, respectively.

2.2 Adhesives

Currently, four cold-setting chemical adhesive systems are prevalent in the production of structural wood products

such as glulam. To obtain a substantial overview of the performance and the potential of these adhesive systems in combination with ash, all four adhesive systems were included in the research:

- Phenol-resorcinol-formaldehyde (PRF)
- Melamine-urea-formaldehyde (MUF): due to the importance of these adhesive systems for the production of structural wood products, two adhesives were tested with variation in the mixing ratio of one adhesive (MUF-1, MUF-2-100/20 and MUF-2-100/50)
- Emulsion polymer isocyanate (EPI)
- Polyurethane (PUR)

While PRF, MUF and EPI are two-component adhesive systems, PUR adhesive consists of one component. The adhesives used in this study meet the requirements of adhesive type I according to EN 301 (2006) and EN 15425 (2008), respectively. Requirements given in these standards are specified with regard to longitudinal tensile shear strength (EN 302-1) as well as resistance to delamination (EN 302-2). Until now, the performance of these adhesives in combination with the medium-density hardwood ash has not been determined.

2.3 Bonding parameters and test specimens

All the bonded members were made in the laboratory at 20 °C and 65 % relative humidity. The two-component adhesives were processed in mixed application. The adhesives were applied one-sided on the lamellas by means of a spatula. The mixing ratio, adhesive quantity, and pressing time were specified in consultation with the adhesive manufacturers. For all bonded members, an open assembly time below 5 min and a pressure of 1.2 N/mm^2 were used. When designing the experiment, the focus was put on the closed assembly time because this bonding parameter has been proven to be important for the bonding quality of beech (Schmidt et al. 2010). The bonding parameters as well as the number of bonded members are listed in Table 1.

The surfaces were produced by means of a conventional surface planing machine. The time period between surface preparation and bonding was at most 6 h. The bonded members consisted of six lamellas with a thickness of 30 mm, a length of 500 mm and a width of 150 mm. The members were assembled with equally oriented growth rings in accordance with the specifications given in EN 302-2 (Fig. 1). The time for post-curing after removal from the press was at least 7 days. The specimens were sawn from the bonded members as shown in Fig. 1. In the delamination test, for each bonding parameter two specimens with the cross-section of the bonded members and a length of 75 mm were tested. The specimens used for shear tests were obtained in the form of two adjacent slats with five gluelines each and with a cross section A of 50 mm x 50 mm.

2.4 Test methods and glueline analysis

2.4.1 Delamination test

In this research study, the delamination test as defined in EN 302-2 was applied. This is due to the fact that in Germany technical approval of a building product such as ash glulam requires proof of bond durability by means of a delamination test for adhesive type I according to EN 302-2.

The test was performed in three cycles with a drying plant (ULWA-E, Ulrich Lübbert Warenhandel GmbH & Co. KG, Germany). Each cycle consisted of an impregnating period at varying pressure levels (two sub-cycles, each for 15 min at 25 ± 5 kPa and 1 h at 600 ± 25 kPa absolute pressure) and a subsequent drying period at 65 ± 3 °C, 12.5 ± 2.5 % relative humidity and an air speed of 2.25 ± 0.25 m/s. One impregnating-drying cycle was finished "when the mass of the test piece was between 100 and 110 % of the original mass" (EN 302-2, 2004, p. 7).

After the test, the lengths of the glueline openings were determined on both end grain surfaces of the specimen.

Table 1 Adhesives and bonding parameters
Tab. 1 Verwendete Klebstoffe und Verklebungsparameter

	PRF	MUF-1	MUF-2		EPI	PUR
Adhesive components	2	2	2		2	1
Adhesive application	Mixed application of resin and hardener					N/A
Mixing ratio resin/hardener	100/20	100/60	100/20	100/50	100/15	N/A
Quantity of adhesive spread in g/m^2	450	450	450		300	200
Closed assembly time in min	15–100	30–120	30–90	30–70	10–25	20–50
Pressing time in h	6	12	12	6	2.5	24
Number of bonded members	8	8	3	5	4	2

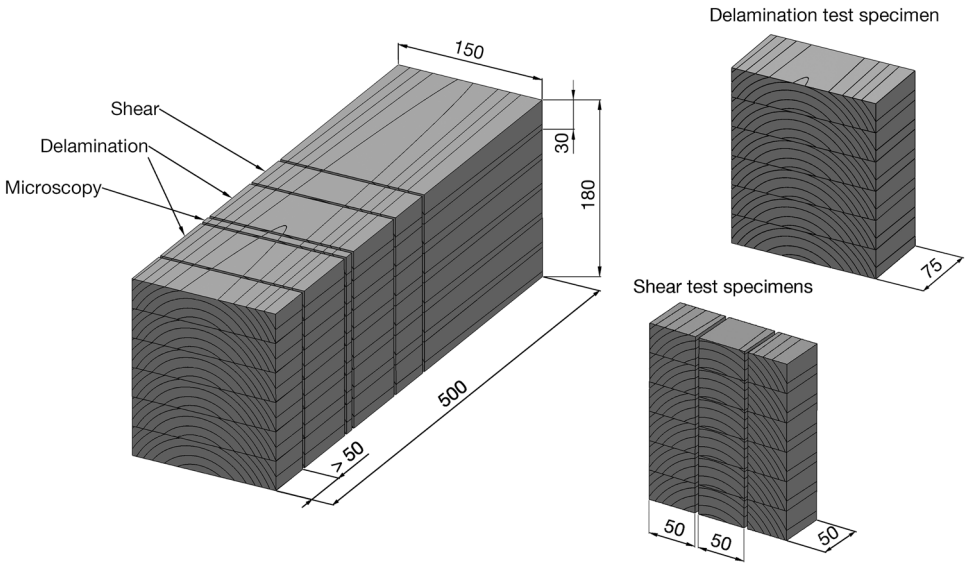

Fig. 1 Cutting of test specimens from the bonded members (dimensions in mm)
Abb. 1 Zuschnitt der Prüfkörper aus den verklebten Kleinträgern (Abmessungen in mm)

The resistance to delamination is described by the delamination values D_{SP} and D_{GL}, which were calculated by means of the lengths of the glueline openings l_{Delam} relative to the glueline lengths l_{GL}.

$$D_{SP} = \frac{\sum l_{Delam,SP}}{\sum l_{GL,SP}} \times 100 \quad \text{in } \% \quad (1)$$

$$D_{GL} = \frac{\sum l_{Delam,GL}}{2 \times l_{GL}} \times 100 \quad \text{in } \% \quad (2)$$

While Eq. (1) refers to the total delamination of a test specimen (D_{SP}), the delamination value for a single glueline D_{GL} is defined in Eq. (2). All delaminated gluelines were opened after the test with a chisel for closer investigation of the delaminations.

2.4.2 Shear test

The shearing behavior was determined in a block shear test according to EN 392. This block shear test is, together with the delamination test, an approved method to verify the bonding quality when producing, for instance, glulam according to EN 386. The specimens were tested parallel to the grain with the shear plane corresponding to the adhesive layer. As a reference, tests with solid timber were performed to determine the shear strength of ash wood. The shear test was performed with a universal testing machine (DOLI Elektronik GmbH, Germany) using a displacement rate of 3 mm/min. The shear strength f_v was calculated by means of the load at failure F_{max} and the cross-section A of the respective specimen (Eq. 3):

$$f_v = \frac{F_{max}}{A} \quad \text{in N/mm}^2 \quad (3)$$

Furthermore, for each shear plane the WF was estimated and rounded to the nearest 5 %.

2.4.3 Microscopy

Analysis of the gluelines was performed with two microscopes. A transmitted light microscope (Zeiss Axiophot with Imaging software Axio Vision 4.8.3.0) was primarily used for qualitative analysis of the penetration behavior of the adhesives in the wood tissue and the glueline formation. For this investigation, microtome sections with a thickness of approx. 20 µm were sliced from small cubes (approx. 10 mm × 10 mm) extracted from a pre-cut part of the bonded members (Fig. 1). Depending on the type of adhesive the slices were stained with safranin and methylene blue, respectively.

For the determination of the glueline thicknesses, a reflected light microscope (Leica MZ FLIII with Imaging software Leica Application Suite 4.1.0) was used. The measurements were performed on the end-grain surface on the upper and the second lowest glueline of a specimen (Fig. 1). These gluelines were chosen to verify the possible influence of slight differences in open assembly times between the layers. For these gluelines, four measurement areas (10 and 50 mm from both edges) were defined and three glueline thickness values were determined for each area. The measuring accuracy was 0.5 µm.

3 Results

3.1 Delamination test

First, the duration of the drying cycles of the delamination test was documented for the ash specimens. The mean drying time to reach the target mass of between 1.0 and 1.1 times the initial mass of the specimens was 32.2 h. Table 2

Table 2 Results of the delamination test according to EN 302-2
Tab. 2 Ergebnisse der Delaminierungsprüfung nach EN 302-2

Adhesive	Closed assembly time in min	N (specimens/gluelines)	D_{SP} (%)	D_{GL} (%)	
			Mean	Min	Max
PRF	15	2/10	38.3	3.3	57.3
	30	2/10	21.2	3.0	48.0
	60	6/30	11.4	0	54.7
	80	4/20	8.8	0	28.0
	100	2/10	4.2	0	19.3
MUF-1	30	2/10	63.8	43.3	89.3
	60	6/30	55.9	15.3	86.3
	90	4/20	45.6	23.0	78.3
	120	4/20	39.2	9.3	74.3
MUF-2, 100/20	30	2/10	43.4	29.3	59.7
	60	2/10	46.2	27.3	65.0
	90	2/10	26.5	6.7	45.3
MUF-2, 100/50	30	2/10	22.4	4.3	65.3
	50	2/10	21.9	4.3	35.3
	70	6/30	14.1	1.3	46.0
PUR	20	2/10	86.9	71.0	100
	50	2/10	77.3	31.0	100
EPI	10	2/10	73.7	48.3	92.7
	25	6/30	70.3	11.7	98.0

shows the results of the delamination test in dependence of closed assembly time for each adhesive. In addition, the results of the delamination test are visualized in Fig. 2 by means of boxplots. The test revealed significant differences in terms of resistance to delamination among the adhesive systems. The highest resistance to delamination was found for PRF bondings. The mean delamination ranged from 38.3 (15 min) to 4.2 % (100 min). The test results showed a clear relationship between closed assembly time and delamination values (Fig. 2). For the MUF adhesives, the results obtained varied considerably between MUF-1, MUF-2-100/20 and MUF-2-100/50. The delamination values for MUF-1 ranged from 63.8 (30 min) to 39.2 % (120 min). As already described for PRF, the influence of the closed assembly time on the delamination behavior is evident. In comparison to MUF-1, the delamination values are significantly lower for MUF-2. While the mean delamination for MUF-2-100/20 ranged from 46.2 (60 min) to 26.5 % (90 min) lower values between 22.4 (30 min) and 14.1 % (70 min) were determined for the mixing ratio 100/50. An increase in the closed assembly time from 30 min to 50 and 60 min, respectively, produced only a marginal effect on the resistance to delamination. In contrast, further prolongation of the closed assembly time (70 min/90 min) showed a clear enhancement of the resistance to delamination.

Statistical analysis showed significant differences between the delamination values D_{GL} with respect to the closed assembly times for PRF, MUF-1 and MUF-2-100/20 (Kruskal–Wallis, $p < 0.05$). Despite substantial variations in the distribution of delamination values for MUF-2-100/50 (Fig. 2), the differences between the closed assembly times were statistically not significant for this adhesive.

PUR and EPI bondings showed the greatest delamination of all adhesives. The influence of a longer closed assembly time on the delamination behavior is noticeable as delamination values decrease, even though the influence seems to be limited. For PUR and EPI bondings, no statistical evidence was found for delamination differences between the various closed assembly times.

The requirement given in adhesive standards of a maximum delamination of 5 % for each specimen could not be fulfilled by any of the tested adhesives with any closed assembly time. The best test result was obtained for PRF bonding and a closed assembly time of 100 min with a mean delamination of 4.2 %. However, one of the specimens demonstrated 7.1 % delamination, exceeding the standard requirement.

After testing, delamination occurrences were inspected following separation of the adherends with a chisel. For the assessment of the failure pattern, the brown PRF and whitish MUF adhesives could easily be distinguished from

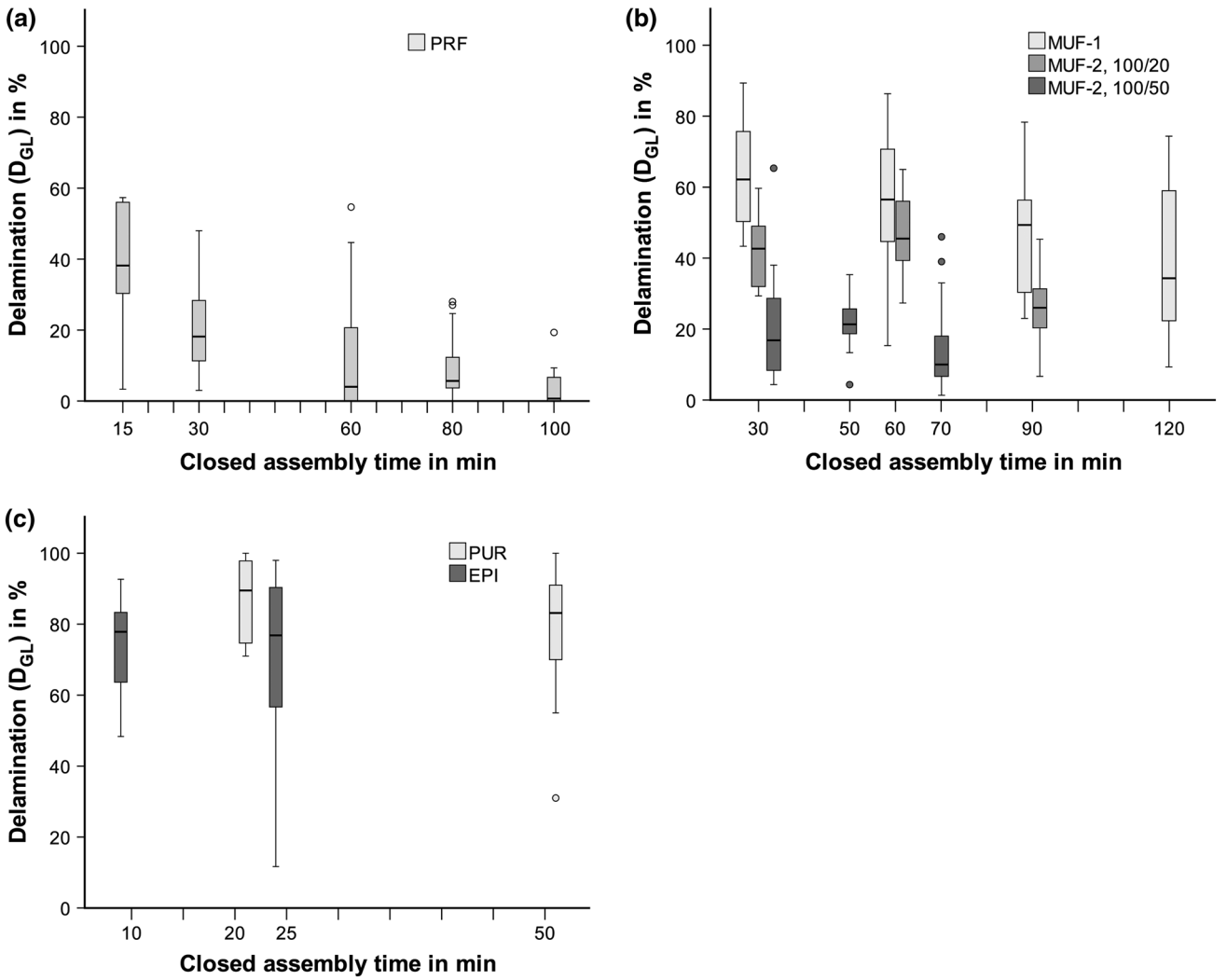

Fig. 2 Resistance to delamination (D_{GL}) for ash bondings with **a** PRF, **b** MUF and **c** PUR- and EPI adhesives in dependence of the closed assembly time. Boxplot elements: *box* values between the 25th and 75th percentile (interquartile range), *horizontal line* median, *whiskers* lowest/highest values, *circles* outliers beyond 1.5 times the interquartile range

Abb. 2 Delaminierungsbeständigkeit (D_{GL}) von Eschenholzverklebungen mit **a** PRF, **b** MUF sowie **c** PUR- und EPI-Klebstoffen in Abhängigkeit der geschlossenen Wartezeit. Boxplot Elemente: Box = Wertebereich zwischen dem 25. und 75. Perzentil (Interquartilsabstand); Horizontale Linie = Median; Whiskers = niedrigster/höchster Wert; Kreise = Ausreißer, die außerhalb der Grenze des 1,5-fachen Interquartilsabstands liegen

the wood. For specimens made of softwood species such as spruce, the glueline openings initially assessed as delamination on the end-grain surface are often covered by thin layers of fiber and therefore, have to be frequently reevaluated as wood failure. In contrast, examination of the ash specimens revealed a predominantly distinct adhesion failure in the interface between adhesive and wood. For PUR and EPI adhesives the differentiation between wood and adhesive was difficult because of their transparency. However, the distribution of the adhesives on the adherends could be visualised by means of ultraviolet light, and failure in the adhesive-wood interface could be confirmed for PUR and EPI. For all adhesives, delaminations frequently occurred in areas where latewood was predominant.

3.2 Shear test

The results of the shear test for solid ash and ash bondings are displayed in Table 3 and Fig. 3, respectively. The mean values for the shear strength f_v ranged between 13.2 N/mm^2 (PUR) and 14.7 N/mm^2 (MUF-1). As a reference, $f_{v,mean}$ of 14.3 N/mm^2 was determined for solid ash. There was no significant difference between the shear strengths of solid ash and PRF and MUF bondings. This fact and the high mean WF (min. mean WF 88 %, MUF-2-100/50) support the assumption that the cause of failure for the bondings lies in the wood itself and not in the glueline.

Somewhat lower mean values both for f_v (13.2 N/mm^2) and WF (63 %) were found for PUR bondings. Statistical

Table 3 Results of the shear test according to EN 392
Tab. 3 Ergebnisse der Scherprüfung nach EN 392

Specimen	N	Shear strength f_v in N/mm^2				CoV (%)	WF (%) Mean
		Mean	Std	Min	Max		
Solid ash timber (reference)	35	14.3	2.3	8.3	18.0	15.9	–
PRF	80	14.6	1.7	9.9	19.0	11.6	96
MUF-1	80	14.5	1.6	7.1	17.7	10.9	89
MUF-2, 100/20	30	14.6	1.4	11.8	17.3	9.6	99
MUF-2, 100/50	50	14.4	2.3	6.4	17.6	15.8	88
PUR	20	13.2	1.0	11.2	15.4	7.8	63
EPI	40	13.7	2.1	9.4	17.8	15.3	90

investigations by means of variance analysis (ANOVA) and Tamhane's post hoc test revealed significant differences ($p < 0.05$) between the shear strengths obtained in PUR bondings and ash bondings made with PRF, MUF-1 and MUF-2-100/20. Moreover, the WF was also found to be significantly lower for the PUR sample (Kruskal–Wallis, $p < 0.05$) when compared to ash bondings made with PRF, MUF and EPI.

The evaluation of the test results is based on requirements given in EN 386 for shear strength f_v and WF both for soft- and hardwood glulam. EN 386 defines minimum values for f_v and WF, both in terms of individual values for a single glueline and the average values for a beam. According to EN 386 every glueline must reach $f_{v,min} = 6.0$ N/mm^2 and a corresponding boundary value for WF that depends on the shear strength. The requirements given in EN 386 were met by all adhesives included in the survey.

Furthermore, the influence of the closed assembly time on the shear strength f_v and the WF was examined within the adhesive samples. The analyses gave statistical evidence that there is no significant difference between shear strengths obtained with various closed assembly times. In addition, for the WF no dependency on closed assembly time was found except for PUR bondings.

3.3 Microscopy

Microscopic examination of the gluelines showed significant differences in adhesive penetration behavior and glueline formation in dependence of the closed assembly times for all adhesives. For short closed assembly times, the penetration into the earlywood vessels can be excessive and may amount to several 100 µm, whereas the adhesive penetration depth into earlywood is reduced with long assembly times (Fig. 4). In contrast, the penetration into latewood fibers seems to be independent of the bonding parameters. In these areas little or no adhesive penetration was found for all adhesives.

For the glueline formation the closed assembly time was found to have significant influence. This could be confirmed using glueline thickness measurements, the results of which are displayed in Fig. 5. All adhesives show very low glueline thicknesses, below 20 µm, with short assembly times. For bondings made with PRF, MUF-1 and

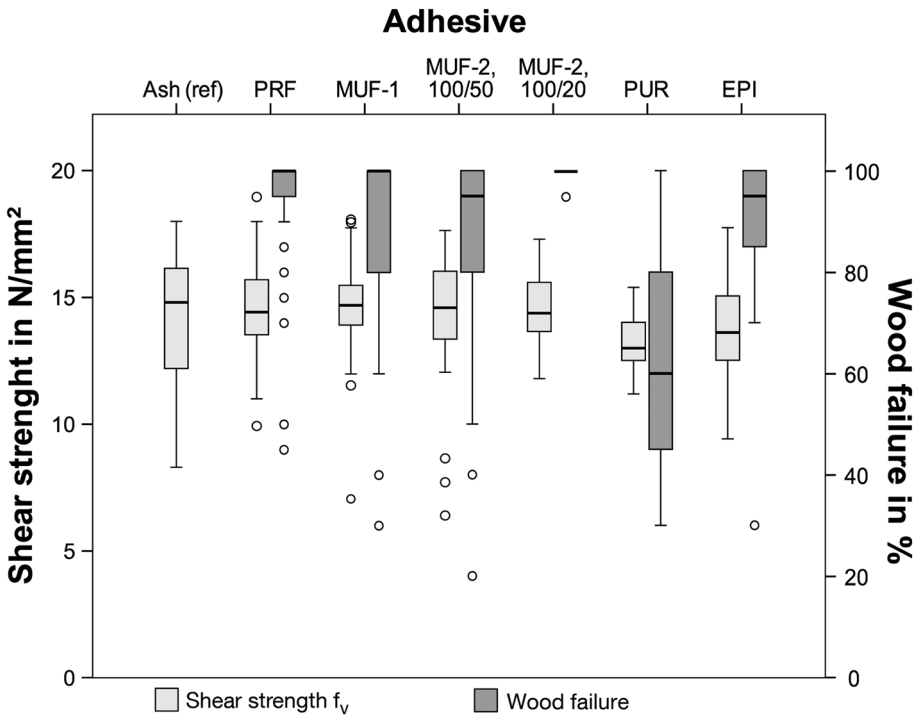

Fig. 3 Shear strength f_v of solid ash wood and ash bondings together with WFs for ash bondings. Boxplot elements are explained in Fig. 2
Abb. 3 Scherfestigkeit f_v von Eschenholz und Eschenholzverklebungen sowie Holzbruchanteil von Eschenholzverklebungen. Die Boxplot Elemente werden in Abb. 2 erklärt

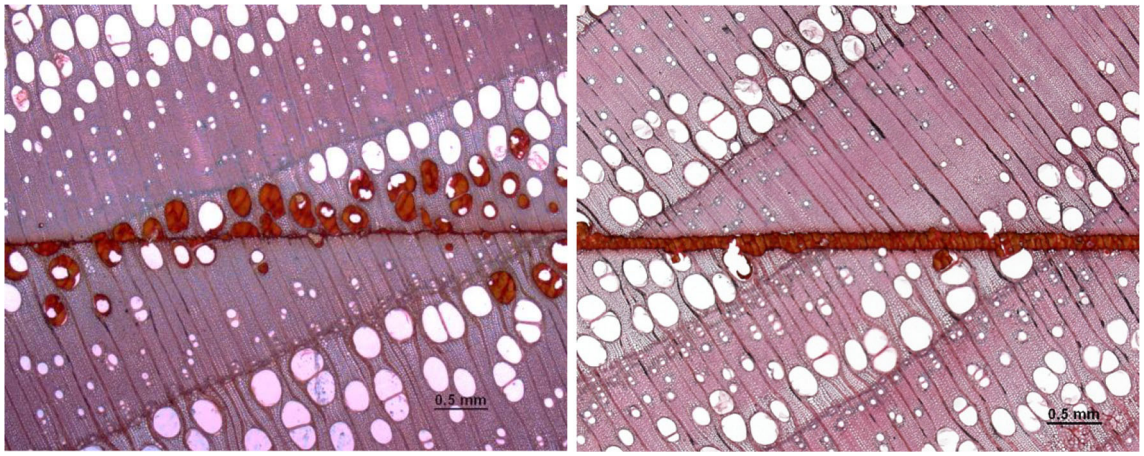

Fig. 4 Microscopic images of ash PRF bondings with closed assembly times of 30 min (*left*) and 80 min (*right*)
Abb. 4 Mikroskopische Darstellung von PRF-Eschenholzverklebungen mit den geschlossenen Wartezeiten 30 Minuten (links) und 80 Minuten (rechts)

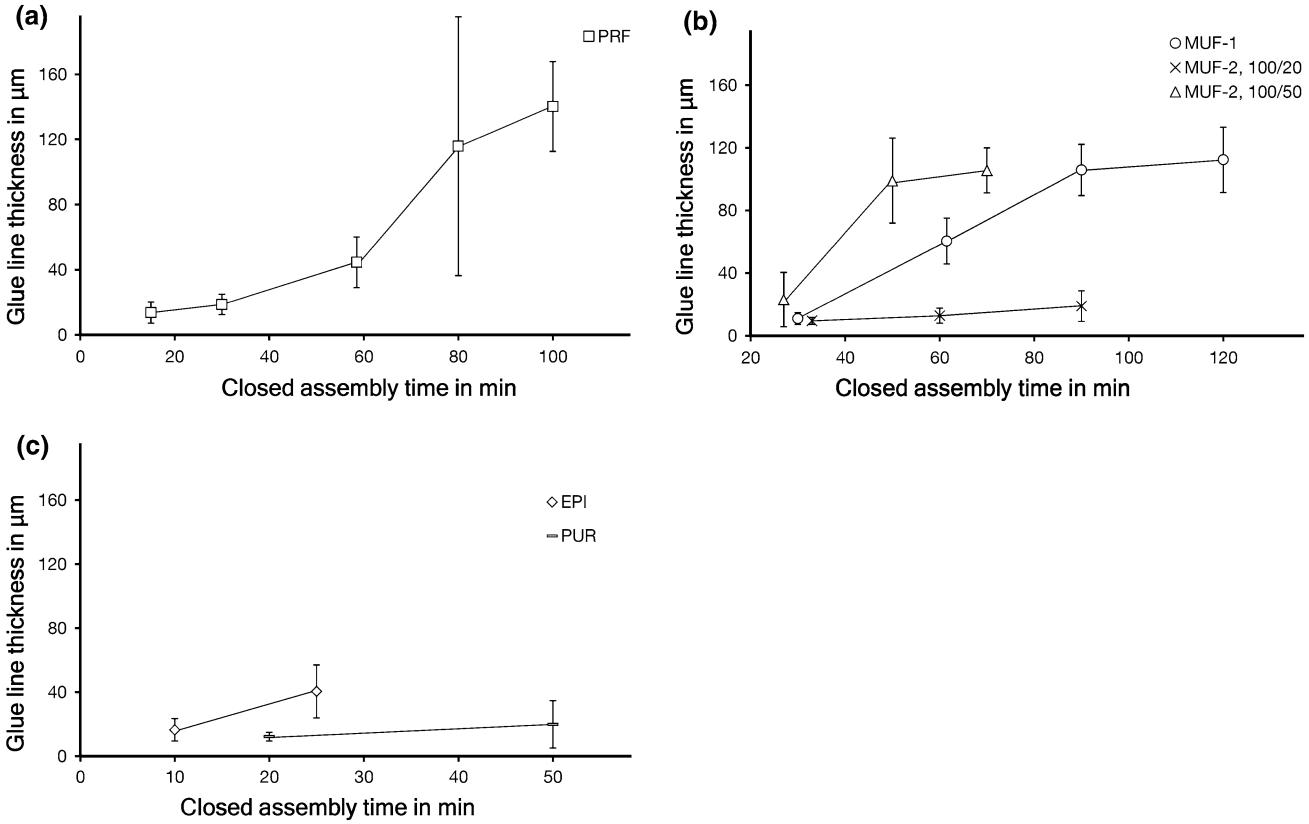

Fig. 5 Glueline thicknesses (mean values ± standard deviation) of ash bondings with **a** PRF, **b** MUF and **c** PUR and EPI adhesives
Abb. 5 Klebfugendicken (Mittelwerte ± Standardabweichung) von Eschenholzverklebungen mit **a** PRF, **b** MUF sowie **c** PUR- und EPI-Klebstoffen

MUF-2-100/50 a significant increase in glueline thicknesses with increasing closed assembly times is evident, reaching single values of up to 250 µm for PRF. In addition, the glueline thicknesses appear to approach a maximum value with increasing closed assembly times for these adhesives, which can be attributed to the quantity of adhesive spread and the corresponding solid content, respectively. In contrast, only a slight increase in glueline thickness can be found for the processing times recommended by the adhesive manufacturers for PUR, EPI and MUF-2, 100/20 (Fig. 5). The glueline thicknesses were measured on two gluelines of a specimen. No deviations

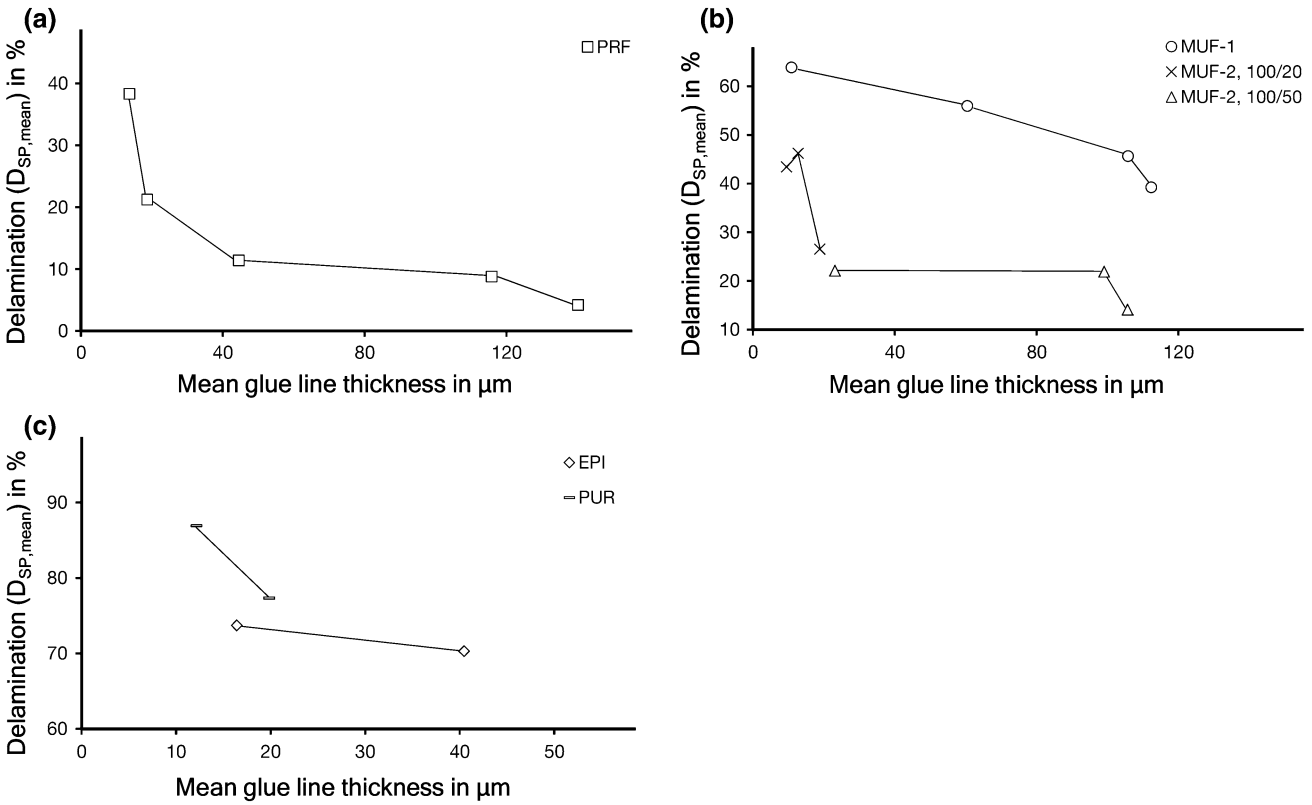

Fig. 6 Resistance to delamination of ash bondings with **a** PRF, **b** MUF and **c** PUR and EPI adhesives in dependence of glueline thickness
Abb. 6 Delaminierungsbeständigkeit von Eschenholzverklebungen mit **a** PRF, **b** MUF sowie **c** PUR- und EPI-Klebstoffen in Abhängigkeit der Klebfugendicke

were found when the thickness values between gluelines within one specimen were compared.

3.4 Glueline thickness and delamination

The results of the glueline thickness measurements and the corresponding delamination $D_{SP,mean}$ are displayed in Fig. 6. For all adhesives, there is a tendency towards increasing glueline thickness being associated with decreasing delamination. This correlation is particularly apparent for PRF and MUF-1 bondings. For both mixing ratios of MUF-2, an enhanced resistance to delamination can only be found for the highest glueline thickness. Despite high delamination values and low glueline thicknesses, also for EPI and PUR a dependency between these two characteristics can be found.

4 Discussion

4.1 Delamination behavior of different wood species and standard requirements

Ash bondings with all adhesives and bonding parameters used in the present research study did not meet the requirements for resistance to delamination of 5 % according to EN 301 and EN 15425, respectively. This is a value which all of these adhesives have met with spruce in approval tests. Since all tests were performed according to EN 302-2, so that test conditions were comparable for spruce and ash specimens, the properties of ash wood clearly influence the delamination resistance. This indicates that the stresses on the glueline vary significantly between species during the delamination test (Marra 1992). Whereas the European standards specify a maximum delamination of 5 % for all wood species, the corresponding ASTM standard for the assessment of bond durability by means of delamination (ASTM D2559) differentiates between softwood and hardwood, setting limit values of 5 and 8 %, respectively. Therefore, delamination limit values could be defined in Europe taking different wood species behavior into consideration. A rise of the maximum allowed value to 8 % would mean that ash bondings with PRF and 100 min closed assembly time in the present survey fulfill the delamination requirements.

The delamination test with ash demonstrated a long mean duration of 32.2 h of the drying cycles. In comparison, Aicher and Reinhardt (2007) determined a mean drying time of only 17.2 h for beech specimens, reaching a mean value of 1.06 times the initial mass of the specimens,

which is, according to the authors, shorter than for softwood specimens. This can be confirmed by own investigations where the mean drying time for spruce specimens was 19.5 h (unpublished). Because drying parameters were comparable for the investigations mentioned above, the deviating drying times imply that the exposure of the bonds varies strongly. Consequently, the severity of the delamination test depends on the drying behavior of the tested wood species. This may apply to varying durations when moisture-related stresses have an impact on the gluelines and for differences with regard to moisture gradients and resulting stress levels between wood species. As a consequence, the delamination test procedure should be modified to take the different moisture behavior of the wood species into account.

4.2 Resistance to delamination in relation to adhesive systems and bonding parameters

The delamination behavior of ash bondings differed significantly depending on the adhesive and bonding parameters. When comparing the adhesives, the highest resistance to delamination was found for PRF bondings. This is presumably attributed to the well-known high moisture resistance of PRF (Dunky and Niemz 2002). However, the results strongly depend on the closed assembly time, with the lowest delamination values found for the longest closed assembly time. This general picture was also found for both MUF adhesives. However, with MUF higher delamination values were obtained when compared with PRF.

Variations in resistance to delamination for MUF adhesives could be observed between the two adhesives as well as with varying mixing ratios for MUF-2. Whereas the performance of adhesives of the same adhesive family may differ because of the formulation of the adhesives, the higher amount of hardener in MUF-2 obviously leads to a higher delamination resistance. This may be due to the fact that the increase of hardener primarily involves higher percentages of the important hardener ingredients formaldehyde and polyvinyl acetate (PVAc) in the resin-hardener mixture. The higher percentage of formaldehyde leads to higher reactivity of the adhesive and a higher degree of cross-linking in the hardened glue (Dunky and Niemz 2002). The higher percentage of PVAc, however, results in more intense wetting of the wood surface and therefore, in a narrower contact between adhesive and wood (Scheikl and Dunky 1996). In addition, the bond formation in terms of glueline thickness and penetration varied for the two mixing ratios. Ash bondings with MUF-2 and a mixing ratio of 100/20 showed high penetration in earlywood and only a marginal increase in glueline thickness for all closed assembly times. This may be because there is still a too little increase in viscosity, even for a long closed assembly time of 90 min.

The highest delamination values were obtained with PUR and EPI bondings. As before with MUF-2-100/20, the closed assembly time had little influence on bond formation and delamination. The bonding parameters used were obviously not effective in avoiding high penetration into the earlywood and low glueline thicknesses. This occurred despite the use of the maximum closed assembly times as specified by the adhesive manufacturers.

4.3 Shear strength and wood failure percentage

The shear test showed high strength values both for solid ash and the ash bondings. The mean shear strength obtained for solid wood in the present study (14.3 N/mm^2) was slightly higher than values documented in literature. For instance, the mean shear strength found by Brandmair et al. (2012) was 13.27 N/mm^2. Kollmann (1941) differentiated between tangential plane and radial plane and determined shear strengths of 11.47 and 12.06 N/mm^2, respectively. Variations in shear strength may, on the one hand, be caused by the deviating shape of the test specimens. For example, Kollmann (1941) used double shear test specimens, whereas Brandmair et al. (2012) tested specimens according to ASTM 2559. In addition, the testing device itself can have significant influence on shear test results (Steiger et al. 2010). Furthermore, variations in material properties within one wood species may account for the deviations between the studies.

For ash bondings with PRF, Brandmair et al. (2012) observed a decrease in shear strength to 10.91 N/mm^2 and a slightly higher reduction in strength values to between 9.11 and 10.71 N/mm^2 for bondings with three PUR adhesives for structural purposes. The corresponding WF amounted to a high value of 90 % for PRF bondings. For PUR bondings, the WF was significantly lower with mean values between 40 and 50 %. In comparison, both the strength values and the WF obtained in this study with PRF and PUR show a slightly higher level. However, the overall picture of the results of both studies is in good agreement. For ash bondings with MUF and EPI adhesives no comparative values could be found in literature.

The differences between shear strengths and WF obtained with various closed assembly times were statistically not significant and therefore, the bonding parameters and the bond formation obviously had no influence on the shear test results. This is in accordance with findings reported earlier for beech by Aicher and Reinhardt (2007) and Schmidt et al. (2010). In addition, as was identified for beech (Ohnesorge et al. 2010; Schmidt et al. 2010), no correlation between the shear strength or WF and the delamination percentage could be found. Accordingly, the

shear test is regarded as not suitable to assess bond durability. As stated before by Ohnesorge et al. (2010), the shear test seems only applicable in order to detect severe adhesion failure.

4.4 Penetration and bond formation

Microscopic examination showed high penetration and adhesive filled vessels up to 1 mm from the glueline in the earlywood for short assembly times. This picture was found for all adhesives together with low glueline thicknesses. With increasing closed assembly times thicker gluelines ($\geq$0.1 mm) together with reduced penetration in earlywood were observed, in particular for PRF, MUF-1 and MUF-2-100/50. On the contrary, the adhesive penetration in the low porosity fibers of latewood seems not to be affected by the applied closed assembly times.

Whereas penetration in general is regarded as important for the bond performance (Kamke and Lee 2007), in the case of the ring porous wood species like ash, this needs to be discussed. For instance, high penetration in the earlywood resulted in low glueline thickness and obviously had no positive effect on the delamination behavior. A higher delamination resistance was obtained with bonds with limited glue penetration in the earlywood and with glueline thicknesses of around 0.1 mm. Adhesive penetration in the latewood was very low or even non-existent and therefore mechanical interlocking is expected to be less than in the case of spruce. This assumingly contributed to a distinct adhesion failure in the adhesive-adherend interface which normally does not occur with softwood species such as spruce. A deeper adhesive penetration in the latewood would probably contribute to a better bond performance, but apparently the anatomy of ash prevents this.

The results indicate that the bond formation and the resulting glueline thickness play key roles for delamination resistance. This echoes the findings for MUF adhesives with beech by Schmidt et al. (2010), who explained the differences in bond formation by different viscosities at the time of pressure application, and moreover, presumed that the delamination resistance of medium-density wood bondings is related to the quality of the glueline formation.

In general, the importance of the glueline formation with regard to the distribution of stresses caused by moisture changes is known. Frihart (2009) states that gluelines that allow for even stress distribution and dissipation of local stress concentrations show higher resistance to delamination. This is in agreement with a research study by Wetzig et al. (2011), who confirmed the capability of gluelines to dissipate moisture-induced stresses in dependence of glueline thicknesses and adhesive elasticities.

5 Conclusion

From delamination and shearing tests on ash specimens structurally bonded with five different adhesives (PRF, MUF-1, MUF-2, EPI, PUR) the following conclusions can be drawn:

- Differences in shear strengths and WFs were found to be minor. This applies both to bondings with the different adhesives and to various closed assembly times.
- The resistance to delamination varied strongly, depending on the adhesives and bonding parameters. In general, longer closed assembly times led to higher delamination resistance for all tested adhesives. None of the tested adhesives was able to meet the current delamination requirements given in EN 301 and EN 15425, respectively. However, PRF adhesive seems to be the most suitable for bonding of ash. If requirements were adjusted to different wood species behavior, as done in the ASTM standard, PRF adhesive would comply with standard requirements. For MUF, it was shown that the resistance to delamination strongly depends on the hardener percentage. Based on the delamination test results, PUR and EPI seem to be the least applicable adhesives for ash bondings, taking the applied bonding parameters into account.
- The delamination test frequently generated large areas with a distinct adhesion failure in the adhesive-wood interface.
- No correlation between resistance to delamination and shear performance was found for all investigated adhesives and bonding parameters.
- Penetration behavior of the adhesives and the resulting glueline thicknesses are clearly related to the closed assembly time. The shorter the closed assembly time, the more excessive penetration could be observed in earlywood, leading to thinner gluelines. In contrast, the adhesive penetration in latewood was insignificant for all closed assembly times.

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Paper II

Influence of surface preparation methods on moisture-related performance of structural hardwood-adhesive bonds

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Influence of surface preparation methods on moisture-related performance of structural hardwood–adhesive bonds

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ABSTRACT

The aim of this study was to investigate the influence of three surfacing methods (peripheral planing, sanding and face milling) on the moisture-related performance of bonded ash assemblies (*Fraxinus excelsior* L.). The different surfaces were tested in combination with four adhesives: phenol-resorcinol-formaldehyde (PRF), melamine-urea-formaldehyde (MUF), polyurethane (PUR) and emulsion polymer isocyanate (EPI). For evaluation, the surface roughness was measured and surfaces and bonds were examined by means of scanning electron microscopy (SEM) and transmitted light microscopy, respectively. To analyze bond performance, tensile shear tests were carried out as per EN 302-1 and the resistance to delamination was determined according to EN 302-2. Microscopy and roughness measurements showed significant differences between the bonding surfaces, notably with regard to cell damage and the level of fibrillation. The surface texture had significant impact on shear and delamination results. While shear tests showed good bond performance when tested in dry condition, moisture treatment revealed differences between surfaces, in particular with regard to wood failure. Based on shear results, the most appropriate surfacing method to produce moisture-resistant bonds appeared to be face milling together with PRF. Delamination results varied strongly with the surfacing method and adhesive types. PRF and MUF showed highest resistance to delamination with sanded surfaces, possibly because damaged cells helped to dissipate strain. PUR and EPI provided lower moisture-related durability. For these adhesives, best results were obtained with face milled surfaces, probably because of a more homogenous strain dissipation in the glueline caused by fibrillation.

1. Introduction

In recent years new fields of hardwood utilization have been subject to intensive discussions in Europe. In this context, ash (*Fraxinus excelsior* L.) is considered an important hardwood species that proves beneficial in load-bearing structures [1] because of valuable mechanical properties [2]. To facilitate application in engineered wood products such as glued laminated timber, it is important that bond strength and durability in ash wood assemblies can be guaranteed. It is generally considered more difficult to obtain durable bonds with hardwoods than with softwoods [3]. However, little information about moisture-related durability of bonded hardwood assemblies can be found in literature. A recent study on bonded ash assemblies revealed low moisture-related durability and extensive failure at the wood–adhesive interface with peripherally planed surfaces in delamination tests [4]. Because of these unsatisfactory results a possible positive effect of different surfacing methods (peripheral planing, sanding, face milling) on bond performance after exposure to moisture changes was investigated in this new study.

The preparation of wood surfaces is of high importance for bonding quality. The texture of a bonding surface is determined by two factors: (i) the kind and the quality of the machining process and (ii) the wood species, i.e., the exposed anatomical structure and the response to the machining process. To evaluate the machining quality or to characterize the bonding surface, the surface roughness has been used in several studies [e.g., 5–8]. However, based on literature, roughness and bonding quality do not clearly correlate. For example, a certain roughness caused by damaged or fibrillated fibers can help to improve the bonding quality when compared to smooth surfaces [9]. On the contrary, high roughness also may cause decreasing bond strength [10] when, for example, crushed and damaged cells become prevalent and create a mechanically weak boundary layer (MWBL) [9].

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Peripheral planing is the most common surfacing method in the woodworking industry and almost exclusively used in the production of glued laminated timber. In the literature, varying observations on the quality of planed surfaces can be found. Singh et al. [11] and Kläusler et al. [8] showed that surface quality significantly depends on the condition of planing knives. When using sharp knives, peripheral planing produced surfaces with open cells that facilitated penetration [7,12]. Bond quality of planed surfaces was heterogeneous compared to other surfacing methods. Therefore, both enhanced [13] and impaired [8] bond performance were found with planed surfaces.

Sanding is a widely-used method to create smooth, homogeneous surfaces previous to wood coating [14]. However, it has been given little attention as a preparation method for bonding of load-bearing products. The sanding process is characterized by a negative rake angle and high normal forces [15] which generally leads to increased surface damage. Sanded surfaces showed crushed and damaged cells [6,16,17] as well as torn-out fibrils [12,18]. The degree of damage highly depended on the grit size [13,16,18]. Damaged cells can have both the effect of inhibiting adhesive penetration into the sound wood tissue [10] and of preventing excessive penetration into earlywood [12]. As mentioned above, fibrillation, i.e., partially detached or slightly crushed cell walls components, is considered to contribute to a good bonding quality. This could be confirmed in studies from de Moura and Hernández [12] and Cool and Hernández [7] with sugar maple and black spruce wood, respectively. While bonds of surfaces sanded with coarse grit (36) performed poorly [13], utilization of finer grit (80–180) resulted in good performance [8,12].

Face milling is a surfacing method characterized by a cutting direction primarily perpendicular to the grain. The blades are mounted on a cutter disk and are characterized by two cutting edges – a peripheral cutting edge for material removal and a face cutting edge to create the surface [19]. As concluded by de Moura et al. [20], the machining principle results in lower cutting forces than a machining process parallel to the grain because of lower strength of the wood in transverse direction. As a consequence, damages at wood surfaces prepared by face milling are comparatively low [6,7]. In addition, face milling produces cell-wall fibrillation which is considered to be beneficial for adhesion [20,21]. When compared to other surfacing methods, face milled surfaces showed equivalent or better bond performance [7,8].

For the present study, the above presented surfacing methods peripheral planing, sanding and face milling were applied to prepare bonded assemblies with four different adhesive types phenol-resorcinol-formaldehyde (PRF), melamine-urea-formaldehyde (MUF), polyurethane (PUR) and emulsion polymer isocyanate (EPI). Surface topology was examined by means of surface roughness measurements and microscopy (SEM, transmitted light microscopy). Bond performance of surfaces and adhesives was evaluated in tensile shear tests according to EN 302-1 [22] and delamination tests following EN 302-2 [23]. Emphasis was placed on performance after exposure to moisture change.

2. Materials and methods

2.1. Wood and surface preparation

The study was performed with ash wood (*Fraxinus excelsior* L.) from southern Bavaria, Germany. Kiln dried timber of approximately 40 mm thickness was cut to 170 mm wide and 1050 mm long boards. The boards were stored at 20 °C and 65% relative humidity (RH) until equilibrium moisture content was reached. The moisture content ω and the density ρ_{12} were (10.8 ± 1.2)% (mean $(\bar{x}) \pm$ standard deviation (sd)) and (633 ± 75) kg/m³, respectively. Boards for tensile shear tests and surface analyses showed angles between 30° and 90° between surface and annual rings. For those examinations, the boards were cut lengthwise into three layers with each layer being divided into three panels of 320 mm length. The nine panels were machined to a cross-section of 5 mm by 140 mm. With eight of these panels, tensile shear specimens were prepared. The remaining panel was used for surface roughness measurements and SEM microscopy. Boards with tangentially aligned annual rings were used for the delamination tests. The boards were cut into two sections to obtain lamellas of 500 mm length, 160 mm width and 30 mm thickness.

The surfaces were prepared by means of the three surfacing methods peripheral planing (hereinafter referred to as planing), sanding and face milling. The surfacing machines and parameters used in this study are displayed in Table 1.

2.2. Adhesives and bonding parameters

The adhesive selection represents the range of chemical systems available on the market for structural face gluing: PRF, MUF, PUR and EPI. As far as known, the four adhesives used in this study have all been successfully applied with spruce and comply with the requirements of European standards [24-26] for adhesives for load-bearing timber products. For the two component adhesives (PRF, MUF, EPI), mixed application was used. All adhesives were spread one-sided with a spatula. Bonding was performed in a climate room at 20 °C and 65% RH within 6 h after surface preparation. Further bonding parameters are displayed in Table 2. The bonding operations were all performed within the specifications of the adhesive manufacturers. A random selection of specimens including all adhesives and surfacing methods showed a mean glueline thicknesses of 81 μm (± 34 μm (sd)).

2.3. Analysis and test methods

2.3.1. Microscopy

SEM was used to examine the surface textures. For this purpose, small specimens with 20 mm length, 10 mm width and 5 mm thickness were prepared and coated with a thin layer of gold to provide conductivity. Micrographs were taken at 20 kV acceleration voltage using a Zeiss “EVO 40 XVP” microscope with “Smart SEM V05.04.03.00” software.

Table 1
Surfacing machines and parameters.

	Planing	Sanding	Face milling
Surfacing machine	Otto Martin “T43”	Kuendig “MAGIQ”	Ledinek “Rotoles 400 D”
Characterization of knives/sanding belt	HSS, freshly sharpened	80 grit, new sanding belt	HSS, freshly sharpened
Number of cutting edges z	4	n/a	48
Cutting speed v_c (m/s)	32.7	17	80
Feed speed v_f (m/min)	6	7	10
Feed f_z (mm)	0.3	n/a	0.07
Cutting depth (mm)	2	0.5	2.5
Rake angle	34°	n/a	Axial: 10°, radial: 15°

Table 2
Adhesives and bonding parameters.

Adhesive	Mixing ratio resin/hardener (parts by weight)	Adhesive spread (g/m^2)	Open assembly time (min)	Closed assembly time (min)	Pressure (MPa)	Press time (h)
PRF	100/20	450	< 5	60	1.2	6
MUF	100/50	450		70		6
PUR	n/a	140		20		2.5
EPI	100/15	300		25		2.5

To evaluate the adherend surface in cross-sectional view and to analyze adhesive penetration, transmitted light microscopy was applied. For this, small blocks with a cross-section of 20 mm by 10 mm and 10 mm length were obtained from tensile shear test specimens. Using a microtome, slices approximately 20 µm thick were cut from the end-grain surfaces and stained with safranin. The microscopic examination was performed with a Zeiss "Axiophot" with imaging software "Axio Vision 4.8.3.0".

2.3.2. Surface roughness

The surfaces were scanned with a Taylor-Hobson "Form Talysurf Series 2" profilometer. The measuring unit was equipped with a stylus tip with 90° cone angle and 2 µm tip radius. Measurements were carried out at 0.5 mm/s on a length of 48 mm. Data was logged with the Taylor-Hobson software "Ultra 4.1.7" at a frequency of 500 Hz. For each surfacing method, 10 specimens with four measuring lengths each were scanned across the grain. A cut-off length of 8 mm and a Gaussian filter were applied to calculate the three roughness parameters *Ra* (arithmetical mean deviation), *Rp* (maximum peak height) and *Rv* (maximum valley depth) according to ISO 4287 [27]. In addition, the material ratio curve following ISO 13565-2 [28] was used to determine the three roughness parameters *Rk* (core roughness depth), *Rpk* (reduced peak height) and *Rvk* (reduced valley depth).

2.3.3. Tensile shear test

The tensile shear test was performed according to EN 302-1 [22]. For each surface and adhesive combination, eight bonded assemblies consisting of two panels were prepared. From each assembly, 10 shear test specimens (Fig. 1) were obtained, resulting in 80 specimens for each combination. Half of the specimens underwent treatment A1 and A4, as specified in EN 302-1 [22]. Treatment A1 involves testing in dry condition and is taken as a reference. Treatment A4 comprises 6 h boiling and 2 h water storage at 20 ± 2 °C before testing the specimens in wet condition. The shear test was performed with a TesT "Modell 112" universal testing machine using a constant displacement rate so that failure occurred after 60 ± 30 s. The tensile shear strength $f_{v,t}$ was calculated as follows:

$$f_{v,t} = \frac{F_{max}}{A} \quad \text{(MPa)}, \tag{1}$$

where F_{max} is the applied load at failure and A is the area of the shear plane. In addition, the wood failure percentage (*WF*) was visually estimated to the nearest 5%.

2.3.4. Delamination test

The resistance to delamination was determined according to EN 302-2 [23]. For the delamination test, 24 bonded members consisting of six lamellas and five gluelines were prepared. This corresponds to two bonded members for each surface and adhesive combination. The bonded members had a cross-section of 180 mm (height) by 150 mm (width) and a length of 500 mm. Four

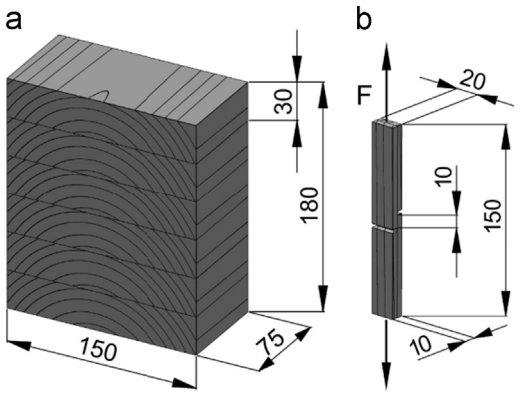

Fig. 1. Delamination test specimen (a) and tensile shear test specimen (b) with notches cut into the adherends, resulting in a shear plane of 10 mm by 20 mm

delamination test specimens with 75 mm length were obtained from each bonded member (Fig. 1). Two specimens each from one bonded member were tested in the high and the low temperature procedure according to EN 302-2 [23]. The two procedures serve as proof for the durability of adhesive bonds under different environmental conditions: the high temperature procedure (HT) is applied to verify the suitability under outdoor conditions and the low temperature procedure (LT) is used when products shall be installed indoors or in sheltered outdoor conditions. In both procedures the test specimens are impregnated with water using the same pressure levels. Subsequently, the swollen specimens are dried at different temperatures and RH. After the last cycle (HT: 3 cycles, LT: 2 cycles), the delamination percentage is calculated by dividing the open glueline lengths by the total glueline lengths measured on both end grain surfaces of the specimens. Both delamination values for each glueline (D_{GL}) and for specimens (D_{SP}=mean delamination of five gluelines within one specimen) have been determined.

3. Results and discussion

3.1. Microscopy

Planed surfaces showed very little damage in SEM micrographs (Fig. 2a). The anatomical structure could easily be identified. Large vessels and low-porosity fibers as well as pits in vessels and fibers were clearly visible. Ray cells and parenchyma cells alongside the vessels were cut open and showed exposed lumens, thus possibly facilitating adhesive penetration. The level of fibrillation seemed to be lower than that reported earlier by Hernández and Cool [21] with birch wood. The utilization of freshly sharpened knives for surface preparation may have contributed to this low fibrillation. In addition, the relatively high density of ash wood may be a further reason for the limited fibrillation, given that density was found to influence both fibrillation as well as cell damage [16,20]. As fibrillation is considered beneficial for adhesion, the absence of torn-out fibrils may have negative consequences for planed surfaces.

Closer examination of the xylem in cross-sectional view by means of transmitted light microscopy (Fig. 2b) confirmed that damage was insignificant also in subsurface areas. This agrees with results from previous studies on other wood species [10,17]. Despite that many cell lumens were cut open and consequently provided good access for adhesive, penetration depth was low with typically between 0 and 2 cells (fibers) for planed surfaces (Fig. 2b, upper adherend). In the lower adherend up to 5 cells were filled with adhesive which was assumingly caused by local grain deviation and the application of adhesive on this surface. Thus, the two adherends in Fig. 2b represent

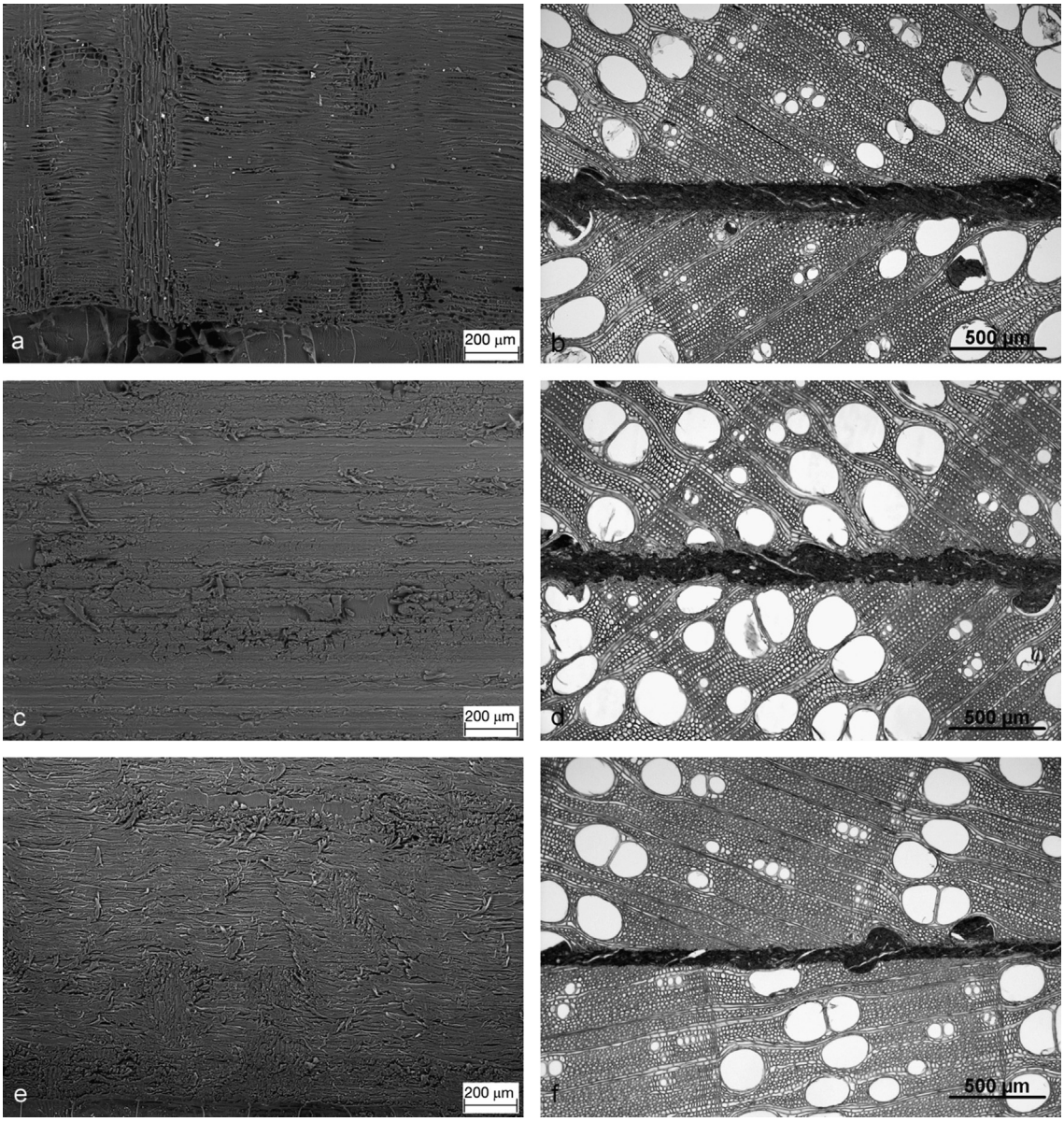

Fig. 2. Scanning electron micrographs of radial surfaces (left) and transmitted light micrographs, showing the cross-sections for the MUF bonded surfaces (right). Surfaces were prepared by planing (a and b), sanding (c and d) and face milling (e and f).

the possible spectrum of penetration depths for planed ash surfaces. Very little penetration was found in vessels which can be explained with the long closed assembly time that is associated with increased viscosity of the adhesive. The fractures in the glueline (Fig. 2b, d, and f) were most likely caused during cutting of the slices for microscopy.

In comparison to planed surfaces, sanding produced substantial cell damage. As a result, anatomical features such as fibers and rays were barely identifiable in SEM micrographs (Fig. 2c). Vessels were partly visible and often filled with fibrils and cells detached by the abrasive surfacing process. The appearance of sanded surfaces ranged from very smooth in areas where the coarse abrasive grit obviously flattened the wood tissue to quite rough in areas where considerable damage and fibrillation could be observed. This resulted in a highly irregular surface in comparison to planed surfaces.

In the cross-section (Fig. 2d), the degree of cell damage became visible. A layer of crushed and deformed cells both in surface and subsurface areas could be identified, creating a barrier that limited adhesive penetration. Consequently, penetration was significantly less than for planed surfaces. Only on rare occasions adhesive was found to pass through the layer of crushed cells and penetrate into the sound wood below. Generally, the irregular surface texture in combination with detached fibrils and torn-out cell fragments in turn provided a larger interfacial surface available for adhesion. These findings for ash are in accordance with results from Murmanis et al. [10] and de Moura and Hernández [12] for Douglas fir and sugar maple wood, respectively.

Face milled surfaces showed significantly lower cell damage than sanded samples (Fig. 2e). Vessels usually were cut open and free of debris. Although fibers and rays could be identified, cell lumens were not exposed as observed for planed surfaces which indicated at least some cell damage. Furthermore, face milling produced a significantly higher level of fibrillation than planed surfaces. Thus, packets of microfibrils were obviously partly detached during surface preparation and moreover, frequently showed a deflection perpendicular to the grain that corresponded to the cutting direction.

In cross-sectional view (Fig. 2f), cell damage could be found in the topmost cell layer. The discovered damages were deformed cell walls and detached fibrils and cell fragments rather than

crushed cells as were found for sanded surfaces. Earlywood fibers with thin cell walls were more severely deformed than latewood fibers, which corresponds to results for black spruce from Cool and Hernández [7]. Subsurface damage was not observed. With face milled surfaces, adhesive penetration was lower than with planed surfaces. Even though a layer of crushed cells could not be observed, slightly deformed cells in combination with fibrillation may have reduced penetration.

3.2. Surface roughness

The results of the surface roughness measurements are shown in Table 3. In addition, exemplary surface profiles in Fig. 3 display differences between the three surfacing methods. Planing produced a plateau-like surface (Fig. 3) that was associated with the lowest mean roughness *Ra* and core roughness depth *Rk* (representing the processing roughness according to Westkämper and Schadoffsky [29]). In addition, the peak values *Rp* and *Rpk* were significantly lower confirming results from the microscopic examination (Fig. 2) which showed an even surface with very little damage and fibrillation. The surface profile moreover showed recurring valleys which were related to large earlywood vessels and therefore, to the anatomic structure. This corresponds to the interpretation of *Rvk* by Westkämper and Schadoffsky [29].

The comparison of sanded and planed surfaces revealed significant differences for all roughness values. Sanded surfaces showed higher roughness *Ra* and *Rk*, and associated with this, a larger surface available for adhesion as already observed in the microscopic examination (Fig. 2d). The surface profile had a uniform appearance with few pronounced peaks and valleys. Deep valleys did not stand out so that the interpretation of *Rvk* as the anatomical component seems inappropriate for sanded surfaces. Furthermore, the sanding process caused high values for *Rp* and *Rpk*. These may have resulted from a lower positioned mean line as suggested by Cool and Hernández [7] and not necessarily from fibrillation as proposed for *Rpk* by Westkämper and Schadoffsky [29]. In addition, roughness values do not take subsurface damages into account even though this may have an impact on the bond performance because of the limited penetration or the creation of a MWBL.

Ra and *Rk* values for face milled samples were in-between those of planed and sanded surfaces. In comparison to planed surfaces, face milling produced a higher level of fibrillation (Fig. 2e) which resulted in a less plateau-like surface profile. Instead, recurring peaks could be observed (Fig. 3) which led to increased *Rp* and *Rpk* values and presumably to an enlarged adhesion surface. Depth and occurrence of valleys could be attributed to large earlywood vessels. In addition, no significant differences were found for *Rv* and *Rvk* between planed and face-milled samples. These consistent results suggest that *Rvk* is a good measure for anatomical structure both for face milling and planing.

Table 3
Surface roughness of ash specimens prepared with planing, sanding and face milling.

Roughness parameters	$\bar{x}$ (μm) (sd (μm))					
	Planing		Sanding		Face milling	
Ra	11.8 (3.2)	A[a]	16.2 (1.6)	B	13.1 (4.0)	A
Rp	20.1 (4.0)	A	57.6 (13.9)	B	40.6 (7.8)	C
Rv	107.3 (13.7)	A	85.6 (13.8)	B	103.5 (23.5)	A
Rk	9.9 (2.6)	A	47.2 (4.4)	B	19.2 (3.8)	C
Rpk	4.5 (2.0)	A	18.1 (5.8)	B	15.7 (4.0)	B
Rvk	57.3 (14.0)	A	37.3 (8.8)	B	64.1(18.9)	A

[a] Roughness values were compared by means of variance analysis and Scheffe post-hoc test using a 0.05 significance level. Different letters for surfacing methods indicate statistically significant differences in roughness.

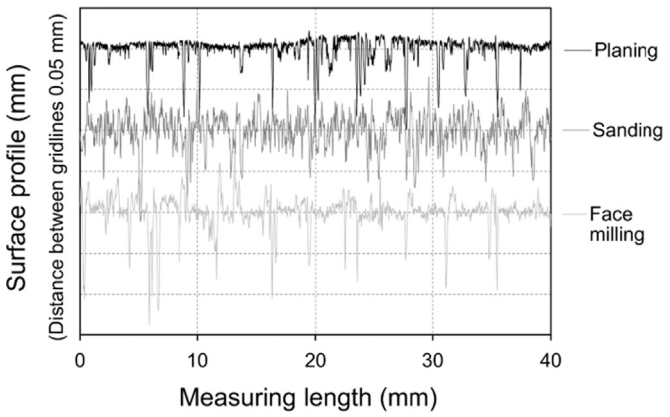

Fig. 3. Surface profiles of planed, sanded and face milled specimens.

Planed and face milled surfaces can be described well using the abovementioned surface roughness parameters. As long as subsurface cells are undamaged, roughness seems to be a good indicator for adhesion purposes. However, interpretability is limited with respect to adhesive accessibility of cells and therefore for penetration behavior. For sanded surfaces, interpretability of roughness parameters seems limited for adhesion purposes because of crushed wood cells on the surface.

3.3. Tensile shear test

Specifications for mean tensile shear strength are given in standards [24-26] for bonds made with beech wood (*Fagus sylvatica* L.). The requirements for treatments A1 and A4 are $f_{v,t,mean} \geq 10$ MPa and $f_{v,t,mean} \geq 6$ MPa, respectively. No requirements apply for *WF* and if wood species other than beech are used, the minimum values for $f_{v,t,mean}$ are not compulsory but rather serve as reference values. The results for $f_{v,t}$ (mean and CoV) and *WF* together with outcomes of statistical analyses are shown in Table 4. To illustrate the performance of the adhesives relative to each other and for the different surfacing methods, reference is made to Fig. 4. After A1, $f_{v,t,mean}$ was higher than 10 MPa for all adhesives and surfacing methods. Even though strength values vary with adhesives and surfaces, the results suggest a good bond quality. As expected, bond strength was lower after A4 because of the moisture influence on wood and wood–adhesive bonds. However, only with PRF bonded samples as well as with the MUF-bonded sample with sanded surfaces $f_{v,t,mean} \geq 6$ MPa was obtained. The results indicate that ash bonds were severely impaired by moisture changes in A4. Based on the facts that all adhesives included in this study have been approved according to European standards and thus have reached the required value of $f_{v,t,mean} \geq 6$ MPa with beech and, in addition, shear strength of ash solid wood is on average higher than that of beech [30,31] higher bond strengths with ash were expected. However, apparently ash bonds are more severely impaired by moisture changes than adhesive bonds with beech.

PRF bonded samples showed the highest shear strength both in dry (A1) and wet (A4) test condition (Table 4). The difference between samples bonded with PRF and with other adhesives was statistically significant for planed surfaces and for face milled surfaces after A4. In addition, the decrease in strength after A4 was lower for PRF (44.4–50.6%) than for MUF, PUR and EPI (51.3–55.9%). In general, MUF, PUR and EPI bonded samples showed a similar strength level.

The analysis of *WF* showed consistently high mean values ≥85% for PRF, MUF and EPI after A1. For PUR bonded ash, *WF* highly depended on the surfacing method. WF_{mean} varied between 33% and 94%, with differences between the surfaces being

statistically significant. *WF* for PRF bonded samples after A4 corresponded to the high values after A1 and can be explained with the high moisture-durability of PRF adhesives [32]. In contrast, treatment A4 caused a significant decrease of *WF* and at the same time, an increase of adhesion failure for MUF, PUR and EPI. For these adhesives, the *WF* strongly depended on the surface preparation.

Table 4
Shear strength $f_{v,t}$ (mean and CoV) together with mean *WF* for ash bonds depending on surfacing method, adhesive and treatment according to EN 302-1 [22].

Treatment	Surface	Adhesive	$f_{v,t,mean}$ (MPa) (CoV (%))			WF_{mean} (%)		
A1	Planing	PRF	14.53 (9.0)	A[a]	a[b]	89	A[a]	ab[b]
		MUF	11.10 (18.7)	B	a	92	A	b
		PUR	11.84 (10.2)	B	a	33	B	c
		EPI	11.76 (21.0)	B	ab	88	A	a
	Sanding	PRF	12.61 (13.9)	A	b	87	AB	b
		MUF	11.16 (15.9)	BC	a	93	A	b
		PUR	12.27 (15.6)	AB	a	62	C	b
		EPI	10.85 (11.3)	C	b	85	BC	a
	Face milling	PRF	13.60 (13.0)	A	ab	94	AB	a
		MUF	11.52 (10.6)	B	a	98	A	a
		PUR	12.74 (11.7)	AB	a	94	A	a
		EPI	12.74 (21.0)	AB	a	89	B	a
A4	Planing	PRF	7.18 (24.9)	A	a	90	A	a
		MUF	4.99 (11.5)	B	c	12	B	b
		PUR	5.61 (12.7)	B	a	5	C	c
		EPI	5.46 (22.1)	B	a	16	BC	b
	Sanding	PRF	7.01 (11.4)	A	a	92	A	a
		MUF	6.29 (20.2)	AB	a	50	B	a
		PUR	5.98 (13.2)	B	a	40	B	a
		EPI	5.15 (20.4)	C	a	24	C	a
	Face milling	PRF	7.34 (23.2)	A	a	96	A	a
		MUF	5.52 (11.6)	B	b	64	B	a
		PUR	5.86 (15.3)	B	a	29	C	b
		EPI	5.62 (17.1)	B	a	37	C	a

Shear strength values were compared by means of variance analysis and Scheffe post-hoc test. *WF* values did not follow a normal distribution for which reason the non-parametric Kruskal–Wallis test and Mann–Whitney *U* post-hoc test were used. The level of significance was set at 0.05 for all statistical tests.
Different letters indicate statistically significant differences in shear strength or *WF*.

[a] Uppercase letters were used for comparison of adhesive performance within one treatment and surfacing group.

[b] Lowercase letters were used for comparison of surfacing methods within one treatment and adhesive group.

When comparing the bond performance of the three surfacing methods, planed surfaces generated both the highest $f_{v,t,mean}$ (PRF—A1) and the lowest $f_{v,t,mean}$ (MUF/PUR—A1/A4). Also, the PUR bonded planed sample showed a low WF_{mean} of 33% when tested in dry condition. After A4, planed surfaces with their plateau-like texture and little fibrillation (Fig. 3) provided low resistance to adhesion failure with MUF, PUR and EPI. For these adhesives, the lowest WF_{mean} was determined, with differences to other surfaces being statistically significant. Adhesion failure primarily occurred in areas where latewood is predominant, thus emphasizing the importance of the surface texture and core and peak roughness in these areas. The increased contact area because of adhesive penetration (Fig. 2b) did not prevent adhesion failure in the interface.

Results with sanded surfaces were heterogeneous and therefore could not clearly be linked to surface texture or roughness. For example, sanding produced a highly irregular surface with a large interface available for adhesion (Fig. 2d). The higher roughness in combination with a significant level of fibrillation may have contributed to the highest $f_{v,t,mean}$ for MUF and PUR after A4. In contrast, the lowest $f_{v,t,mean}$ were determined for PRF and EPI both after A1 and A4 (statistically significant for EPI after A4). In these cases, a MWBL may have been created during surface preparation (Fig. 2c and d). This, in combination with an increase of cell damage because of moisture change as determined by Jokerst and Stewart [13] for Douglas fir and southern pine may have accounted for a lower strength. In addition, cell damage may also have favored the increased *WF* (PUR—A1, MUF, PUR, EPI—A4) as suggested by Jokerst and Stewart [13].

Face milled surfaces clearly showed the best results in the shear test. Except for the PRF bonded sample, the highest $f_{v,t,mean}$ was obtained after A1. In addition, the highest WF_{mean} was determined for face milled surfaces with all four adhesives after A1. In particular, *WF* was enhanced with 94% for PUR bonds, thus reaching the *WF* level of the other adhesives in dry tests. This improvement for PUR bonds

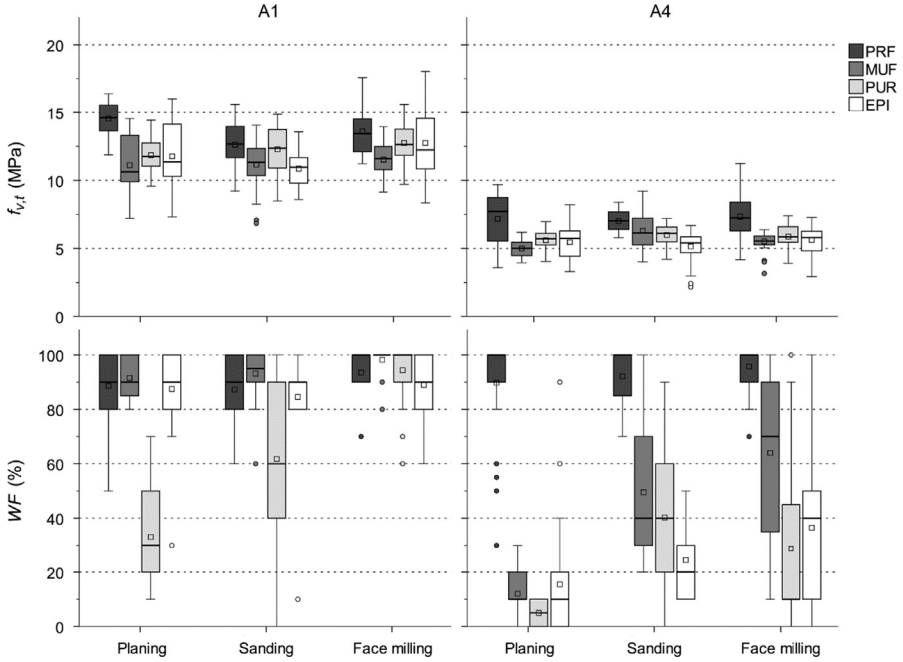

Fig. 4. Shear strength $f_{v,t}$ and *WF* for ash bonds depending on the surfacing method, adhesive and treatment according to EN 302-1 [22]. Boxplot elements: box = values between the 25th and 75th percentile (interquartile range); horizontal line = median; whiskers = lowest/highest values, circles = outliers beyond 1.5 times the interquartile range; squares = arithmetic mean.

after face milling could also be verified in statistical tests. The good test results after A1 could be confirmed by high $f_{v,t}$ and *WF* after A4 when compared to the other surfaces. Face milling produced a significant level of fibrillation without creating cell damage in the subsurface. This combination obviously contributed to a good bond quality and apparently provided a higher resistance to moisture treatment than planed and sanded surfaces.

The results did not show a clear correlation between roughness and $f_{v,t}$ or *WF*. This is in agreement with findings from Kläusler et al. [8] with beech wood. As described above, one reason for the limited correlation may be that subsurface damage, as observed for sanded surfaces, cannot be quantified in roughness measurements. However, with limited subsurface damage, specific roughness parameters such as *Rk* (processing roughness) or *Rpk* (fibrillation) may help to explain differences between planed and face milled surfaces. For these surfaces, a trend was observable towards higher $f_{v,t,mean}$ and WF_{mean} with increasing *Rk* and *Rpk* values. This trend applied for all four adhesives.

3.4. Delamination test

The delamination values have been evaluated with regard to standard requirements [24-26]. Accordingly, the delamination D_{SP} may not exceed 5% after HT and 10% after LT, for any specimen within an adhesive/surface sample. Therefore, the specimen with the highest delamination ($D_{SP,max}$) is decisive for evaluation. After HT, the best results were obtained for PRF with face milled ($D_{SP,max}$=6.1%) and sanded surfaces ($D_{SP,max}$=6.8%). MUF showed the best performance with sanded surfaces ($D_{SP,max}$=13.7%) whereas PUR and EPI produced best delamination results with face milled surfaces (PUR: $D_{SP,max}$=45.2%; EPI: $D_{SP,max}$=30.3%). It can be concluded that after HT the requirements of the standards could not be met by any combination of adhesive and surfacing method. After LT, the delamination was generally lower than after HT which can be explained by the less severe drying parameters and the lower number of test cycles. However, the general picture of the test results for adhesives and surfaces showed good agreement between HT and LT (Fig. 5). Again, PRF showed the highest resistance to delamination after LT for all three surfaces (Planing: $D_{SP,max}$=4.4%; Sanding: $D_{SP,max}$=2.9%, Face milling: $D_{SP,max}$=4.7%). In addition, the standard requirement was met with sanded surfaces bonded with MUF ($D_{SP,max}$=4.5%). Therefore, the condition for use in indoor or sheltered outdoor applications was fulfilled by four adhesive/surface combinations. PUR and EPI again performed best with face milled surfaces (PUR: $D_{SP,max}$=33.6%; EPI: $D_{SP,max}$=11.6%), nevertheless did not meet standard requirements.

PRF and MUF generally provided higher resistance to delamination than PUR and EPI in both test procedures (Fig. 5). This is supported by statistical analyses as shown in Table 5. For a better understanding of moisture-related durability of wood–adhesive bonds Frihart [32] developed a model that links bond durability with strain in the glueline caused by moisture change. This model differentiates between two adhesive groups, with PRF and MUF belonging to the group of “in situ polymerized” adhesives and PUR and EPI being “pre-polymerized” adhesives. Unlike “pre-polymerized” adhesives, PRF and MUF are able to diffuse into the cell walls before curing. This allows for a stabilization of the wood interphase (“region from the wood surface to the maximum penetration depth of the adhesive” as defined by Frihart [32], p. 614) and for a decrease of stress concentration in the interface in case of moisture change. Due to rigidity of these adhesives, strain is then mainly dissipated through the wood. In contrast, dissipation of interfacial strain for “pre-polymerized” adhesives takes place in the glueline because of a higher adhesive flexibility rather than in the wood. This model is in agreement with findings from Schmidt [33] who explained variations in fracture toughness after accelerated aging of PF-bonds with differences in molecular interpenetration into the cell walls between resins with varying molecular weights. An increased interlocking and a different stress distribution of PRF and MUF bonds may therefore have contributed to the higher resistance to delamination of ash assemblies. Considerably better delamination behavior of PRF and MUF bonds was also determined in combination with other medium-density hardwoods when compared to PUR or EPI bonds [34,35].

Closer examination of the differences between surfaces revealed the remarkable result that planing as the most common surfacing method in industry provided the lowest resistance to delamination for ash assemblies bonded with PRF, MUF and PUR (statistically significant for PUR). In contrast, EPI bonds showed a slightly better delamination behavior with planed surfaces than with sanded surfaces (Fig. 5).

When compared to planed surfaces, the delamination behavior was slightly improved for PRF and MUF with face milled surfaces. A main difference between planed and face milled surfaces was the level of fibrillation. Stehr and Johansson [9] suggested that fibrillation increases the adhesion surface area and fortifies the adhesive layer. However, the integration of fibrils into the PRF and MUF glueline showed only little improvement. This may be due to the fact that cured PRF and MUF adhesives are quite rigid compared to wood perpendicular to the grain. Strain as a consequence of moisture change is then distributed through the wood rather than in the glueline. In contrast, a more significant improvement in delamination performance was associated with sanded surfaces. One reason for this enhancement may be the higher roughness of sanded surfaces and therefore, a larger interface. Furthermore, the considerable subsurface damage caused by sanding assumingly helped to enhance the resistance to delamination. In case of moisture change, damaged cells adjacent to the interphase could have worked as a buffer zone and helped to dissipate strain away from the wood–adhesive interface. In general, the results obtained with PRF and MUF correspond well to the model of Frihart [32] as previously described. Consequently, the applicability of this model to interpret moisture-related durability can be confirmed for ash assemblies bonded with PRF and MUF.

The delamination behavior of PUR and EPI bonds is clearly related to the surfacing method. PUR bonds revealed a significantly improved delamination behavior with sanded surfaces and more notably, with face milled surfaces. EPI showed equally low resistance to delamination with planed and sanded surfaces. The resistance to delamination with face milled surfaces, however, was significantly improved, as could be confirmed in statistical analysis. PUR and EPI as “pre-polymerized” adhesives are not able to penetrate into cell walls [32] and develop gluelines with low stiffness. In case of moisture changes, the occurring stress is distributed by the adhesive because of its flexibility. The model of Frihart [32] seems applicable when comparing planed and face milled surfaces. Apparently, the higher fibrillation of face milled surfaces in comparison to planed surfaces led to a fortification of the adhesive layer. This helped to reduce the stress on the wood–adhesive interface and consequently, led to a higher resistance to delamination for both PUR and EPI. On a macroscopic level the stress becomes more uniform [6,9], whereas on the microscopic level the stress distribution may become more complex in the vicinity of semi-detached fibrils. Bonds with sanded surfaces did not show consistent results for PUR and EPI.

The correlation between roughness and delamination was limited. As for shear results, one reason for this is that subsurface damage is not taken into account in the roughness assessment. However, subsurface damage possibly contributes to a higher resistance to delamination with sanded surfaces bonded with PRF and MUF.

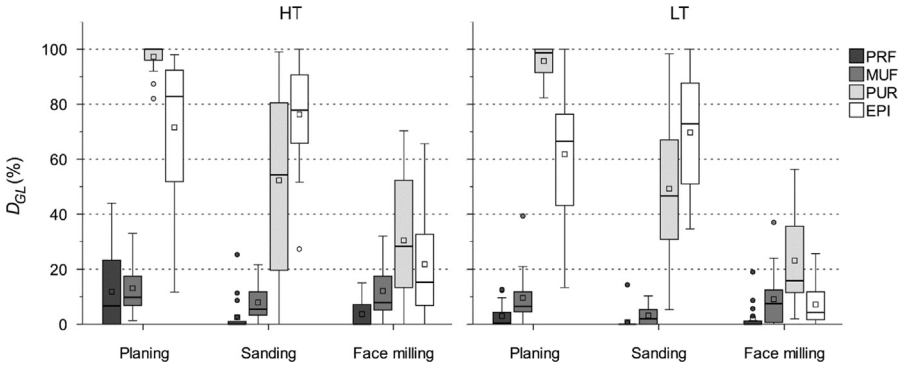

Fig. 5. Delamination D_{GL} of bonded ash assemblies depending on the surfacing method, adhesive and test procedure according to EN 302-2 [23]. Boxplot elements are explained in Fig. 4.

Table 5
Delamination $D_{GL,mean}$ of bonded ash assemblies depending on the surfacing method, adhesive and test procedure according to EN 302-2 [23].

Surface	Adhesive	$D_{GL,mean}$ (%) HT			LT		
Planing	PRF	11.8	A[a]	b[b]	3.0	A[a]	a[b]
	MUF	13.1	A	a	9.6	B	b
	PUR	97.3	C	b	95.7	D	c
	EPI	71.6	B	b	61.8	C	b
Sanding	PRF	2.5	A	a	0.8	A	a
	MUF	7.9	B	a	3.2	B	a
	PUR	52.3	C	a	49.3	C	b
	EPI	76.3	C	b	69.7	C	b
Face milling	PRF	3.7	A	ab	2.0	A	a
	MUF	12.1	B	a	9.1	B	b
	PUR	30.5	C	a	23.2	C	a
	EPI	21.8	BC	a	7.2	B	a

Delamination values were compared by means of the non-parametric Kruskal–Wallis test and Mann–Whitney *U* post-hoc test. The level of significance was set at 0.05.
Different letters indicate statistically significant differences in delamination.

[a] Uppercase letters were applied for comparison of adhesive performance within one surfacing group.

[b] Lowercase letters were used for comparison of surfacing methods within one adhesive group.

Face milling produced a higher processing (*Rk*) and peak roughness (*Rpk*) when compared to planed surfaces. Associated with this were a larger adhesion surface and consequently, an improved delamination resistance for all adhesives. These results show that roughness values *Rk* and *Rpk* can be used as an indicator for delamination performance as long as cell damage from surfacing is limited.

4. Conclusions

Bonded ash assemblies have been prepared with four adhesives (PRF, MUF, PUR and EPI) and moisture-related performance was determined in tensile shear and delamination tests. The wood components of the assemblies have been prepared using three surfacing methods (planing, sanding and face milling) and analyzed with respect to surface roughness and microscopy of the wood–adhesive interface. The following conclusions can be drawn:

- The surfacing methods resulted in significantly different bonding surfaces, notably with regard to cell damage and the level of fibrillation. Planed and face milled surfaces showed little damage and could be well characterized with surface roughness parameters. In contrast, sanding produced significant cell damage, leading to more complex bonds.
- The adhesive type had considerable influence on the performance of ash assemblies in both shear and delamination tests. The best moisture-related performance was determined with PRF. Based on the applied test methods and bonding parameters, PUR adhesive was the least performing adhesive.
- The surfacing method had significant impact on shear and delamination results. Planing, the most commonly used surfacing method, produced bonds with the lowest moisture resistance.
- Shear tests showed good bond performance in dry testing. Treatment A4 revealed differences between surfaces, in particular with regard to wood failure *WF*. Based on shear results, the most appropriate surfacing method to produce moisture-resistant bonds appeared to be face milling.
- In delamination tests, in-situ polymerized adhesives PRF and MUF showed highest resistance to delamination with sanded surfaces because damaged cells helped to reduce stresses. Pre-polymerized adhesives PUR and EPI provided the highest moisture-related durability with face milled surfaces as in this case a more homogenous stress distribution in the glueline is expected because of fibrillation.

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Paper III

Measurement of moisture-related strain in bonded ash depending on adhesive type and glueline thickness

Markus Knorz, Peter Niemz and Jan-Willem van de Kuilen (2015)

Holzforschung DOI: 10.1515/hf-2014-0324

Measurement of moisture-related strain in bonded ash depending on adhesive type and glueline thickness

Short title: Moisture-related strain in ash bonds

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Abstract: Structural wood-adhesive bonds (WAB) have to be durable while subjected to considerable stresses caused by mechanical loads and moisture content changes. To better understand the moisture-related durability of WABs, knowledge is important of how moisture changes generate strain in the bond. In this paper, strain on end-grain surfaces of bonded ash specimens was analyzed by means of digital image correlation. Strains were generated by wood shrinkage, and the evaluation was focused on shear strain (SStr). The bond lines were studied depending on the adhesive type – phenol resorcinol formaldehyde (PRF), melamine urea formaldehyde (MUF), polyurethane (PUR), and emulsion polymer isocyanates (EPI). Moreover, three different glueline (GL) thicknesses of MUF were taken into consideration. Comparing the adhesive types, SStr distributions (SStrD) were strongly influenced by adhesive elasticity. MUF and PRF bonds were quite rigid and were associated with pronounced strain amplitudes in and close to the GL together with strain dissipation reaching deep in the wood. PUR and EPI adhesives were more elastic and therefore allowed for smoother strain transition showing less distinct strain peaks. GL thickness had significant impact on SStrD. A high strain level and direct strain transition between adherends was found for the 0.01 mm GL, whereas a pronounced strain decrease was observed in the 0.1 and 0.2 mm GLs. This indicates different stress levels in the wood-adhesive interface dependent on GL thickness.

Keywords: adhesive, ash, digital image correlation, EPI, glueline thickness, moisture-induced strain, MUF, PRF, PUR

Introduction

Adhesive bonds in timber structures must be durable and stable. The durability of wood-adhesive bonds (WAB) is affected by the climate and can be impaired by induced swelling and shrinkage stresses in wood and WABs due to moisture changes. Stresses can develop at the beginning of the service life during the initial hygrothermal conditioning of the product when lamellas with different moisture content (MC) are adhesively bonded (Niemz et al. 2005). In addition, stresses can occur as a consequence of MC differences, for example, when lamellas with relatively high MC are bonded and MC decreases to a seasonal minimum in winter. Stresses are also generated when laminated timber is subjected to varying temperature and humidity during service life. When stresses exceed bond strength, delaminations can develop and affect the remaining service life (van de Kuilen and Gard 2013). Although the importance of moisture-dependent behavior of structurally laminated timber is recognized and has been examined in several studies (e.g., Aicher et al. 1998; Jönsson and Svensson 2004; Gereke and Niemz 2010; Angst and Malo 2012), the moisture-related durability (MRD) of WABs is still not well understood.

The level of moisture-induced stresses in WABs depends on various factors, such as the geometrical properties of the lamellas and the glued element as well as the wood species. With increasing density, shrinkage, and swelling coefficients as well as stiffness and strength properties, moisture changes generate higher stress levels (Marra 1992). Bonds between high-density wood have to tolerate higher moisture-induced stresses than those between lower-density wood. In addition, the strength difference between wood and WAB is lower for wood species with higher density; therefore, it is more likely that bond failure occurs than with low-density wood species. This effect gained importance when developing high-capacity glulam made of ash (*Fraxinus excelsior* L.) or beech (*Fagus sylvatica* L.), because these wood species have significantly higher densities than other woods typically used for glulam in Europe [e.g., spruce (*Picea abies* L. Karst.)]. Although the properties of ash and beech are beneficial for the load-carrying capacity, it is difficult to obtain high moisture-related bond durability (MRBD) with these species (Ohnesorge et al. 2010; Schmidt et al. 2010; Knorz et al. 2014). In the quoted studies, delamination tests were in focus that generate stresses on the WAB both perpendicular to the grain and in shear to evaluate MRBD. In addition, investigations with adhesively bonded hardwood (HW) were performed aiming at the determination of factors that influence MRD. On a microscopic scale, the glueline (GL) thickness with dimensions up to 0.2 mm (Knorz et al. 2014), the surface texture (Knorz et al. 2015), and the application of a primer (Kläusler et al. 2014) showed an impact on MRBD.

The results of the latter studies indicate that strain distribution (StrD) in the GL and in its vicinity has significant impact on MRBD. This was also proposed by Frihart and Wescott (2008) and Frihart (2009), who developed a model that explains the distribution of swelling strain in the case of moisture absorption for two different adhesive types. According to this model, “in-situ polymerized” adhesives, such as phenol resorcinol formaldehyde (PRF) and melamine urea formaldehyde (MUF), penetrate into the cell walls and stabilize them and, in addition, develop a rigid GL. Swelling strain is then distributed away from the wood-adhesive interface and dissipated in the wood. In contrast, “pre-polymerized” adhesives [e.g., polyurethanes (PUR) and emulsion polymer isocyanate (EPI)] do not penetrate into cell walls and produce a more flexible GL that

results in a different StrD. This model is supported by several experimental studies aiming at the analysis of adhesive penetration into cell walls (Gindl et al. 2002, 2004) and the determination of material properties of adhesives (Konnerth et al. 2006, 2007; Follrich et al. 2010; Clauß et al. 2011). Konnerth et al. (2010) and Kläusler et al. (2013) demonstrated that the mechanical properties of cured adhesives depend on their MC and therefore have to be considered in the case of moisture change. To investigate the influence of adhesive penetration into cell walls and cell lumens on the stress distribution in WABs, Gindl et al. (2005) compared PRF- and PUR-bonded spruce in compression tests perpendicular to the grain. It was shown that compression strain was reduced in the GL area compared to wood and that strain in the GL area was lower for PRF than for PUR. This is in agreement with the data of Müller et al. (2005), who observed higher deformation for PUR-bonded than for PRF-bonded lap joint shear specimen, and these results were explained with different Young's moduli of the adhesives.

The delamination tests with HW and several studies quoted above indicate that StrD has a significant impact on MRBD. However, little is known about the complex micromechanics in WABs and how strains are distributed as a result of moisture change. Therefore, the goal of the present study is to examine moisture-induced strain in adhesively bonded ash wood, taking different GL thicknesses in a relevant range between 0.01 and 0.2 mm and adhesive types (PRF, MUF, PUR, and EPI) into account. The specimens are designed with a primary focus on investigating shear strain (SStr) in the GL area. For strain measurements, digital image correlation (DIC) will be applied. DIC is well suited for the visualization of strains with a high spatial resolution on wood surfaces (Zink et al. 1995; Valla et al. 2011; Keunecke et al. 2012; Lanvermann et al. 2014a) and wood-adhesive joints (Serrano and Enquist 2005). The expectation of the present study is that a better comprehension of stress fields in WABs due to moisture changes will contribute to adhesive developments, numerical simulations of WABs, and a better understanding of the durability of laminated timber under varying climatic conditions.

Materials and methods

Defect-free ash boards were selected with tangentially (T) oriented growth rings and dimensions 40 x 185 x 550 mm^3 (thickness x width x length). After storage at 20 °C and 95% relative humidity (RH; 20/95) for approximately 8 weeks, the mean MC before bonding was 19.7% with 0.6% SD. The density averaged 618 kg m^{-3} with 46 kg m^{-3} SD (related to 12% MC). The boards were planed to a cross-section of 180 x 30 mm^2 and cut into pieces with 80 mm length. The members were assembled from three pieces (Figure 1) with the same growth ring orientation. This assembly agrees with the specimen layup applied in delamination tests according to EN 302-2 (2013) for MRBD evaluation. The members were bonded with four adhesives (PRF, MUF, PUR, and EPI), which are approved for load-bearing soft wood bonds in Europe. The GL thickness d_{gl} of 0.1 mm was generated by means of an aluminum frame for all four adhesives. In addition, MUF-bonded members with d_{gl} of 0.01 and 0.2 mm were prepared. From each bonded member, one specimen with 150 mm width, approximately 90 mm height, and 15 mm length (Figure 1) was obtained. For each adhesive and GL thickness, five specimens were tested, which corresponds to a total number of 30 specimens. The bonding parameters open assembly time (< 5 min), pressure (1.2 MPa), press time

(12 h), one-sided application, and bonding conditions (20/95) were constant for all adhesives. The amount of adhesive applied for the 0.01 mm MUF GL ($GL_{0.01}$) was 450 g m^{-2}. The 0.1 and 0.2 mm GLs ($GL_{0.1}$, $GL_{0.2}$) were prepared with an excess of adhesive. Further data of adhesive properties and processing parameters are summarized in Table 1.

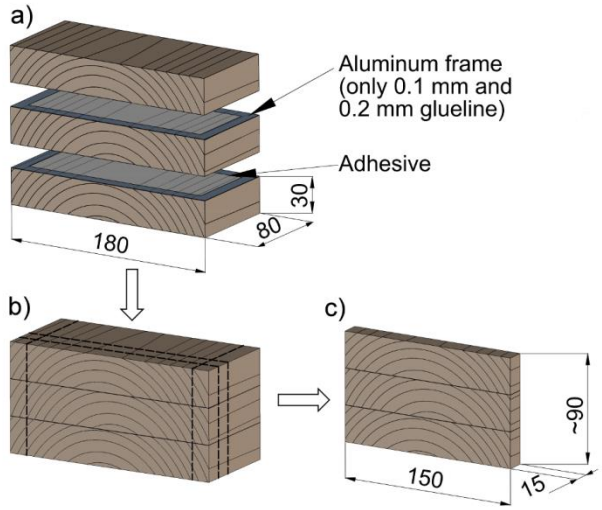

Figure 1 **Schematic drawing of specimen preparation: (a) Assembly of boards with application of adhesive and aluminum frame for defined GL thickness as required, (b) cutting of test specimen from bonded assembly and (c) test specimen.**

Table 1 **Adhesive properties and processing parameters for the four adhesives indicated.**

Adhesive	MUF	PRF	PUR	EPI
Ratio resin/hardener (-)	100/50	100/20	n/a	100/15
Solid content (%)[a]	50.2	55.8	100	66.4
Closed assembly time (min)	30 ($GL_{0.01}$) 70 ($GL_{0.1\text{-}0.2}$)	80	5	10

[a] Solid content for MUF, PRF and EPI was determined according to EN 827 (2005), value for PUR corresponds to the manufacturers' declaration

For strain measurement by DIC, an artificial speckle pattern was applied on one end-grain surface of the specimens. The approach to analyze end-grain surfaces corresponds to that of standardized delamination tests for MRD evaluation. The speckle pattern was generated with acrylic paint sprayed with an airbrush and comprised a white base layer and a layer of stochastically distributed dark speckles. The experimental setup consists of a specimen mount, two cameras, and cold light sources for illumination (Figure 2). The surface observed was aligned perpendicular to the optical axes (z) of the cameras. The specimens were placed on three supports and images were taken from underneath. This setup guaranteed a constant distance between measurement surface

and the cameras, thus avoiding errors because of dimensional change of the specimens in the z-direction. In the x- and y-directions (Figure 2), the specimens were positioned by means of aligning pins to provide consistency. The two cameras recorded monochrome images (2048 x 2048 pixels) of an overall specimen view (image size: 160.6 x 160.6 mm^2, pixel size: 78.4 x 78.4 μm^2) and a detailed view of a corner area (image size: 23 x 23 mm^2, pixel size: 11.2 x 11.2 μm^2), where high SStrs in the GL were expected (Figure 3a and c). Reference images showing the initial condition at 20/95 were taken and the specimen mass was determined. Then, the specimens were split in groups of five specimens and exposed to 20 °C and 40% RH (20/40) in a climate chamber.

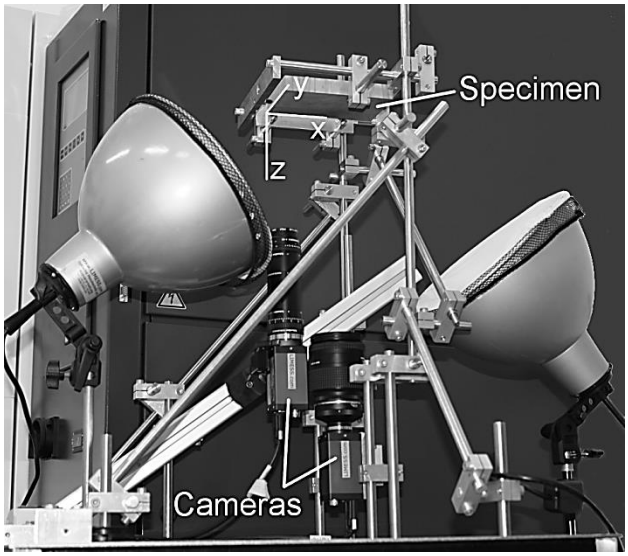

Figure 2 **Test setup with specimen mount with two cameras and cold-light sources for DIC measurements.**

After predefined intervals (1, 2, 4, 6, 8, 24, 72, and 144 h), images were acquired and the specimen mass was measured. Images were processed with the DIC software VIC 2D (Correlated Solutions, Inc., Columbia, SC, USA). The area of interest (AOI) was defined in the reference image for both views (Figure 3a and c) to narrow data down to the relevant content and to reduce file size and computing time. Based on the results from Valla et al. (2011), the parameters subset and step were specified in the software. Subset defines the size of a pixel array that is used to trace displacements between corresponding images based on the gray values of the speckle pattern and was assigned 21 (i.e., the array had a size of 21 x 21 pixels). For step, the values 3 for the overall view and 1 for the detailed view were selected. Step specifies the grid spacing; for example, step 3 means the correlations between images were calculated for every third pixel in the x- and y-directions (Figure 3a and c). The in-plane displacements *u* and *v* in the x- and y-directions were determined by cross-correlating the images. Based on these displacement values, strain values ε_{xx} and ε_{yy} and SStrs ε_{xy} (in mm mm^{-1}; the results in the following sections are given without dimension) were calculated (Figure 3b and d). The choice of the respective AOI and step value resulted in strain field sizes of 620 x 340 for overall view and 1800 x 1300 for detailed view. To retrieve the strain values from the GL area and the wood, the data were further processed with MATLAB

(The Mathworks, Inc., Natick, MA, USA; Figure 3e and f). The adhesive penetration behavior was evaluated by a reflected light microscope Leica MZ FLIII with the imaging software Leica Application Suite 4.1.0 (Leica Microsysteme Vertriebs GmbH, Wetzlar, Germany). For the observations in detailed view area, two specimens per adhesive and GL thickness were randomly selected.

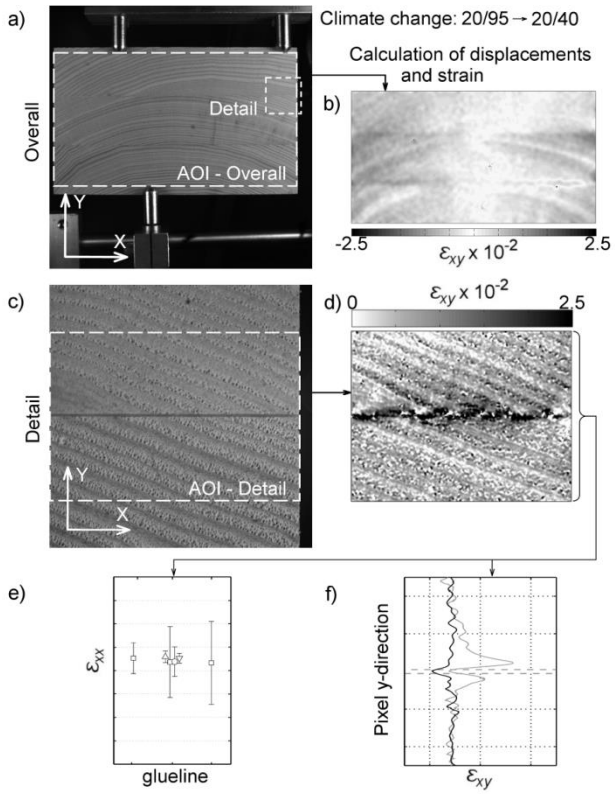

Figure 3 **Overall view (a) and detailed view (c) of the specimens with definition of areas of interest (AOIs) together with strain distributions after conditioning at 20/40 (b, d) and exemplary evaluation schemes (e, f).**

Results and discussion

Moisture content and strain development

During climate change from 20/95 to 20/40, the mean MC decreased from 19.7% to 8.2% (± 0.1%), which equals a mean MC change of 11.5%. The most significant decrease of MC occurred within the first 24 h after exposure to 20/40 and was presumably caused by the rapid drying of the surfaces. A MC change of only approximately 1% was observed between 24 and 144 h until equilibrium MC (EMC) was very likely reached after 144 h (Figure 4). The decrease of MC mean values for ε_{xx}, ε_{yy}, and ε_{xy} are presented over time separately for wood and one exemplary PRF GL. The strain values after 144 h for wood were retrieved from detailed view for all specimens and averaged -0.0296 (± 0.0029) for ε_{xx} and -0.0203 (± 0.0033) for ε_{yy}. The negative values reflect the shrinkage deformation. The mean SStr ε_{xy} after 144 h amounted to 0.0054 (± 0.0019) and was mainly caused by the displacements of pixels in the x-direction (*du*) relative to their y-position (*dy*). The greater variation of ε_{xy} was possibly

caused by an increased range of strain values covering high values in positions close to the GL and smaller values at some distance to the GL. In addition, a different deformation behavior between earlywood (EW) and latewood (LW) may have contributed to an increased variation of ε_{xy}. The development of strain on wood surfaces is due to shrinkage and therefore correlated closely with the decrease of MC. The close relationship between MC and strain development also applies for the GL area. Thus, the most significant changes are seen within the first 24 h after exposure to 20/40, whereas the changes are lower later on.

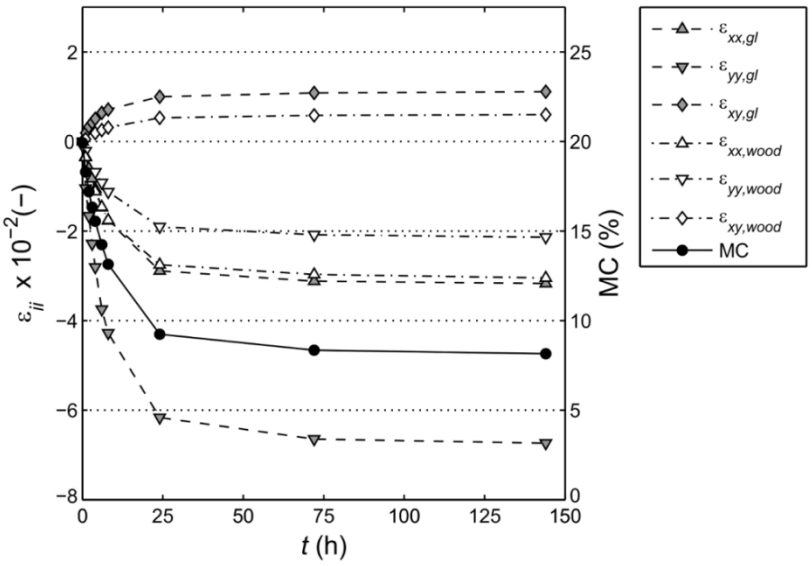

Figure 4 **Development of MC over time after climate change from 20/95 to 20/40 together with development of strain ε_{xx}, ε_{yy} and ε_{xy} on specimen surfaces in wood (mean values from all wood surfaces from detailed view excluding the GL area) and in the GL area (mean values for one exemplary PRF GL).**

From ε_{xx}, ε_{yy}, and ε_{xy} obtained after 144 h, strain in the radial (R; ε_r) and T (ε_t) directions were calculated by means of coordinate transformation:

$$\varepsilon_t = \varepsilon_{xx} \cos^2\theta + \varepsilon_{yy} \sin^2\theta + 2\,\varepsilon_{xy} \sin\theta \cos\theta \quad (1)$$

$$\varepsilon_r = \varepsilon_{xx} \sin^2\theta + \varepsilon_{yy} \cos^2\theta - 2\,\varepsilon_{xy} \sin\theta \cos\theta \quad (2)$$

where θ is the angle between the x-axis positioned at the wood-adhesive interface and the tangent aligned to the growth rings at the intersection point with the x-axis. As a result, ε_r and ε_t averaged -0.0178 (± 0.0027) and -0.0245 (± 0.0037), respectively. For comparison, literature-based shrinkage values were collected for ash. For this evaluation, the differential shrinkage β_d in the R and T directions was first calculated based on the maximum shrinkage values $\beta_{max,r}$ = 5% and $\beta_{max,t}$ = 8% (Kollmann 1951) and a fiber saturation level of MC_{FS} = 32.2% for ash (Popper and Niemz 2009). The calculation resulted in differential shrinkage values $\beta_{d,r}$ = 0.155%/% and $\beta_{d,t}$ = 0.248%/% and consequently in $\beta_{r,11.5\%}$ = 1.78% and $\beta_{t,11.5\%}$ = 2.86% shrinkage for an MC change of 11.5%. The comparison of experimental and literature-based values shows a good agreement of ε_r and $\beta_{r,11.5\%}$, whereas ε_t was lower than $\beta_{t,11.5\%}$. One reason for the difference between ε_t and $\beta_{t,11.5\%}$ is likely due to the specimen design. In the case of MC change, the bonds between lamellas with mainly T-oriented growth rings cause a

constraint situation predominantly in the T direction. Consequently, strain in the wood in the T direction is likely to be reduced due to this constraint. Furthermore, intraspecific variations in ash wood may have contributed to the observed difference.

Strain distribution (StrD) in overall view

The analyses in this and the following sections are based on strain values that were determined at the end of the 144 h test period, after an EMC at 20/40 was reached. In an overall view of AOI (Figure 3a), the differences in deformation behavior between samples bonded with different adhesives and GL thicknesses were minor. The representative StrDs for ε_{xx}, ε_{yy}, and ε_{xy} are displayed in Figure 5. The ε_{xx} StrD was homogeneous for the entire specimen surfaces (Figure 5a). In contrast, pronounced differences were observed in ε_{yy} strain fields that can be explained by the shape and position of the growth rings (Figure 5b). For comparison with strain values from the detailed view, the ε_{xx} and ε_{yy} values have been analyzed for wood areas (excluding the GLs) from the overall view. The mean ε_{xx} value amounted to -0.0320 (± 0.0024) and thus compared well to the mean ε_{xx} from detailed view [-0.0296 (± 0.0029)]. An even better agreement was found for the mean ε_{yy} values with -0.0202 (± 0.0031) for overall view and -0.0203 (± 0.0033) for detailed view.

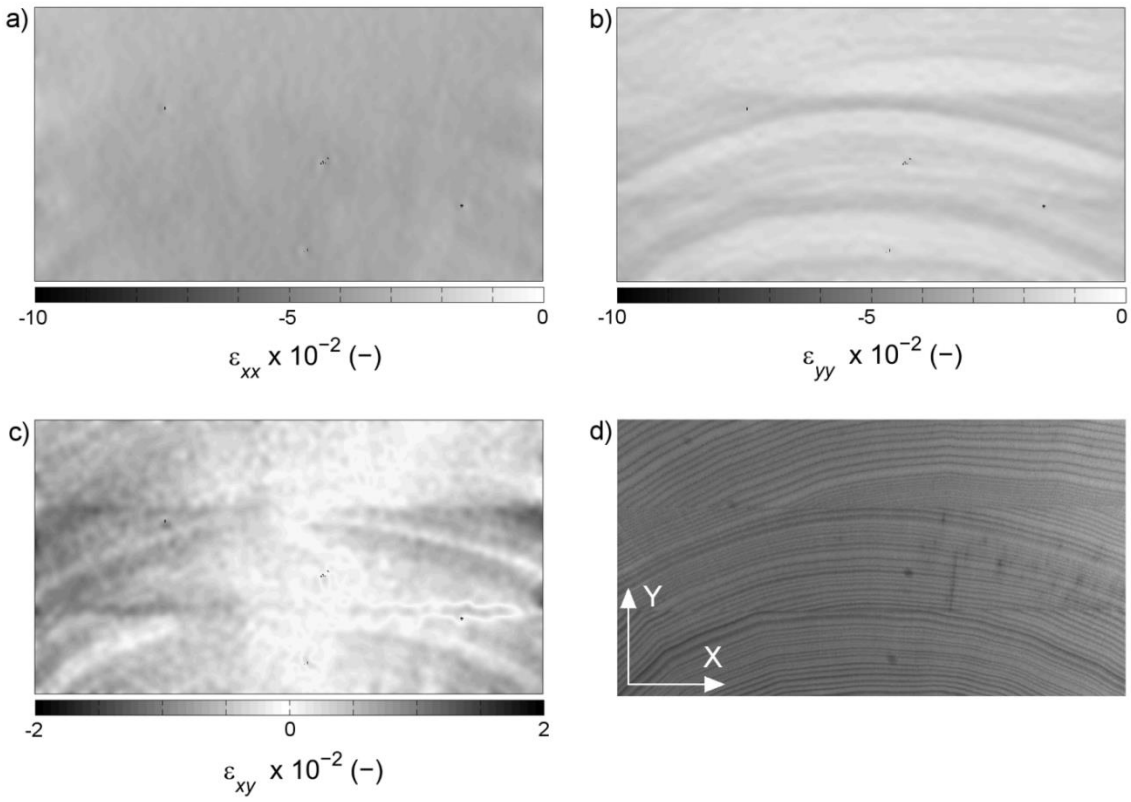

Figure 5 **Strain distributions for ε_{xx} (a), ε_{yy} (b) and ε_{xy} (c) from overall view AOI for one representative EPI-bonded sample together with the corresponding image of the test specimen before test (d).**

In both ε_{yy} and ε_{xy} strain maps, the position of the GLs could be visually identified (Figure 5b and c). However, no conclusive numerical analysis of strain in the GL was performed

with overall view data. This is because the cross-correlation does not allow for a clear differentiation between wood and GL data in the case of the pixel size of 78.4 x 78.4 µm^2 together with a higher grid spacing value (*step*: 3).

The ε_{xy} strain was inhomogeneously distributed in the x-direction. In the center of the specimens, a white or light grayish zone could be observed, representing no or little SStr. The SStr increased toward both ends of the GL near the specimen sides. In the wood itself, strains correspond to the shape of the growth rings (i.e., with the wood structure). The highest SStr values are in and close to the GLs. This picture confirms that the test results comply with theoretical considerations with respect to specimen design. Normally, a reduction of MC in lamellas with T-oriented growth rings would lead to cupping of the cross-section because of different shrinkage in the R and T directions and the curvature of annual rings. The adhesive between the lamellas now prevents such deformations, causing restraints toward the edges of the specimens in the GL.

Strains in glueline (GL) and wood in detailed view

Strains in GLs have been retrieved from detailed view strain maps and analyzed depending on adhesive type and GL thicknesses. For comparison, strain values were also evaluated for the wood surfaces. The wood strain data were retrieved from a part of the AOI (Figure 3c) that was at least 2 mm away from the wood-adhesive interface and therefore were assumed not to be directly affected by the bond. The transition zone between wood and GL is not taken into account for evaluation in this section. For all samples, the mean strain values and SDs were calculated. The results are displayed in Figure 6. In addition, the samples were compared by means of variance analysis and the post hoc test Dunnett-C, where required, at a 0.05 significance level.

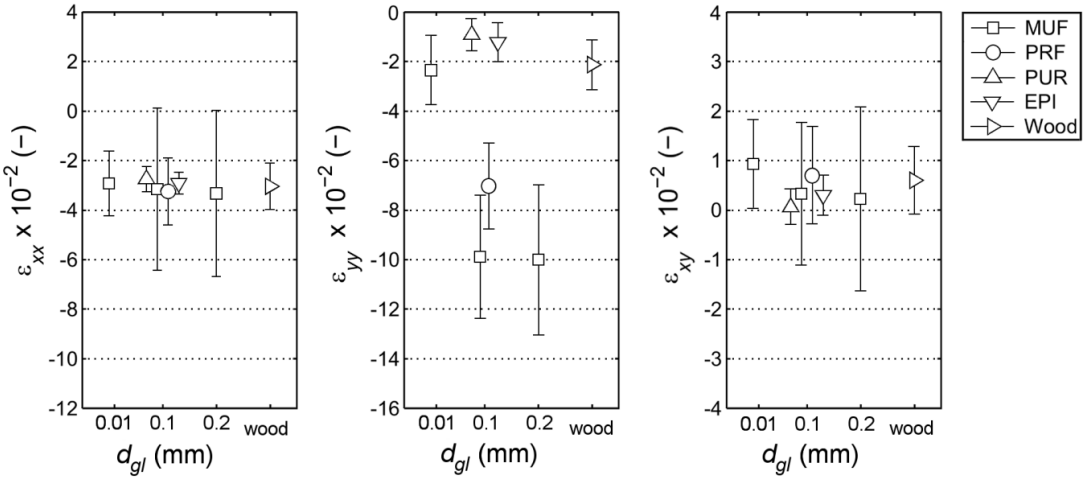

Figure 6 Strains ε_{xx}, ε_{yy}, and ε_{xy} (mean with error bars representing the SD) in the GLs in dependence on adhesive and GL thickness and in wood.

The mean values for ε_{xx} ranged between -0.0275 (PUR) and -0.0333 (MUF $GL_{0.2}$) and therefore deviated only little from ε_{xx} for wood (-0.0305). The comparison of strain in the

x-direction in the GLs shows no statistical difference between the four adhesives as well as between the GL thicknesses. Also, as can be seen from Figure 6, strains ε_{xx} in the GL are about equal to the shrinkage strains in wood. Although differences between mean values were insignificant, the SD varied considerably for the samples. For EPI and PUR GLs, the SD was low and strain was more homogeneous than in wood. In contrast, MUF GLs, particularly those with 0.1 and 0.2 mm thickness, showed a high SD.

In the y-direction, strain varied considerably between the samples and can be divided into three groups (Figure 6). Strain ε_{yy} for the MUF $GL_{0.01}$ ($\varepsilon_{yy,mean}$ = -0.0235) obviously was closely related to the behavior of the wood ($\varepsilon_{yy,mean}$ = -0.0214), and in a statistical sense, these two samples were similar. The closer examination of the images of MUF bonds with thin GL does not reveal a clear separation between wood and adhesive; thus, strain values represent a combined strain of adhesive with wood rather than for the isolated MUF GL. A separate analysis of wood and adhesive for these samples would require significantly smaller pixel sizes than available in the present study. ε_{yy} strain values were more homogeneous for PUR and EPI GLs but significantly lower than for wood and MUF $GL_{0.01}$. In contrast, strain in the y-direction was found to be significant for PRF with a mean ε_{yy} of -0.0703 and for MUF $GL_{0.1}$ and $GL_{0.2}$ with mean values of -0.0989 and -0.101. One factor that most likely contributed to this considerable shrinkage is the water sorption behavior of adhesives that varies considerably depending on their chemistry. For example, Wimmer et al. (2013) determined significant water uptake for formaldehyde resins (MUF 22% and PRF 18%), whereas PUR only absorbed 3.5% for a humidity range between 0% and 98%. In addition, formaldehyde resins are water based and moreover release water when curing. As bonding was performed at a relatively high wood MC, the water release from the GL may have been inhibited in the bonding process and cure shrinkage may have been limited. Postcuring shrinkage then may have occurred during conditioning at 20/40.

The mean SStr ε_{xy} ranged between 0.0093 for MUF $GL_{0.01}$ and values close to zero, 0.00065 for PUR. Although ε_{xy} varied considerably between wood, adhesives, and GL thicknesses, no significant differences between the samples were found in a variance analysis. The SStr ε_{xy} for MUF $GL_{0.01}$ was increased when compared to the strain level of wood and indicates elevated shear stress between the two adherends. In contrast, strain levels with $GL_{0.1}$ and $GL_{0.2}$ were similar (PRF) or significantly lower (MUF, PUR, and EPI) than in wood; therefore, a considerable influence of the GL is assumed.

In general, a higher SD of strain values could be observed for PRF and MUF bonds. This applies, in particular, for the $GL_{0.1}$ and $GL_{0.2}$. One reason for this is very likely that adhesives such as PRF or MUF can generate local strain maxima when compared to PUR because of different elasticities. This was found by Serrano and Enquist (2005) when comparing strain characteristics of adhesives with different stiffness in shear tests, and this can be confirmed in our tests when examining strain characteristics along the GL. However, Serrano and Enquist (2005) showed this behavior in tests with forces applied uniformly and parallel to the fiber direction, whereas strain in the present study was generated by the wood itself mainly due to shrinkage in the R and T directions. Therefore, the growth ring alignment and differences in shrinkage in the R direction within one growth ring, as determined by Lanvermann et al. (2014b) for spruce, may have influenced strain occurrences in our tests. In ash wood, differences within one growth ring with EW vessels with large diameters and fibers with thin cell walls and, in

contrast, LW cells with small lumens and thick and stable cell walls are more distinctive than for spruce. These intra-ring variations of ash wood therefore may have generated more pronounced strain peaks along the GL than what for example may be induced with spruce. Furthermore, cracks were found in the MUF $GL_{0.1}$ and $GL_{0.2}$ most likely as a consequence of restrained shrinkage of the rigid adhesive. Although the frequency of cracks was significantly lower than in UF GLs as observed by Hass et al. (2012), cracks in the MUF GLs contributed to the increased strain variance. Localized strain maxima next to cracks are most likely associated with stress concentrations that may possibly act as starting points for bond failure and thus reduce bond strength.

Shear strain distribution (SStrD) perpendicular to GL (detailed view)

For a detailed analysis of SStr ε_{xy}, average values were calculated across the detailed view AOI. The evaluation showed similar characteristics both for the distributions for PRF and MUF and for PUR and EPI. For reasons of clarity, only representative distributions that compare PUR and MUF as well as different GL thicknesses with MUF are displayed in Figure 7. The ε_{xy} distributions exhibited a homogeneous strain level for the surfaces that are beyond the sphere of influence of the bond. With increasing proximity to the GLs, significant changes of ε_{xy} can be found for some samples, thus indicating a considerable impact of the bond on the SStrD.

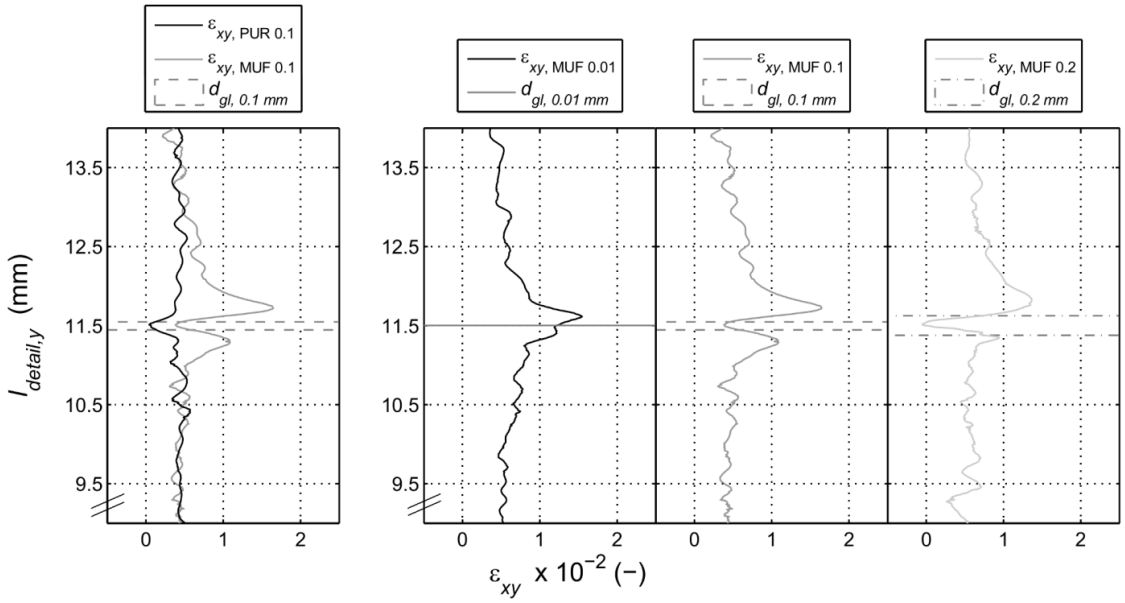

Figure 7 Representative strain distributions ε_{xy} in assemblies with homogeneous GL thickness for adhesive types MUF and PUR (left) and in MUF-bonded joints in dependence of GL thicknesses d_{gl} 0.01 mm, 0.1 mm and 0.2 mm (right).

When comparing the behavior of GLs with the same thickness (Figure 7, left), considerable differences between MUF and PUR can be observed. For example, strain values for the MUF-bonded sample started deviating from the strain level of the wood in significant distance to the GL, whereas the PUR-bonded sample showed an effect only

in close proximity to the GL. Also, a more pronounced strain decrease close to and in the GL could be found for MUF than for PUR. The minimum strain value in GL center, however, was lower for PUR than for MUF.

Obviously, the various adhesive types contributed to the differences in SStrD. For example, SStr in MUF bonds seems to be mainly distributed in the wood. This corresponds to theoretical considerations of Frihart (2009), who suggested that moisture-induced interfacial strain in PRF or MUF bonds is likely to be distributed in the wood and explained this with a stabilized wood surface due to the ability of these adhesives to penetrate into the cell walls together with a rigid GL where strain can barely be distributed. Referring to this, several research studies have shown that the stiffness of cured MUF and PRF adhesives is several times higher than of PUR or EPI (e.g., Konnerth et al. 2006; Clauß et al. 2011; Stoeckel et al. 2013) and, in addition, of ash wood in the R and T directions (Clauß et al. 2014). In contrast, only little effect on SStr can be found in the vicinity of the PUR GL. The reason for this is most likely that Young's moduli (E) of EPI and PUR are similar or lower (e.g., E_{PUR} = 330 MPa according to Clauß et al. 2011) than the elastic properties of ash wood in the T and R directions (E_t = 578 – 625 MPa and E_r = 1143 – 1193 MPa as determined by Clauß et al. 2014) and therefore allow for a smoother strain transition. This aspect indicates that stress concentrations can be avoided with PUR and EPI. When comparing the distribution of moisture-induced strain of different adhesives in GL vicinity, the influence of adhesive penetration has to be considered as shown by Gindl et al. (2005) for PUR- and PRF-bonded spruce.

Furthermore, it needs to be taken into account that the adhesives in our survey were processed at high humidity and high wood MC; therefore, MUF and PRF polymers presumably contained a significant content of water. Water in the cured adhesive polymer was observed to act as a softener and to reduce the stiffness (Konnerth et al. 2010; Kläusler et al. 2013). Accordingly, this may have influenced strain occurrences for these adhesives, for example, in terms of an increased minimum strain value in GL center. In contrast, bonding at lower humidity and MC as applied in industrial production might prevent a reduced adhesive stiffness and lead to even more pronounced strain occurrences.

Looking only at SStrDs, one may conclude that EPI or PUR is more suitable for structural bonding. However, the application of EPI and in particular PUR also involves drawbacks such as limited resistance against moisture in the wood-adhesive interface. For example, Kläusler et al. (2014) observed a loss of adhesion in PUR bonds in wet condition along with regained bond strength in re-dried specimens. Therefore, it is difficult to draw a general conclusion about the suitability of adhesives based on the SStrD only.

The SStrDs for MUF bonds with $GL_{0.01}$, $GL_{0.1}$, and $GL_{0.2}$ are significantly different (Figure 7, right). While increasing strain values were found for all thicknesses when approaching the GL, differences could be observed close to and in the GL. For $GL_{0.01}$, no strain decrease was seen in the GL area, whereas a significant strain decrease adjacent and in the GL as well as a reduced strain level in the wood-adhesive interface were noticed for $GL_{0.1}$ and $GL_{0.2}$ bonds. Comparing $GL_{0.1}$ and $GL_{0.2}$, strain decrease seems to be more pronounced and a lower minimum strain value in GL center was found for $GL_{0.2}$. This leads to the conclusion that the high adhesive stiffness relative to the wood has an increased effect on strain reduction in the GL center for a higher GL thickness. The direct strain transition together with comparatively high strain values indicates that the wood-

adhesive interface in $GL_{0.01}$ has to withstand stresses that are significantly higher than in the case of $GL_{0.1}$ and $GL_{0.2}$.

Although the effective stress levels cannot be quantified due to various reasons (e.g., microscale variabilities, relaxation, and plastic deformation), the different strain levels between the samples lead to the assumption that the GL thickness influences the MRD. These findings are a possible explanation for the dependence of MRD of ash bonds on GL thicknesses as determined by Knorz et al. (2014). Also, the results seem to be contrary to the common opinion that an ideal bond should exhibit a GL as thin as possible, at least when MRD testing is involved. As mentioned above, penetration possibly influences strain transition.

SStrD in GL vicinity (detailed view)

To evaluate further the dependence of SStrD ε_{xy} in GL vicinity from the adhesive type, the distances on both sides of the GL were determined, where the ε_{xy} strain level adjacent to the GL deviated from the average ε_{xy} strain level in the wood. These regions with deviating strain level were regarded as influenced by the bond. Two distance values $s_{def,1}$ and $s_{def,2}$ were determined for each GL based on the detailed view SStr diagrams (Figure 8). s_{def} is the distance between GL and intersection point of ε_{xy} fit-lines for unaffected wood and the peak area. The beginning of the peak area was specified before as the intersection point of the ε_{xy} curve with ε_{xy} fit-line for wood increased by 10%.

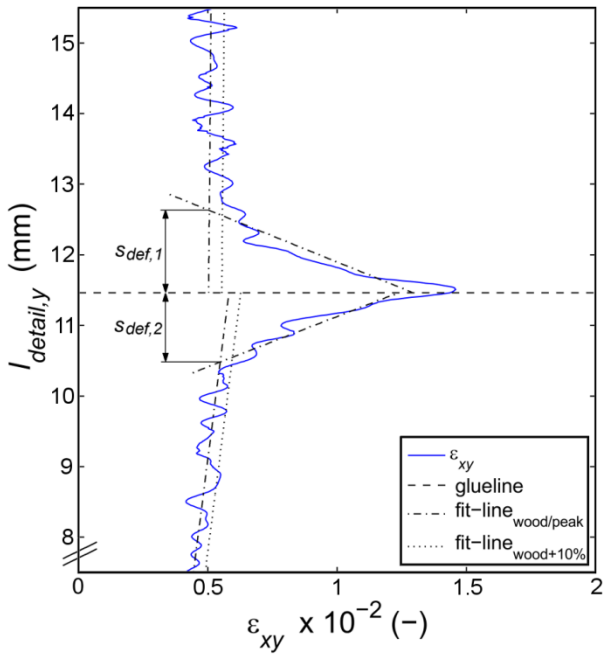

Figure 8 **Distances $s_{def,1}$ and $s_{def,2}$ in GL vicinity where increased shear strains are recorded, shown for a detailed view strain distribution of an exemplary MUF bonded specimen with 0.01 mm GL.**

The mean distances s_{def} together with their SD are displayed in Figure 9. These data for the four adhesives with $GL_{0.1}$ show considerable differences between MUF (1.1 mm) and PRF (1.28 mm), on the one hand, and PUR (0.19 mm) and EPI (0.17 mm), on the other hand. This is supported by the statistical analysis, where these two groups were found to be significantly different, whereas the data within the groups were statistically similar (variance analysis with Dunnett-C post-hoc test, $p < 0.05$).

Accordingly, this analysis confirms the observations in the previous section. The way of SStrD presumably varies with the elasticity of adhesives and their ability to diffuse into cell walls and stabilize them. The latter indicates that also the penetration behavior into the cell lumen network and from there into adjacent cell walls is of importance. Adhesive penetration as an influencing factor on StrD was also determined by Gindl et al. (2005), who found that the penetration of PUR and PRF mainly into spruce cell lumens changes the elastic properties of the wood-adhesive interphase and thus has an effect on StrD.

In this context, it would be important to know to which extent s_{def} values can be explained by adhesive penetration into ash wood. For this reason, the lumen filling portion was observed by microscopy. In fibers of EW and LW, no or very little penetration (up to three cell layers) was found for all adhesives and GL thicknesses. In contrast, the penetration in EW vessels varied strongly with the adhesives. The lowest penetration for $GL_{0.1}$ was found for MUF. Only individual vessels bordering the GL were filled with MUF adhesive and corresponded to a maximum penetration depth of 132 µm. Because long closed assembly times were used for both MUF and PRF, a limited penetration was expectable based on the data of Knorz et al. (2014). Nevertheless, the highest penetration depth in EW vessels of all adhesives was found in the case of PRF (841 µm). The maximum penetration depths for PUR and EPI were 353 and 360 µm, respectively.

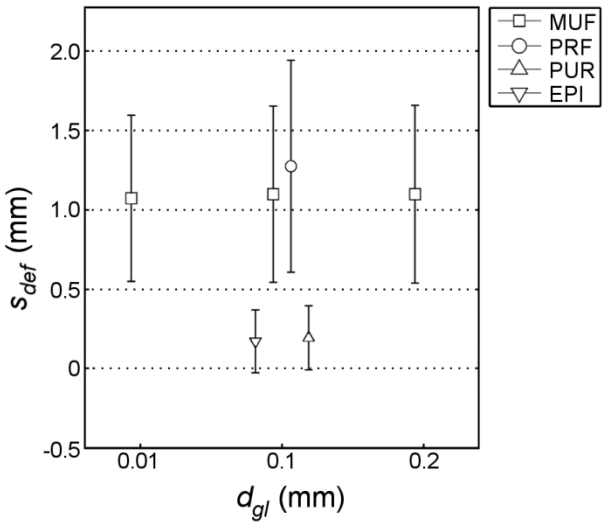

Figure 9 Distance s_{def} (mean with error bars representing the SD) in dependence on adhesive and GL thickness.

MUF bonds with different GL thicknesses showed statistically insignificant differences between s_{def} values ($GL_{0.01}$ 1.07 mm, $GL_{0.1}$ 1.1 mm, and $GL_{0.2}$ 1.1 mm). To obtain a thin GL, MUF bonds with $GL_{0.01}$ were prepared with a shorter closed assembly time than

bonds with $GL_{0.1}$ or $GL_{0.2}$ (Table 1). As a consequence of these varying bonding parameters, differences in adhesive penetration into EW vessels could be observed. For short closed assembly times, the adhesive penetrated up to 706 µm into the EW vessel network. In contrast, only cut-open EW vessels that adjoined the GL were filled with adhesive (up to 132 µm) in the case of longer closed assembly times.

The s_{def} values (i.e., the distances where strains are influenced by the bond) for MUF and PRF bonds were significantly higher than the maximum penetration depths of these adhesives. Thus, moisture-related strains extend deeper in the case of rigid adhesives than expectable by the sole penetration of the adhesives. In addition, s_{def} values were similar for MUF $GL_{0.01}$, $GL_{0.1}$, and $GL_{0.2}$, whereas penetration was significantly different. The previous data can be interpreted that SStrD in GL vicinity is independent from the penetration in EW vessels of ash. In contrast, the penetration depths for PUR and EPI exceed the mean s_{def} values. This indicates that either the influence of penetrated vessels on strain is insignificant or that strain transition is, in general, smoother due to similar or lower elasticity of these adhesives when compared to ash wood perpendicular to the grain.

Conclusions

The deformation behavior of bonded ash was investigated by means of DIC while the specimens were dried from 20/95 to 20/40. The ash lamellas were bonded with the adhesives MUF, PRF, PUR, and EPI with 0.1 mm GL thickness. In addition, $GL_{0.01}$ and $GL_{0.2}$ have been prepared with MUF.

DIC proved to be a valuable method to analyze 2D deformation on different scales. The analysis of strain in the GL in detailed view showed that ε_{xx} was significantly influenced by wood shrinkage. ε_{yy} differed strongly with adhesive and presumably depended on the ability of adhesives to absorb water or to store water from the preparation process.

SStrDs (ε_{xy}) were significantly different between rigid (PRF and MUF) and more elastic (PUR and EPI) adhesives. A more pronounced strain decrease in and close to the GL together with increased strain dissipation in wood regions was found for the rigid adhesives MUF and PRF.

A pronounced strain decrease was seen close to and in the MUF $GL_{0.1}$ and MUF $GL_{0.2}$, whereas, for the MUF $GL_{0.01}$, a direct strain transition together with higher strain values was found. This indicates a higher stress in the case of a very thin GL, which possibly explains the lower resistance to delamination of thin GLs.

SStr for MUF and PRF bonds was predominantly distributed in the wood and influenced wood regions that were significantly beyond adhesive penetration depths and regions where adhesive diffusion may have stabilized cell walls. In contrast, SStrD in the wood was limited for PUR and EPI adhesives.

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