Samuel Ammann* and Peter Niemz

Mixed-mode fracture toughness of bond lines of PRF and PUR adhesives in European beech wood

Abstract: The fracture behavior of bond lines in hard-wood has been studied. The joints of a phenol resorcinol formaldehyde (PRF) resin and a one-component polyure-thane (PUR) adhesive with European beech wood (*Fagus sylvatica* L.) adherends were examined for their fracture toughness (K_c). The initial crack tip was placed directly in the adhesive as a thin silicon film. Thereby, the exami-nation of the bond line, and not the solid wood, can be assured. Five different load angles were applied, from fracture mode 1 (M1) to mode 2 (M2), with an Arcan test mount. Additionally, three sample series conditioned at the relative humidities (RH) of 50%, 65%, and 95% of the surrounding air were tested. The results clearly show an increasing K_c of both adhesives with increasing shear stresses. This observation is valid for all RHs, but the differences decrease with increasing RH. The mois-ture dependency is more pronounced in PUR than in PRF glue joints. PUR generally shows a lower K_c than PRF, with the only exceptions being $K_{I,c}$ and $K_{II,c}$ in dry climate. The subsequent crack propagation in the PRF samples mainly takes place in the wood adherend, whereas, in the PUR samples, the cracks remain within the bond line (adhesive failure). Nevertheless, the performance of PUR glue joints is not worse than that of the solid wood, which can be attributed to the ductile behavior of the adhesive.

Keywords: adhesive failure, European beech, fracture toughness, glue joints, mixed-mode, one-component pol-yurethane (PUR), phenol resorcinol formaldehyde (PRF) resin

*Corresponding author: Samuel Ammann, Institute for Building Materials, Department of Civil, Environmental and Geomatic Engineering, ETH Zurich, HIF E 27, Stefano-Franscini-Platz 3, CH-8093 Zurich, Switzerland, e-mail: ammanns@ethz.ch
Peter Niemz: Institute for Building Materials, Department of Civil, Environmental and Geomatic Engineering, ETH Zurich, Zurich, Switzerland

Introduction

Hardwoods are gaining popularity in the European timber construction industry. The common hardwoods have a good potential to either locally support softwood con-structions at the zones of increased load or even replace softwood constructions by a slimmer design, on account of the higher values of their elementary parameters of strength and stiffness (Wagenführ 2007). However, due to the hygric nature of wood, higher mechanical properties also result in higher internal stresses caused by environ-mental changes, most notably changes in the surround-ing humidity. Therefore, in timber engineering, softwoods cannot simply be replaced with hardwoods based on their superior parameters, and the purpose of the construction and its local conditions also has to be taken into account.

Nowadays, wooden structural elements are mostly glued either simply to get the desired cross-section or to achieve a certain homogeneity within a wooden element. Such glue joints are crucial for a structure's safety. None-theless, the processes responsible for the performance of glue joints, especially in hardwoods, are not entirely apparent and are still a topic of many scientific investiga-tions (Yan et al. 2013). Most wood adhesives are designed for softwood and have proven their quality countless times. However, hardwood applications are rare due to the lack of knowledge and missing standards. Consequently, a better understanding of the bonding mechanics is nec-essary to produce reliable, adhesively bonded hardwood elements and to broaden the application of hardwoods in general.

A good approach to determine the safety of a structure would be a numerical simulation. However, the task to model the wood itself is already a considerable challenge (Hanhijärvi 1995; Ormarsson 1999; Serrano 2000; Dan-ielsson and Gustafsson 2011; Hering 2011; Qiu et al. 2014) that must be accompanied with simplifications due to the lack of experimental data and the complexity of the mate-rial itself. Therefore, most numerical investigations focus only on one specific problem. Danielsson and Gustafsson (2011) analyzed quadratic holes in glulam elements with a probabilistic approach and achieved results close to the experimental tests. Finger joints and their modeling

were elaborately investigated by Serrano (2000). Hanhijärvi (1995) analyzed the mechanosorptive behavior, and Ormarsson (1999) focused on the distortions induced by moisture changes. Crack propagation in solid wood was analyzed by Qiu et al. (2014). Another approach is modeling the materials themselves, not limiting to a specific application. Hering (2011), for example, developed an extensive material model [finite element method (FEM)] for beech wood and glued elements, but the work was focused only on the elastic range due to the complexity of wood and the numerous parameters.

As also stated by Danielsson and Gustafsson (2011), the application of existing standards (for softwoods) would also not deliver satisfactory results, since they are based on empirical values and their test setups are not based on realistic scenarios (EN 302-2 2013). Therefore, it is important to examine glue joints in hardwoods experimentally and from different perspectives for a reliable prediction of a structure's behavior and to guarantee its structural safety for decades.

Here, an attempt is made to study the fracture behavior of cracks within the bond lines in hardwood. To characterize the fracture behavior, the fracture toughness was measured under five different load angles [mode 1 (M1) to mode 2 (M2)]. The relative humidity (RH) was varied and two different adhesives types were observed.

Materials and methods

European beech wood (*Fagus sylvatica* L.) was in focus. The samples were prepared from the same trunk lumbered in the region of Zurich, Switzerland. Prior to sample preparation, the raw beech wood boards were conditioned at standard atmospheric conditions (20°C, 65% RH) until the equilibrium moisture content (EMC) was reached. At that point, the wood had a density of approximately 706 kg m^{-3} and a MC of 14.5%. After conditioning, the slats of approximately 50×65×400 mm³ were cut out and glued according to the manufacturers' guidelines. The investigated adhesives are as follows: (1) phenol resorcinol formaldehyde (PRF) resin Aerodux (glue 185 RL with hardener HP 155) provided by Bolleter Composites AG (Arbon, Switzerland) and (2) one-component polyurethane (PUR) HB S 709 provided by Purbond AG (Sempach-Station, Switzerland).

Before joining the slats, thin silicon-coated films with a thickness of 35 µm were inserted into the bond line for the initiation of the crack (Figure 1). This procedure was adapted from ISO 25217 (2009). After bonding, the slats were first stored again at standard atmospheric conditions for 7 days and then conditioned to the desired environmental humidity. Three sample series were conditioned. (1) Dry climate: 20°C, 50% RH, MC 11.0%. (2) Standard climate: 20°C, 65% RH, MC 14.6% and (3) wet climate: 20°C, 95% RH, MC 21.1%. Finally, samples of 6×50×130 mm³ were cut out, so that they contained an initial crack (i.e., silicon film) of approximately 25 mm length (see Figure 1 for the illustration). The orientation of the anatomic
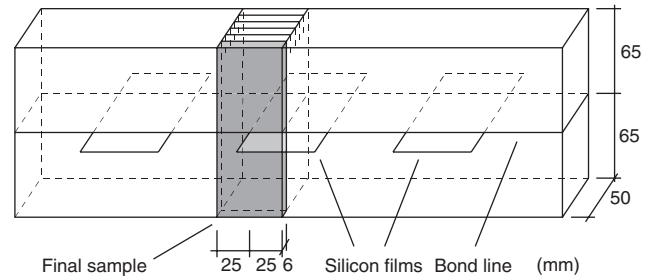

Figure 1 Sample preparation and geometry (units in mm).

directions was chosen so that the crack should propagate in the longitudinal (L) direction and both latewood (LW) and earlywood (EW) are present on the crack plane, that is, with an annual ring angle of 60°–90°, respective to the glue joint.

The orientation of the annual rings in the bonded samples was chosen so that they coincide with industrial bonded elements, and the crack tip was placed directly in the adhesive to assure the investigation on the adhesive, not the solid wood. Therefore, the role of the solid wood was of minor importance, and the focus clearly was on the adhesive systems. The fracture behavior of solid wood is well investigated in different anatomical directions and with different MCs. Ozyhar et al. (2012a) completed an extensive research, which included fracture mechanics, on the same source of beech wood as observed here.

The experiments were conducted on a Zwick/Roell Z010 universal testing machine with a 10 kN load cell and an Arcan test mount (Figure 2). The Arcan test mount can be modified to realize load angles on the glue joint from 0° to 90° in 22.5° steps. A load angle $\alpha=0°$ therefore corresponds to the fracture mode 2 (M2), and $\alpha=90°$ corresponds to the fracture mode 1 (M1). Mixed modes (MM) are denoted with the respective load angle as index (e.g., MM2$_{2.5}$ for $\alpha=22.5°$). In Figure 2, an exemplary test setup for M1 (90°) is shown. For greater clarity, the other load angles are also denoted in this figure.

The tests were performed with a constant displacement rate of 0.5 mm min^{-1} resulting in an experimental duration between 30 and 90 s. The displacement and corresponding load were recorded during the experiments. The displacement, however, is not relevant for the
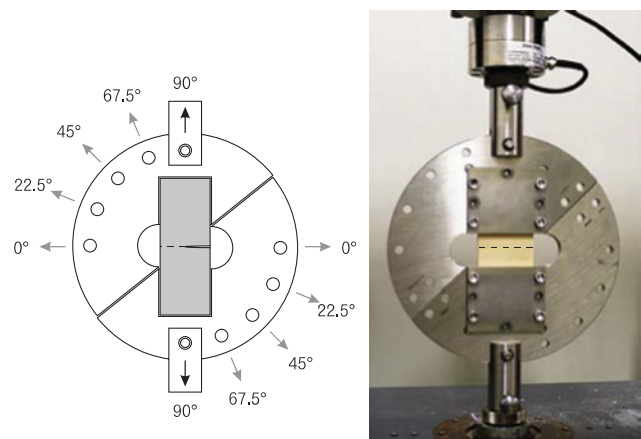

Figure 2 Arcan test with mounted sample.
The illustration on the left indicates the different load angles. The photo shows an experimental setup for M1. The dashed line indicates the glue joint.

data evaluation [see Eqs. (1)–(3)]; therefore, the crosshead movement was taken as a measure. The exact initial crack length and sample geometry were measured straight after testing the samples.

The fracture toughness K_c was calculated using the following formulas:

$$K_{I,c} = \zeta_I(\alpha) \cdot \frac{F_{max}}{w \cdot t} \cdot \sqrt{\pi \cdot \alpha} \tag{1}$$

$$K_{II,c} = \zeta_{II}(\alpha) \cdot \frac{F_{max}}{w \cdot t} \cdot \sqrt{\pi \cdot \alpha} \tag{2}$$

$$K_{\alpha,c} = \sqrt{K_{I,c}^2 + K_{II,c}^2} \tag{3}$$

where F_{max} is the load, at which crack propagation is initiated; w and t are the width and thickness of the sample, respectively; a is the initial crack length; and ζ_I and ζ_{II} are the respective correction factors, depending on the load angle α (Table 1). These form factors were adapted from Richard and Benitz (1983).

After testing the samples, the wood failure percentage (WFP) was analyzed according to EN 302-1 (2013). The statistical significance was evaluated according to DIN 53804-1 (2002) with a 5% level of significance. The standard deviations (SDs) are given for the measured values and the confidence intervals (CIs) are given for the derived results (i.e., K_c).

Results and discussion

The above-mentioned calculation of K_c with the corresponding correction factors is based on isotropic materials and might therefore not be valid for wood or wood-based products. However, this evaluation is still a passable approximation due to the lack of a numerical analysis (FEM) and allows the comparison of the results. For a more precise analysis (i.e., based on the FEM), all experimental results are presented in Table 2 and the actual sample geometries are listed in Table 3. The material properties for such an analysis can be found in the work of Kläusler et al. (2013) for the adhesives and Ozyhar et al. (2012a, b) for beech wood, as both materials are exactly from the same source as that studied here.

For a better overview, the K_c values with the corresponding CIs are illustrated in Figure 3 for PUR and Figure 4 for PRF. In these figures, the monotone piecewise cubic interpolation (according to Fritsch and Carlson

1980) was applied to consider any local extrema, most evident at $K_{I,c}$ and $K_{II,c}$, respectively.

The bulk density of the wood lies within the common range found in the literature, being at 706 kg m^{-3} at standard climatic conditions. The MCs (Table 2) are slightly higher compared with literature values (Scheffler et al. 2004; Sonderegger et al. 2011; Ozyhar et al. 2012b) but are within the normal range.

No significant differences of the K_c of the PUR glue joints can be found between load angles from M1 to MM$_{22.5}$, but a significant increase is visible from MM$_{22.5}$ to M2. This observation applies to all three climate steps. It can be assumed that PUR glue joints perform equally well independent of the load angle until a certain percentage of shear force is reached. In the regular to dry climates, wood failure starts to occur occasionally when decreasing the load angle to 22.5°. As for K_c, a distinct increase in WFP is notable between MM$_{22.5}$ and M2. It thus seems plausible that, at that angle, toward pure shear load, bonding mechanisms come into account that significantly increase the glue joint's performance. The possible mechanisms can be differences in the stress field, surface roughness, and mechanical interlocking.

Similar observations can be made for the PRF in the dry and standard climates as for the PUR, except that $K_{I,c}$ is lower than those at the MM. However, at the wet climate, an unexpected behavior was observed. The measured $K_{I,c}$ of the PRF is not only higher than at standard atmosphere, but they are even at the same level as $K_{II,c}$ in the wet climate. The minimum K_c can be found at a load angle of 22.5° respective to the bond line, being significantly lower than $K_{I,c}$ and $K_{II,c}$. An explanation for this behavior was not found, neither in the evaluation nor in the literature.

Regarding the WFP of both adhesives (Table 2), it becomes apparent that shear stresses at glue joints promote wood failure in the system. In general, PRF predominantly produces wood failure, whereas PUR mostly fails in adhesion [Ammann and Niemz (2014) made similar observations]. However, both adhesives show increasing WFP with decreasing load angle. As already stated, the adhesive systems seem to benefit from decreasing load angles because of the better distribution of stresses at the glue joint and better physical bonding (roughness, mechanical interlocking) at the interface parallel to the joint, which is also notable in the WFP.

Despite the generally low WFP of PUR, these glue joints do not perform worse than the solid wood (Scheffler et al. 2004; Ozyhar et al. 2012b). This behavior might seem surprising at first sight, but it can be explained with the material characteristics of the adhesive. PUR, in contrast to PRF or solid wood, shows a ductile behavior

Table 1 Correction factors for M1 (ζ_I) and M2 (ζ_{II}) fracture toughness for different load angles α, adapted from Richard and Benitz (1983).

	$\alpha=90°$	$\alpha=67.5°$	$\alpha=45°$	$\alpha=22.5°$	$\alpha=0°$
ζ_I	2.810	2.539	1.918	1.004	0
ζ_{II}	0	0.571	0.990	1.295	1.448

Table 2 Material properties and results for different load angles.

Material properties	Adhesive	α (°)	$K_c \pm$CI (MPa√m)	$F_{max} \pm$SD (N)	n	WFP (%)
50% RH, 20°C	PRF	90	1.34±0.25	563±125	8	85
ρ_b: 708±14 kg m^{-3}		67.5	1.90±0.27	868±130	8	90
MC: 11.0%		45	1.65±0.06	924±45	8	95
		22.5	1.66±0.14	1225±131	8	95
		0	2.43±0.15	2026±185	8	90
	PUR	90	1.07±0.11	462±62	8	0
		67.5	1.01±0.23	461±118	7	0
		45	1.08±0.10	613±71	8	0
		22.5	1.00±0.18	736±153	7	10
		0	2.21±0.37	1866±257	5	80
65% RH, 20°C	PRF	90	1.11±0.10	453±53	8	80
ρ_b: 706±13 kg m^{-3}		67.5	1.62±0.27	730±150	8	90
MC: 14.6%		45	1.42±0.15	775±111	8	95
		22.5	1.51±0.10	1093±85	8	100
		0	2.30±0.15	1883±159	8	100
	PUR	90	0.71±0.07	295±45	10	0
		67.5	0.77±0.11	355±73	11	0
		45	0.82±0.08	452±65	11	0
		22.5	0.79±0.11	571±106	10	5
		0	1.85±0.18	1524±184	10	25
95% RH, 20°C	PRF	90	1.70±0.13	740±80	8	90
ρ_b: 720±27 kg m^{-3}		67.5	1.51±0.26	719±159	8	80
MC: 21.1%		45	1.40±0.13	800±92	8	80
		22.5	1.28±0.11	950±100	8	95
		0	1.73±0.10	1463±124	7	100
	PUR	90	0.74±0.13	317±67	7	0
		67.5	0.80±0.08	367±35	6	0
		45	0.71±0.09	407±63	6	0
		22.5	0.69±0.08	514±95	10	0
		0	1.12±0.13	942±126	7	0

ρ_b, bulk density; K_c, fracture toughness; F_{max}, maximum load initiating crack propagation; n, number of specimens.

under tensile load with a distinctive plastic deformability (Kläusler et al. 2013). This characteristic allows for a better stress distribution along the glue joint and hence leads to a lower stress concentration at the crack tip (River 2003; Smith et al. 2003; Müller et al. 2005). The reduced stress concentration in the bond line also diminishes the

Table 3 Mean sample geometry and SD of the different sample groups.

Specimen	n	Width (mm)	Length (mm)	a_0 (mm)
PRF				
50% RH	40	6.78±0.17	49.17±0.07	24.61±0.59
65% RH	40	6.55±0.10	49.56±0.19	24.37±0.97
95% RH	39	6.76±0.13	49.30±0.21	23.49±0.72
PUR				
50% RH	35	6.82±0.12	49.09±0.22	24.28±1.06
65% RH	52	6.59±0.08	49.49±0.26	23.99±0.77
95% RH	36	6.81±0.24	49.40±0.10	24.62±1.06

n, number of specimens; a_0, initial crack length.

likelihood of wood failure. The fact that PUR has a higher tensile strength than beech wood perpendicular to the grain confirms that the weakest zone in PUR glue joints in beech wood is the adhesion (Ozyhar et al. 2012a; Kläusler et al. 2013; Ammann et al. 2014). However, having the superior $K_{I,c}$, a failure in the adhesion is preferred over wood failure, at least for M1 loads.

Both adhesive systems show generally decreasing K_c with increasing RH. The effect is more pronounced on M2 than on the other modes. For European beech wood, Ozyhar et al. (2012a) found that, in general, all strength parameters (including $K_{I,c}$) decease with increasing RH. Kläusler et al. (2013) found that the strength of PUR also decreases with increasing RH, but PRF is not influenced. Thereupon, it can be concluded that the decrease of K_c of PRF glue joints is mainly attributed to the wood properties and not to the adhesive itself. With regard to PUR, a greater decrease in K_c has to be expected when the humidity is increased. On assessing the mean values of the $K_{II,c}$ of PUR (Table 2), when switching from an RH of 50% to
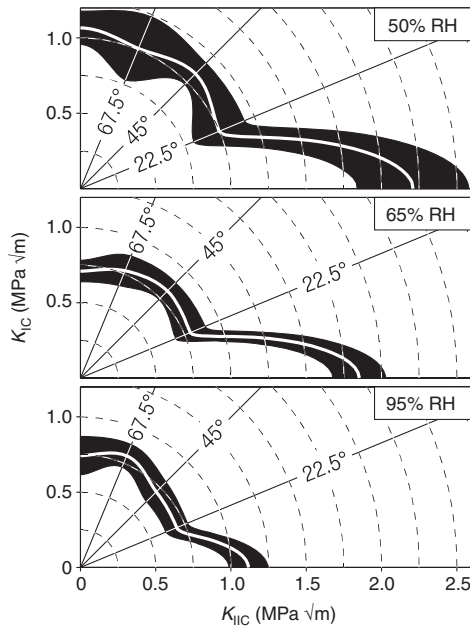
Figure 3 K_c of PUR: mean with CIs (black area).

95%, one can clearly see that a factor of 2 exists, but PRF only changes by a factor of 1.5 between these values. Thus, PUR glue joints are indeed more influenced by RH than the PRF ones. However, as already stated, this effect is most notable for M2. The K_c of PUR at M1 and the MM are also lower at the standard climate than at the dry climate. However, no further changes are present between the standard and wet climates and all K_c for the mentioned load angles stay at approximately 0.75 MPa√m. The K_c of PRF still decreases at the MM, when switching to wetter climates, but only at a statistically insignificant rate.

The PRF glue joints mostly perform better than those of PUR. The differences are most distinct at the MM, where PRF already has increased K_c, but PUR remains at the same level of $K_{I,c}$. Because PUR is influenced by a different RH and PRF is not (Kläusler et al. 2013), the divergence between the performances of these adhesives increases together with the surrounding RH. Furthermore, at 50% RH, there are no significant differences between $K_{I,c}$ and $K_{II,c}$ of the adhesives. Therefore, if the RH is further decreased, one can assume that the PUR might even outperform the PRF.

Conclusions

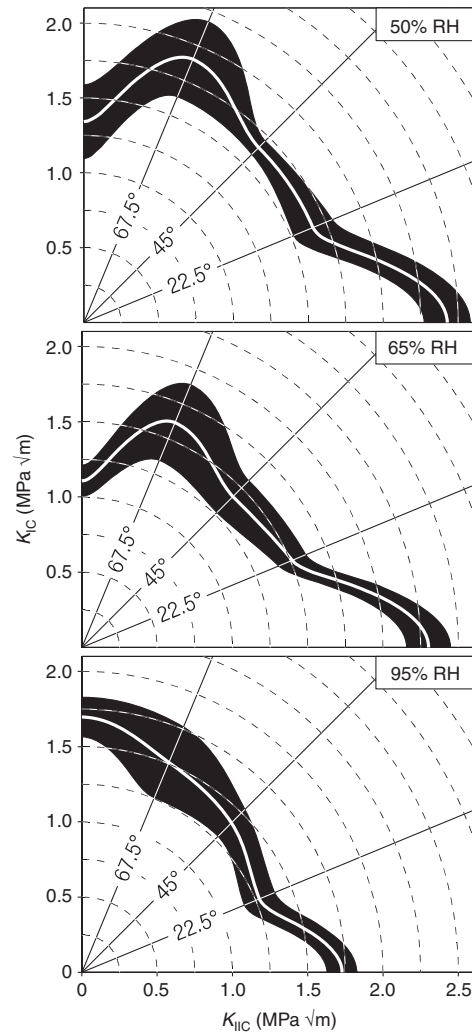PUR glue joints perform equally well when the applied load angle on the bond line is >22.5°. At load angles

Figure 4 K_c of PRF: mean with CIs (black area).

below 22.5°, the K_c of PUR increases rapidly. The K_c of PRF increases with decreasing load angles. The shear stresses at glue joints promote wood failure. PRF joints mainly fail in the wood, but PUR fail in the joint itself. The weakest link of PUR glue joints in beech wood is the adhesion. Nevertheless, such glue joints do not perform worse than the solid wood. The moisture dependency is more pronounced in PUR than in PRF glue joints, which can be attributed to the adhesives' properties. At wet climates, PRF glue joints achieve higher K_c than those of PUR. However, at a moderate dry climate (50% RH), there are no significant differences between PUR and PRF for $K_{I,c}$ and $K_{II,c}$.

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