Jiang Y., Schaffrath J., Knorz M., Winter S., Technische Universität München

# Bonding of various wood species – studies about their applicability in glued laminated timber

## Abstract

In this research project, the gluability of five soft- and hardwoods (ash, beech, Douglas fir, larch and spruce) in combination with four adhesives (EPI, MUF, PRF and PUR) are investigated. Factors such as extractive content, pH value, wettability, etc., that may have significant influence on the bonding strength and durability of adhesive joints, are analyzed. The results of the research project achieved so far will be reported in this article.

## Keywords

Gluability of softwood and hardwood, glued laminated timber, wood extractive, wettability, tensile shear strength

## 1. Introduction

Ongoing climate change has a severe impact on our forests. Hence, the forest conversion in Germany is going to reduce the population of weak monocultures of pine and spruce (Bayerische Staatsregierung 2007). They are replaced by adaptable, climate tolerant and species-rich mixed forests, so that the percentage of tree species, e.g. Douglas fir and beech, is distinctly growing (Bundeswaldinventur 2009). Therefore, the supply of raw material from the forest may change in the near future. To adapt to these changes, the woodworking industry needs to adjust the production processes. Alternatively, advantages can be taken by developing new and improved wooden products. In this context, the bonding technology plays a key role. Bonding of various wood species with higher strength and/or durability, will lead to benefits for wood engineered products. However, experience shows that gluing of timber without sufficient technical knowledge and elaborateness can lead to serious damages. Investigations after the collapse of the Bad Reichenhall ice rink in 2006 showed that damages in structural elements made of glued laminated timber (GLT) are often caused by delaminated glue lines (Blaß and Frese 2007, Dietsch and Winter 2009, Wolfrum and Winter 2007).

Today, in Central Europe GLT is almost exclusively made of spruce or pine (Mack 2006, Ohnesorge et al. 2009). However, in the recent past, research and industry have shown increasing interest in using hardwoods such as beech. At present, for the first time a building with GLT made of beech as structural elements is being erected in Germany. In Switzerland, where building regulations are less restrictive, some experience with GLT made of hardwoods such as ash and beech has been gained in the last decades.

The research project “Bonding of various wood species and studies about their applicability in glued laminated timber” focuses on the gluability of five soft- and hardwoods (ash, beech, Douglas fir, larch and spruce as reference). It is assumed that the selected wood species show increasing importance due to their growth conditions, resistance or strength. The main objective of the ongoing research work is to examine the performance and characteristics of the used wood species in combination with the adhesive systems EPI, MUF, PRF and PUR. These investigations shall give information about the applicability of these wood species in glued laminated timber.

Chemical properties of wood, such as the extractive content and pH-value, play an important role in adhesive bonding. Bikerman (1961) reports that the water-soluble and volatile substances in the wood, such as the hydrophobic resin and fatty acids, move towards the wood surfaces during drying and storage and form a so-called “chemical weak boundary layer”. Highly acidic extractives can accelerate or decelerate the pH-dependent curing reaction of some adhesive systems (Scheikl and Dunky 1996). Zeppenfeld and Grunwald (2005) point out that bonding failure caused by wood extractives are always to be expected when their content (locally) rises above 5%. Therefore, the chemical properties of the wood species used in this work were determined to provide a basis to increase the understanding of the chemical interaction between wood and adhesives.

The attractive forces between adhesive and wood, such as Van der Waal´s forces, dipole-dipole forces and hydrogen bonding, which are necessary for the integrity of a glue line, act in the boundary layers with a thickness that varies from nanometers to micrometers (between 0.1 and 1 nm). In order to provide the optimal intermolecular attraction, the molecules of adhesive and wood should be brought close enough to each other (Habenicht 2009). This requires a sufficient wetting of the wood surfaces. The wettability of the selected wood species has been quantified by means of contact angle measurements.

In addition to the determination of the chemical properties and wettability, strength tests have been carried out to show the performance of different wood-adhesive-combinations. Processing conditions are carefully chosen after evaluating the results of the foregoing tests. Not only the breaking load and the percentage of wood failure, but also the quality of the glue line, which was examined under the microscope, are indicators for the reliability of each bonding. As the research work is in progress, further results, regarding long-time tests for example, are expected.

## 2. Materials and Methods

### 2.1 Materials

Five soft- and hardwoods were selected for this study: ash, beech, Douglas fir, larch and spruce as reference. Sawn timber of these wood species was chosen with densities that more or less match the average value of the respective wood species as given by DIN 68364: spruce (460 ± 50) kg/m$^3$, larch (600 ± 50) kg/m$^3$, Douglas fir (580 ± 50) kg/m$^3$, beech (700 ± 50) kg/m$^3$ and ash (700 ± 50) kg/m$^3$ at (12 ± 1) % moisture content. All specimens were conditioned at 20 °C and 65 % RH (standard climate), so that the equilibrium moisture content of (12 ± 1) % was reached.

Four commercial adhesive systems for load-bearing timber structures were selected:

- Aerodux 185 with Hardener HRP 155 (phenol-resorcinol-formaldehyde adhesive (PRF))
- Prefere 6151 with hardener Prefere 6651 (emulsion-polymer-isocyanate adhesive (EPI))
- Kauramin Leim 690 with hardener 1690 (melamine-urea-formaldehyde adhesive (MUF))
- Jowapur 686.60 (polyurethane adhesive (PUR))

The development of modern adhesives is currently based on MUF-, EPI- and PUR-systems. They are regularly optimized regarding curing speed and flexibility in processing. The PRF adhesives have nowadays only a small market share, especially due to their dark color. With regard to the existing long-time experience, the combination of PRF adhesive with spruce is useful as a reference system.

## 2.2 Chemical analysis

### 2.2.1 Sample preparation

The chemical properties of the wood can highly vary within the same species. Therefore, instead of using reference values, the chemical properties of the timbers, which are used and evaluated in this study, were precisely measured. To obtain reliable results, samples were taken from five different boards of each wood species and cut into small wood cubes (≤ 1.5 $cm^3$).

### 2.2.2 Wood extractives analysis

The wood extractive content was determined by means of cold water and organic solvent extraction. For the extraction process wood flour was prepared from small wood cubes. The cold water extraction was performed at room temperature. Air-dried wood flour was mixed with deionized water. The extraction time period was 48 hours. Afterwards, the pecentages of wood extractives that were soluble in cold water were determined. The organic solvent extraction was carried out at approximately the boiling temperature of the solvents using a Soxhlet extraction apparatus. Three different organic solvents were used successively as extractants: petroleum ether, acetone and methanol. The percentage of wood extractives that were soluble in organic solvents was determined.

The mixtures of wood extractives, which have been obtained though the extraction process, were analyzed. The components of the extractive solutions were separated and characterized using chromatographic separation techniques such as thin layer chromatography (TLC), gas chromatography/mass spectrometry (GC/MS) and ion exchange chromatography (IC).

### 2.2.3 Chemical characterization of wood surfaces by means of pH-measurements

The influence of the surface aging on the chemical properties of wood surface was quantified by means of surface pH measurement. Therefore, from each wood species ten wood cubes were taken and subsequently three veneers with a thickness of 1 mm were prepared using a microtome (see Figure 1, left). The surface pH was measured directly after the veneers were cut off, after 24 hours and after 7days storage at standard climate, respectively. The measurements were taken at room temperature. The thin veneer was placed in the center of a specifically manufactured Teflon container and fixed with the lid of the container (see Figure 1, right). The Teflon lid has a round opening in the center, through which the thin veneer is visible from above. Pure water with a pH of

7 was applied on the wood veneer. A pH flat membrane electrode (InLab® surface electrode, Mettler Toledo GmbH) was used to measure the surface pH.

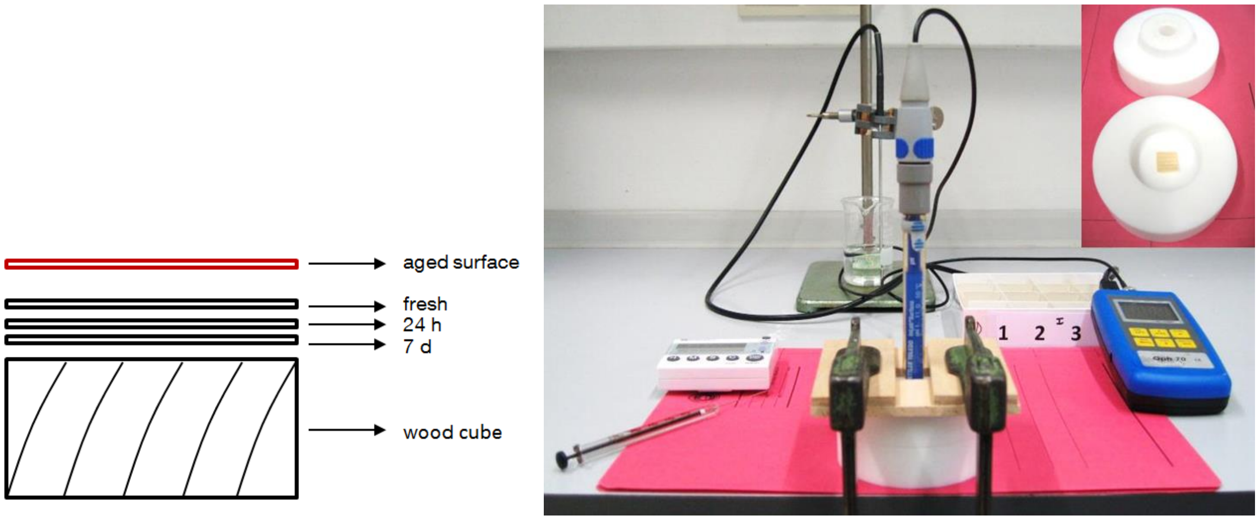


**Figure 1: Preparation of the veneers (left) and measuring device with pH flat membrane electrode (right)**

### 2.3 Contact angle measurement

Five specimens of each wood species with a thickness of 5 mm and with a radial surface area of 40 mm × 100 mm were prepared. Contact angles were measured directly after planing as well as after 24 hours and 7 days conditioning at standard climate, respectively. In addition, some specimens were slightly sanded prior to measurement using sandpaper with grit size 100. During the entire process the surfaces of the specimens were carefully kept free from contamination. The contact angles were determined with the measuring system EasyDrop (KRÜSS GmbH, Germany) with the so-called dynamic method.

### 2.4 Longitudinal tensile shear tests

The performance of each adhesive system in combination with the selected wood species was evaluated by means of the tensile shear tests according to DIN EN 302-1. Specimens were prepared for each combination of wood species and adhesive system from two bonded boards with a thickness of 2 × 5 mm. Processing parameters were carefully chosen after consulting the adhesive manufacturers. After curing, ten specimens with a length of 150 mm, a width of 20 mm and a defined shear plane of 200 mm$^2$ were cut from the bonded boards. The specimens were conditioned at 20°C/65% RH for 7 days. Prior to the tensile shear tests, the specimens were subjected to one of the following treatments according to EN 302-1: A1 (without further conditioning), A2 (4 days in cold water), and A4 (6 hours in boiling and 2 hours in cold water). After the shear test the glue line quality of different wood-adhesive-combinations was evaluated by means of microscopy. Sections of 20 µm thickness were prepared from the end face of the tested shear specimens.

# 3. Results and Discussion

## 3.1 Wood extractives analysis

The extractive contents of the five different wood species are summarized in Figure 2.

A comparatively high extractive content of approximately 10% was found for larch when using cold water as extractant. That is because larch contains a lot of sugars, which are highly soluble in water. The extractive content of the ash is slightly above 5%. Spruce and beech, on the other hand, contain only small amounts of wood extractives (< 2%).

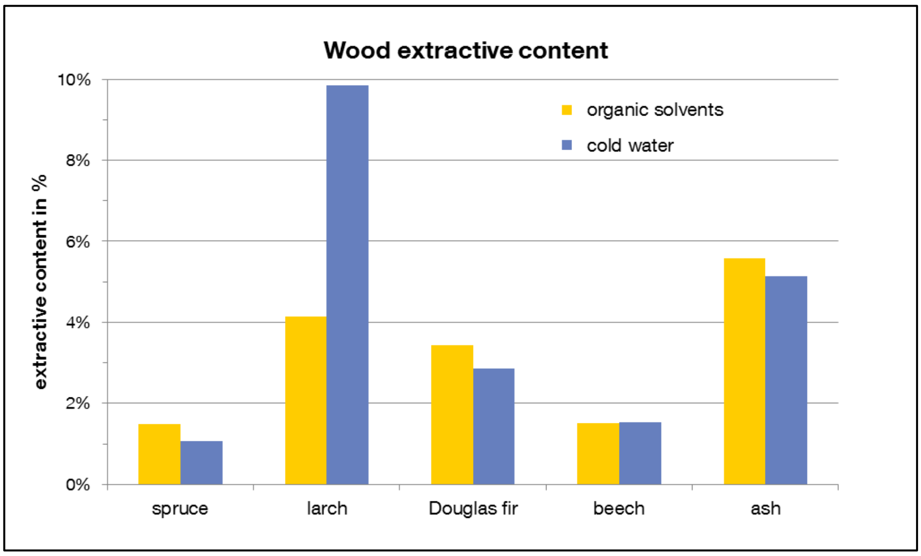


**Figure 2: Extractive contents of spruce, larch, Douglas fir, beech and ash, determined with organic solvents and cold water as solvent**

An overview of the main extractives of the five analyzed wood species can be found in Table 1. The extractives are subdivided into the following substance classes: fatty acids, terpenes/steroids, sugar/sugar derivatives and phenols/tannins. Extractives such as linoleic acid, sitosterol, arabinose and taxifolin are found in most of the examined wood species.

**Table 1: Overview of the extractives of the analyzed wood species**

| wood species | fatty acids | terpenes/ steroids | sugar/ sugar derivatives | phenols/ tannins |
|---|---|---|---|---|
| spruce | oleic acid | dehydroabietic acid<br>sitosterol | arabinose | hydroxymatairesinol |
| larch | linoleic acid<br>oleic acid | isopimaric acid<br>dehydroabietic acid | arabinose<br>galactose | dihydrokaempferol<br>taxifolin |
| douglas fir | linoleic acid | isopimaric acid<br>dehydroabietic acid<br>sitosterol | arabinose | dihydrokaempferol<br>taxifolin |
| beech | - | Sitosterol | fructose<br>glucose | - |
| ash | linoleic acid<br>palmitic acid | stigmasterol<br>sitosterol | glucose<br>D-mannitol<br>saccharose | - |

## 3.2 Wood surface pH

The lowest pH value is shown by Douglas fir, which varies between 3.25 and 4.25 (Figure 3). The surface pH of the two hardwood species are the highest and vary between 4.75 and 5.50. The difference between the highest and lowest pH value is assumed to affect the pH-dependent curing reaction of adhesive systems (e.g. MUF, PRF).

An influence of surface aging on the pH value can be stated for all five wood species. As shown in Figure 3, the surface pH of the majority of the wood species increase slightly after 24 hours storage at 20°C/65% RH. After 7 days, the surface pH of spruce, larch, Douglas fir and ash rise again. Only the pH value of beech shows a slight reduction.

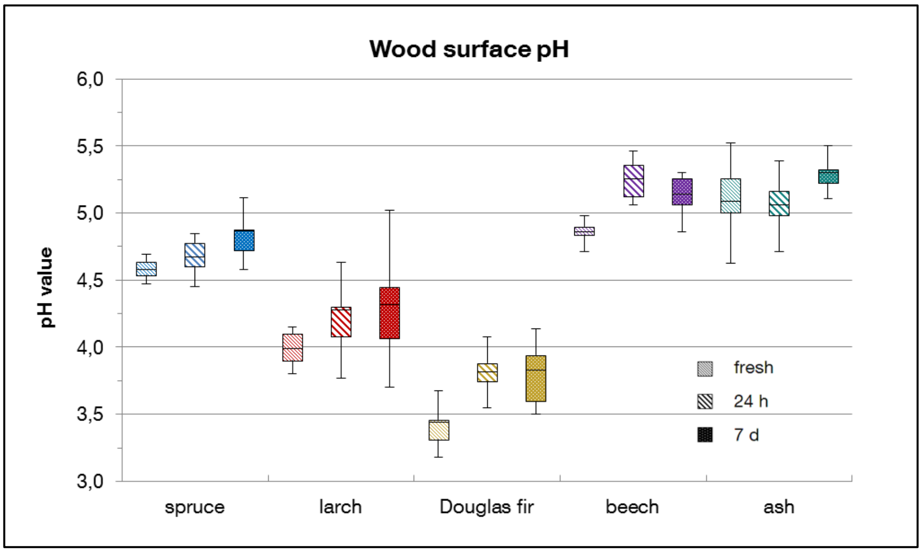


**Figure 3: Surface pH of the fresh and aged wood surfaces**

### 3.3 Contact angle

In general, the low contact angles (< 90°) indicate a good wettability (Dunky and Niemz 2002) of all five wood species (see Figure 4). Compared to the planed wood surfaces, the surfaces, which are slightly sanded, can be more easily wetted by water (Figure 4, left). Regardless of which surface preparation is used the lowest wettability is shown by Douglas fir.

It is clear from the data presented in Figure 4**Fehler! Verweisquelle konnte nicht gefunden werden.** (right) that the surface aging affects the wettability of the wood surface. Only after 24 hours an increase of the contact angle can be seen for almost all wood species, except spruce. After 7 days the wettability of spruce has been reduced most though.

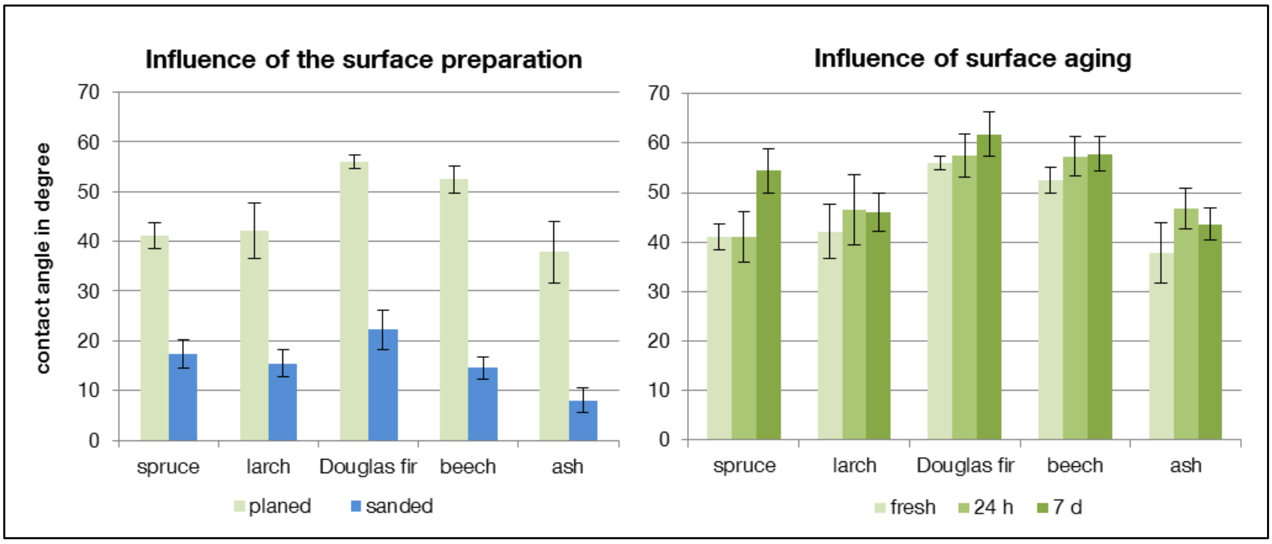


**Figure 4: Influences of surface preparation (left) and aging (right) on the wettability of wood surfaces**

### 3.4 Tensile shear strength

The bond strengths of different wood-adhesive-combinations, which were determined by longitudinal tensile shear tests, are displayed in Figure 5. The minimum requirements on the average longitudinal tensile shear strength of adhesive type I and glue line thickness of 0.1 mm are given in DIN EN 301 with 10 N/mm² after treatment A1 and 6.0 N/mm² after treatments A2 and A4, respectively. However, these requirements can only be applied to specimens made of beech. As a further reference value, the tensile shear strength of solid wood samples with dimensions as specified in DIN EN 302-1 was determined.

Low bond strength was found for the specimens glued with PUR adhesive. Nevertheless, the tensile shear strength, which is achieved by the combination of PUR adhesive and larch after the treatments A2 with cold water and A4 with boiling and cold water, exceed the expectation of the adhesive manufacturer.

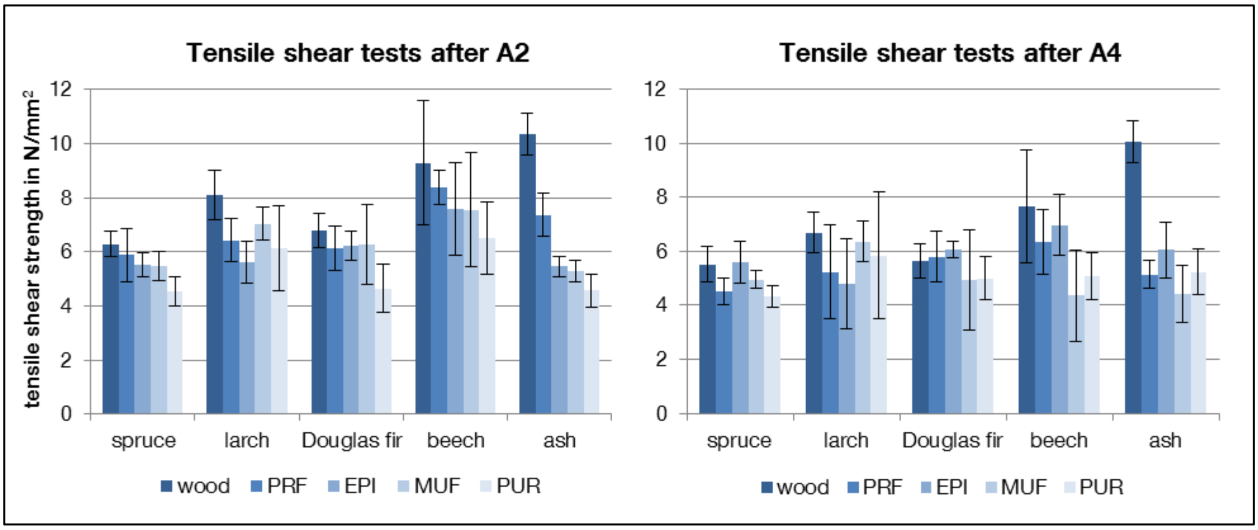


**Figure 5: Longitudinal tensile shear strength of different wood-adhesive-combinations**

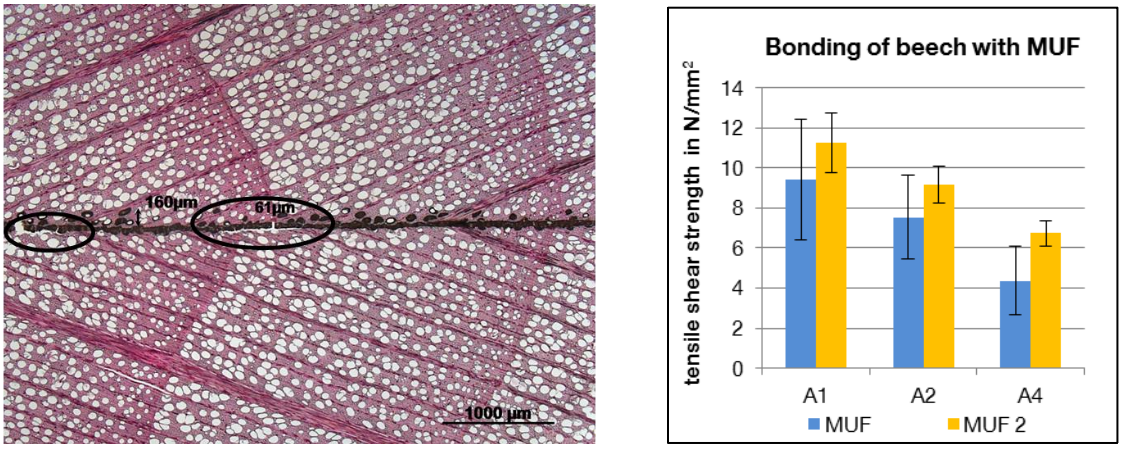


**Figure 6: Micrograph of beech specimens produced using MUF adhesive (left) and comparison between the shear strengths achieved by the specimens of first and repeated test (right)**

Figure 6 (left) shows the electron micrograph of beech specimen glued with MUF adhesive. It is visible that the adhesive has penetrated only into the upper wood surface. The lower surface is only partially wetted. Besides that, considering the amount of adhesive applied on the wood surface, the glue line seems to be very thin. After consultation with the adhesive manufacturer, the test series "beech specimen glued with MUF adhesive" was repeated with changed process parameters (increased application amount and shortened open assembly time). Doing so, a significant increase of the tensile shear strengths was achieved (see Figure 6, right).

## Conclusions and outlook

In this research project, differences regarding the gluability of various wood species are studied. Detailed information about the chemical extractives and surface pH is presented. The results of contact angle measurements show the wettability of surfaces with regard to age, preparation and wood species. The bond strengths of different wood-adhesive-combinations are evaluated by means of the tensile shear tests.

The influence of the above mentioned characteristics on the curing behavior and bonding strength will be further investigated. In addition, various processing conditions on selected combinations of wood species and adhesive system will be analyzed. Hereby, delamination tests, block shear tests and microscopic analysis are carried out on small test beams. With the knowledge of the best performing processing conditions larger test beams will be subjected to long-term testing under load and various climate conditions. The test setup will lead to information about durability and creep behavior. Finally, the residual strength will be compared through a four-point bending test.

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