

PENETRATION DEPTH OF ADHESIVE DEPENDING ON APPLIED PRESSURE DURING GLUING PROCESS

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Abstract

Wood is one of the key raw materials that are used in constructions. The research of the adhesive joint properties is important to characterize the overall design of the wood construction. To achieve good joint performance and durability it is important to adhesive penetrate into wood substrate deep enough. Adhesive penetration is influenced by many factors such as wood, adhesive and process factors. The objective of the present paper is comparison of the polyurethane (PU) glues penetration depth into Scots pine (*Pinus sylvestris* L.) and Norway spruce (*Picea abies*) wood depending on applied pressure. Penetration depth of cured adhesive was determined using fluorescence microscopy. Obtained results of the bond-line width had the good correlation with applied pressure value.

Key words: softwood, polyurethane glue, applied pressure, penetration, fluorescence microscopy.

1. INTRODUCTION

Wood is one of the key raw materials that are used in load bearing and non-load bearing constructions. The increasing demand for sustainable construction caused a need to develop more new material bonding technologies. Nowadays modern timber engineering strongly depends on the adhesive joint and bonding process. The research of the adhesive joint properties is important to characterize the overall design of the wood construction.

Creation of wood construction accurately of bond between adhesive and wood substrates requires glue to sufficiently penetrate into wood. Good penetration of adhesive is promoted by excellent wood-to-adhesive-surface interaction and excellent adhesive mobility and flowability which in turn may be affected by factors of bonding process.

Gluings of wood elements is rather simple processing step, but the details of the penetration and hardening of the adhesives into substrate, like the porous wood, is not fully understand. Wood-adhesive bond-line performance or behavior depends on wide range of different factors, such as the bonding process (spreading of adhesive, pressing time, applied pressure and temperature, environmental factors (level and rate of change in both temperature and relative humidity, etc.) used adhesive properties (adhesive type, viscosity, wetting properties, chemical structure, etc.) and wood properties (wood species, surface roughness and morphology, porosity, chemical composition, presence of wood extractives, pH, density, wood grain orientation, moisture content, etc.) (Hass et al. 2012; Custódio, Broughton & Cruz 2009).

The aim of this study was to compare penetration depth of polyurethane (PU) glues into Scots pine (*Pinus sylvestris* L.) and Norway spruce (*Picea abies*) wood at bond-line to evaluate effect of the applied pressure.

2. MATERIALS AND METHODS

Wood samples of Scots pine (*Pinus sylvestris* L.) and Norway spruce (*Picea abies*) with approximate surface area 30×30 mm and thickness of 50 mm were cut from rectangular timber so that the wood tracheids are open and the grain orientation to the surface was 45° to diminish influence of the wood heterogeneity between radial and tangential direction. Afterwards wood samples were conditioned in desiccators above saturated salts solutions (NaCl, $(\text{NH}_4)_2\text{SO}_4$, KNO_3 , K_2SO_4) until constant mass of samples was reached.

In this study were used two kinds of one-component commercial PU glues with different curing time (Glue I and Glue II). Viscosity of glues was determined with rheometer Bohlin CVO-100 (Malvern Instruments Ltd.). In first regime polyurethane glues were applied to one or both surfaces of wood samples ($100 - 400 \text{ g m}^{-2}$) and clamped with 40 - 60 kPa pressure for 6 hours at room temperature (20 °C). The second technological regime – PU glues

were applied to both surfaces of wood samples (200 g m^{-2}) and compressed with 295 kPa pressure for 3 hours at room temperature. The pressure of 295 kPa was chosen to simulate the used glulam manufacturing technology. Then samples were sawn in smaller ones to fit in the microtome HM 315 (Microm International GmbH) and plasticized into ethylene glycol and red beet juice solution at least for two days. Red beet juice were used to stain samples and separate glue from wood in fluorescence microscopy.

From plasticized wood with microtome were cut off thick samples ($10 - 20 \text{ }\mu\text{m}$). Bond-line thickness and penetration depth of glues were examined with microscope Axioskop 40 (Carl Zeiss Jena GmbH) (with 16x objective magnification) using UV blue filter.

The wetting behavior of the wood samples and glues was characterized by the contact angle method (goniometer technique). The contact angle values of the sessile drop were measured with OCA20 device with E-MD unit (Data Physics instruments GmbH)

The surface free energies, their Lifshitz–van der Waals component and an acid-base component of solid (wood or glue) surface were calculated using Acid-Base (AB) approach (Gindl et.al. 2001). Numeral values were obtained from measured values of contact angles of three test liquids: diiodomethane as non-polar one (produced by ACROS organics), and two polar (purified water (reverse osmosis filtered and demineralised with purifying system TKA, conductivity $0.2 \text{ }\mu\text{S}$) and ethylene glycol (produced by ACROS organics) with known surface tension components. Surface tension characteristics of test liquids used in calculation by software of OCA 20 are listed in the Table 1.

1. Table. Surface free energy characteristics of test liquids by H.Y. Erbil used to measure contact angles (Erbil, 1997)

Test liquids	Surface tension, γ_L , mN m^{-1}	Dispersion component, γ_L^D , mN m^{-1}	Polar component, γ_L^P , mN m^{-1}	Acid component, γ_L^A , mN m^{-1}	Base component, γ_L^B , mN m^{-1}
Diiodomethane	50.80	50.80	2.60	0.00	0.00
Ethylene glycol	48.00	29.00	19.00	1.92	47.00
Water	72.80	21.80	52.20	25.50	25.50

Drops of test liquids on the wood surface were recorded with video mode at the maximal speed - 25 frames per second due to rapid test liquids suction into wood. After that the video was analyzed frame by frame with the dynamic tracking mode and the contact angle was determined with droplet shape smoothing optimization and fitting by Laplace-Young method that is included in DataPhysics OCA20 software.

We supposed that the initial value of receding contact angle of the test liquid is at quasi-equilibrium state that corresponds to the equilibrium state. It can be obtained by extrapolating the trend of changing with time receding contact angle values to time moment 0 s, when the drop touches wood surface and drop is formed in the same way as described by Rubina et al. (2009).

3. RESULTS AND DISCUSSION

Adhesive penetration is influenced by wood factors (wood species, structure, location in the trunk, anatomical orientation, grain direction, size of the cellular elements or surface roughness, its moisture content, the degree of degradation, air content in its structure), adhesive factors (type and viscosity, which are dependent on its molecule size, polarity, and wetting ability) and process factors (applied pressure, time, temperature) (Hass et al. 2012; Kučerová 2009). It has concluded by Hass et al. (2012), that the penetration in beech wood is affected by the vessel network characteristics and by the viscosity of the adhesives. Determined viscosity for Glue I was $5.32 \pm 0.01 \text{ Pa s}$ and for Glue II was $12.05 \pm 0.03 \text{ Pa s}$.

The PU glue pre-polymer have a smaller molecule size and is able to penetrate into the structure of wood and only after then cure. Tracheary elements of wood provide channels and the polymer can flow through for hundreds of micrometers. It has concluded by Smith et al. (2002) that the polypropylene (PP) adhesive can flow into wood openings as small as $1 \text{ }\mu\text{m}$ and flow into parenchyma cells that are in distance from $15 \text{ }\mu\text{m}$ up to 180

μm (Smith, Dai & Ramani 2002). The glue's main fraction with high molecular weight did not penetrate into wood on a nanometer scale, but its small fraction of sufficiently low molecular weight can penetrate and after curing of adhesive acts as internal or external plasticizer into the wood (Mader et al. 2011). Adhesive is able to penetrate in and fill cut cell lumens and mostly the interconnected network of vessels, and the open space between the two contacting rough surfaces (Hass et al. 2012).

The actual penetration depth of solution or adhesive depends on the wood permeability to liquids, the technological methods, and physic-chemical properties of the specific adhesive. Liquid flow through porous material under ideal conditions (the porous material is homogeneous, no interactions occur between the liquid and the porous material, permeability is independent of the porous material length in the flow direction) is defined by Darcy's law (Equation 1) (Kučerová 2012):

$$Q = K * \frac{A}{L} * \frac{1}{\eta} * \Delta P \quad (1)$$

where Q is the liquid volume flow [$\text{m}^3 \text{s}^{-1}$], K is the specific permeability of wood [m^2], A is the area perpendicular to the liquid flow [m^2], L is the sample length in the direction of flow [m], η is the dynamic viscosity of the liquid [Pa s], and ΔP is the pressure gradient [Pa]. As described by Darcy's law the pressure gradient ΔP , being the cause for the liquids penetration into wood, is related to the technological procedure (based on the capillary or outer pressure). Due to the fact that in real processes are changing conditions like heterogeneous structure of wood, interactions between wood and liquid, the theoretical penetration depths calculated by mathematical models differ from the real penetration (Kučerová 2012; Fernandes, Kjellow & Henriksen 2012). Thus, it is necessary to search for experimental methods that allow the study of the penetration of adhesive into the wood structure. In our studies we use fluorescence microscopy to evaluate bond-line thickness of cured adhesive depending on applied pressure. In the further studies we will evaluate and measure adhesive flow rate in horizontal capillaries of wood structure depending on applied pressure.

Surface free energy is a fundamental characteristic parameter that influences surface related processes such as surface wetting phenomena, adhesives spreading and penetration into wood and also wood adhesion with glues (Gindl et al. 2001) and can be calculated and quantified from measured contact angle values (Chau et al. 2009). Obtained results of contact angle and surface energy values of glues and Scots pine and Norway spruce wood are summarized in Table 2.

2. Table. Results of contact angle and surface energy values

Surface	Contact angle ($\pm$ SD), $^\circ$			Surface energy, mN m^{-1}	
	Water	Diiodomethane	Ethylene glycol	Lifshitz – van der Waals component	Base component
Glue I (on polyethylene)	80.12 ± 0.64	47.20 ± 1.03	60.63 ± 1.35	36.21 ± 2.60	9.91
Glue II (on polyethylene)	73.04 ± 0.99	42.00 ± 0.78	57.53 ± 0.55	34.03 ± 1.73	14.45
Scots pine (un-coated) (MC = 15.3 %)	63.97 ± 6.12	26.75 ± 2.15	43.60 ± 4.45	44.37 ± 0.11	17.31
Scots pine (un-coated) (MC = 21.2 %)	78.58 ± 2.79	32.70 ± 2.43	55.73 ± 4.14	37.61 ± 2.62	9.95
Norway spruce (un-coated) (MC = 9.9 %)	70.10 ± 2.52	22.22 ± 1.32	58.90 ± 1.92	47.10 ± 0.08	17.86
Norway spruce (un-coated) (MC = 18.5 %)	56.10 ± 2.91	28.88 ± 1.12	38.30 ± 9.13	43.58 ± 0.88	25.35

Determined contact angle values are higher for Glue I comparing with values of Glue II. Also calculated Lifshitz – van der Waals component is higher due to the difference in glue chemical composition. Due to different wood species, chemical composition, wood extractives and artifact of surface roughness there are difference between obtained contact angle and surface energy values. These values also are affected by moisture content into wood.

The greater moisture content increases contact angle values in case of Scots pine, but decreases in case of Norway spruce.

One of the most important parameter influencing wood bonding is moisture content, both in terms of wood species and resin type and therefore bond-line and penetration depth of glue can vary in relatively large range (Broughton, Hutchinson 2001; Tankut 2007). Water may damage the adhesive physically and chemically by hydrolysis or cracking processes, also by plasticization and swelling of the adhesive or wood and as result induces stresses. In general, the amount of moisture in wood greatly affects wetting, flow and penetration of adhesives (Custódio, Broughton & Cruz 2009).

To achieve good images in microscopy the slides from samples should be as thin as possible, but the cutting process is negatively influenced by the fact that the PU is a thermoplastic adhesive and cannot be plasticized with ethylene glycol. If we cut off a thinner sample, we get better quality of sample image in fluorescence microscopy to measure bond-line thickness. In some cases sample thickness was thicker as 20 μm .

Correlation coefficients of Scots pine and Norway spruce bond-line thickness of both glues with various applied pressures are presented in Tables 3-4. In this study bond-line is assumed as the whole region, where the adhesive can be found: bulk adhesive between the wood surfaces and the area, where the adhesive has penetrated into wood structure. From the firsts investigations was concluded that the penetration depth of glue was deeper in wood sample surface which was previously covered with glue. When glue was applied on both sample surfaces, this fact was not observed; therefore in the further investigations both sample surfaces were covered with glue. The same was expressed by Custódio et al. (2009) that it is important that applied glue properly flows and uniformly covers both of wood surfaces and penetrate into wood pores, cracks and other anatomical features (Custódio, Broughton & Cruz 2009).

3. Table. Scots pine and Norway spruce wood (moisture content 14.4 – 14.7 %) correlation coefficient results between applied pressure (40; 60 and 295 kPa) and bond-line thickness

Wood species and glue type	Correlation coefficient, R
Scots pine with Glue I (MC = 14.4 %)	0.760596
Scots pine with Glue II (MC = 14.4 %)	0.999973
Norway spruce with Glue I (MC = 14.7 %)	0.997169
Norway spruce with Glue II (MC = 14.7 %)	0.999663

4. Table. Scots pine and Norway spruce wood (moisture content 18.5 – 18.9 %) correlation coefficient results between applied pressure (40; 60 and 295 kPa) and bond-line thickness

Wood species and glue type	Correlation coefficient, R
Scots pine with Glue I (MC = 18.7 %)	0.670374
Scots pine with Glue II (MC = 18.9 %)	0.995310
Norway spruce with Glue I (MC = 18.5 %)	0.947294
Norway spruce with Glue II (MC = 18.5 %)	0.996757

Obtained results shows close correlation between bond-line thickness and applied pressure. For most cases obtained relevance is if the higher pressure is applied, the wider bond-line obtained and glue penetrates deeper into wood substrate. Correlation is dependent on wood species, glue type and moisture content. The closest correlation is in the Norway spruce with Glue II (MC = 14.7 %) case, the weakest correlation is in the Scots pine with Glue I (MC = 18.7 %) case.

These results are in accordance with the results of other authors (Flores, Pastor, Martinez-Gabarron et al. 2011) that tested particleboard performance dependence on applied process pressure. They concluded that pressure has a significant contribution to particleboard performance, improving the behavior with a higher pressure during manufacturing process and the use of higher pressures does substantially improve particleboards properties such

as bending strength, modulus of elasticity and modulus of rupture (Flores, Pastor, Martinez-Gabarron, Gimeno-Blanes & Frutos 2011).

By other authors (Fernandes, Kjellow & Henriksen 2012) are concluded that the permeability of wood material measured at higher pressure is higher than that measured for the lowest pressure. Data indicates that at higher pressures the CO₂, formed due to PU hardening, is able to flow through paths that are not available at lower pressure, and tends to increase with working pressure. It is hypothesized that the increased permeabilities result from the softening of the tori of aspirated inter-tracheid bordered pits (Fernandes, Kjellow & Henriksen 2012). If the permeability of wood increases with higher process pressure due to new paths, then those paths more or less can be used by adhesive at higher pressure to penetrate deeper into wood.

Since bond-line thickness is strongly influenced by higher applied pressure there was an interest how will moisture content of wood influence bond-line thickness at 295 kPa working pressure. Bond-line thickness depending on wood species, glue type and wood moisture content is represented in Figure 1.

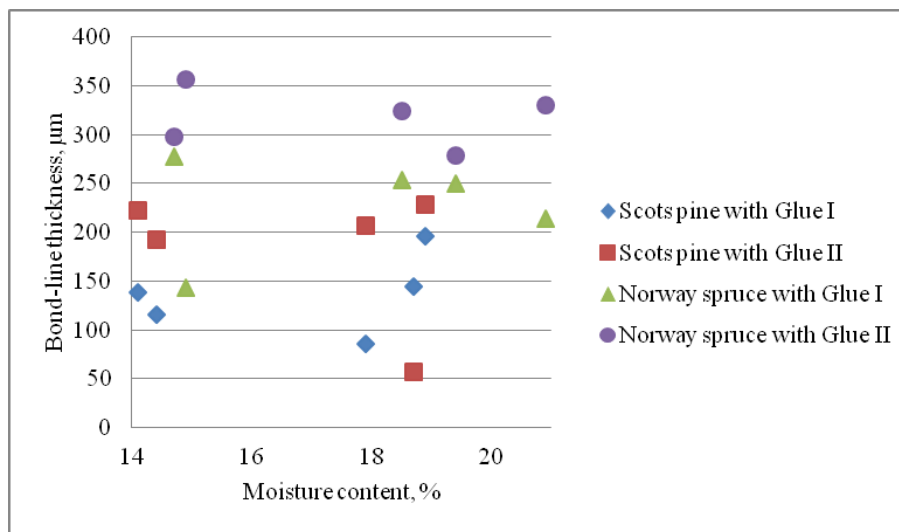


Figure 1. Bond-line thickness with 295 kPa working pressure depending on wood species, glue type and wood moisture content.

Obtained results do not show close correlation between bond-line thickness and moisture content. Nevertheless these results indicate that bond-line thickness is dependent on wood species, glue type and moisture content. In the Norway spruce with Glue II case we obtained the deeper glue penetration into wood. Therefore authors express hypothesis that the penetration depth of adhesive is deeper in Norway spruce wood due to longer average tracheids length than in the case of Scots pine that will be verified in further studies. Bond-line thickness dispersion was large and its values varied from $56.95 \pm 27.60 \mu\text{m}$ (Scots pine with Glue II (MC = 18.7 %)) to $356.82 \pm 47.41 \mu\text{m}$ (Norway spruce with Glue II (MC = 14.9%)).

These results are close with the results of other authors (Ren, Frazier 2012), and the average penetration depth of polyurethane glue was about $55 \mu\text{m}$ (the sum of the both sides from glue-line) and the glue bulk layer had average thickness about $100 \mu\text{m}$, therefore overall bond-line thickness was about $155 \mu\text{m}$. To achieve good mechanical bond stability, it is necessary that the adhesive penetrates deep enough to bond with undamaged cell walls (Ren, Frazier 2012).

4. CONCLUSIONS

The red-beet juice could be used as staining agent for wood and glue separation in microscopic images to determine penetration depth of PU glues and measure bond-line thickness.

Penetration depth of glue into wood is dependent on bonding process, environmental factors, used glue properties and wood properties. Obtained results shows close correlation between bond-line thickness and

applied pressure - the higher is applied pressure, the greater is bond-line thickness and glue deeper penetrates into wood substrate.

Obtained results do not show close correlation between bond-line thickness and moisture content of wood. Bond-line thickness is dependent on different factors, therefore its dispersion is in quite a wide range from $56.95 \pm 27.60 \mu\text{m}$ (Scots pine with Glue II (MC = 18.7 %)) to $356.82 \pm 47.41 \mu\text{m}$ (Norway spruce with Glue II (MC = 14.9%)). The Norway spruce wood has obtained deeper glue penetration with both adhesives at various moisture content. Therefore authors express hypothesis that the penetration depth of adhesive is deeper in Norway spruce wood due to longer average tracheids length than in the case of Scots pine and that will be verified in further studies.

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