PENETRATION DEPTH OF ADHESIVE DEPENDING ON WOOD ANATOMICAL STRUCTURE

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Abstract. Wood is one of the key raw materials that are used in constructions. Thereby it is very important to understand all the processes that are involved in bonding technology. Adhesive penetration into wood at bond-line 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* L.Karst) wood depending on the wood anatomical structure. The bond-line width of cured adhesive and the anatomical structure of wood were determined using fluorescence microscopy. From investigation of wood anatomical structure results were obtained with close correlation between tracheid width and early or late wood width. The calculated surface energy of Norway spruce was higher than of Scots pine wood due to higher measured surface roughness. Good correlation was obtained between Lifshitz–van der Waals component of surface free energy and wood moisture content (R = 0.94153 for Norway spruce, R = 0.77721 for Scots pine). The obtained results shown that the penetration depth of adhesive was deeper in Norway spruce wood (356.82±47.41 μm) due to longer average tracheids width, thickness and length than in the case of Scots pine (56.95±27.60 μm).

Keywords: softwood, polyurethane glue, anatomical structure of wood, penetration, fluorescence microscopy.

Introduction

Wood is one of the key raw materials that are used in constructions. Thereby it is very important to understand all the processes that are involved in bonding technology such as substrate wetting with adhesive, adhesive spreading on the wood surface and adhesive penetration into wood.

On the microstructural or cellular level, wood is an irregular cellular porous solid and microstructural parameters, such as tracheid length and width, cell wall thickness, cell shape, composition of different cell types, and porosity have influence on the wood macroscopic characteristics [1; 2]. The wood tracheid and fibre morphology directly influence fibre flexibility, plasticity and resistance to processing and, therefore, influence the physical properties of the end products [3].

The growth rate of Norway spruce and Scots pine has influence on wood density and tracheids dimensions and cell wall composition. Tracheids length and width increase rapidly during the first years of radial growth. The main factor causing variation in tracheid characteristics is maturation of the cambium. Differences in tracheid length, diameter and cell wall thickness reflect the effects of environmental factors. Growth conditions and geographical location can modify or control early and late wood formation, therefore changing the tracheid properties, wood density and other physical properties [3; 4].

Since Norway spruce consists of 95 % tracheids [1] there is an interest to clarify the relationships between the tracheids dimensions and penetration depth of adhesive into wood and how these characteristics influence each other.

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

Materials and methods

Wood samples of Scots pine (*Pinus sylvestris* L.) and Norway spruce (*Picea abies* L.Karst) with the 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 45º to diminish the influence of wood heterogeneity between the radial and tangential direction. Afterwards the wood samples were conditioned in desiccators above saturated salt solutions (NaCl, $(NH_4)_2SO_4$, KNO_3, K_2SO_4) until constant sample mass was reached.

The wood moisture content was determined using the drying method (105±5) ºC until constant sample mass was reached according to the LVS EN 13183-1:2003 standard [5].

To determine the anatomical structure by microscopy the wood samples were cut from rectangular timber in radial and tangential directions – five pieces for each direction.

Two kinds of one-component commercial PU glues with different curing time (Glue I and Glue II) were used. Viscosity of glues was determined with rheometer Bohlin CVO-100 (Malvern Instruments Ltd.). Polyurethane glues were applied to both surfaces of the wood samples (200 $g \cdot m^{-2}$) and compressed with 295 kPa pressure for 3 hours at room temperature (20 ºC). The pressure was chosen to simulate the industrial glulam manufacturing technology. Then the samples (two from every species with different moisture content each) were sawn in smaller ones (30×30×30 mm) to fit in the microtome HM 315 (Microm International GmbH) and plasticized and stained with red beet juice and ethylene glycol solution at least for two days. Red beet juice was used to separate glue from wood in fluorescence microscopy.

From plasticized wood with microtome thick samples (10-20 μm) were cut off – five from each kind of samples. Bond-line thickness, penetration depth of glues and wood anatomical structure 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). Two samples for each moisture content and species were taken.

Drops of the test liquids on the wood surface were recorded with video mode at the maximal speed – 25 frames per second due to rapid test liquid suction into wood. For each test liquid and sample ten drops were recorded. 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 the receding contact angle of the test liquid is at the 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 the time moment 0 s, when the drop touches the wood surface and the drop is formed in the same way as described by Rubina et al. [6].

The surface free energies, their Lifshitz–van der Waals component and the acid and base components of solid (wood or glue) surface were calculated using Acid-Base (AB) approach [7]. Numeral values were obtained from the measured values of the contact angles of three test liquids: diiodomethane as non-polar one (produced by ACROS organics), and two polar (purified water reverse osmosis filtered and demineralized with purifying system TKA, conductivity 0.2 μS) and ethylene glycol (produced by ACROS organics) with known surface tension components. The surface tension characteristics of the test liquids used in calculation by software of OCA20 are listed in Table 1.

Table 1

Surface free energy characteristics of test liquids by H.Y. Erbil used to measure contact angles [8]

Test liquids	Surface tension, γ_L, $mN \cdot m^{-1}$	Dispersion component, γ_L^D, $mN \cdot m^{-1}$	Acid component, γ_L^A, $mN \cdot m^{-1}$	Base component, γ_L^B, $mN \cdot m^{-1}$
Diiodomethane	50.80	50.80	0.00	0.00
Etyhlene glycol	48.00	29.00	1.92	47.00
Water	72.80	21.80	25.50	25.50

The smoothness behavior of the wood surface (20×20 μm area) was determined with Atomic Force Microscopy (AFM) Solver P47-PRO (NTMDT Co.) in the semi-contact topography mode with the NSG10 platinum coated conducting tip. The surface roughness was determined also with Mahr Perthometer M2 (Mahr GmbH) in 5.6 mm long line.

Results and discussion

Adhesive penetration into wood samples is influenced by wood factors (wood species, structure, 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 (pressure, time, temperature) [9; 10]. It has been indicated by Hass et al. [9], that the adhesives penetration in beech wood is affected by the vessel network characteristics and viscosity. The determined viscosity for Glue I was 5.32±0.01 Pa·s and for Glue II was 12.05±0.03 Pa·s.

Though surface free energy is a principal characteristic parameter that influences the wood surface related processes such as surface wetting, adhesives spreading and penetration into wood, it affects also wood adhesion with adhesives [7] and can be calculated and quantified from the measured contact angle values [11]. The measured and calculated results of the contact angle and surface energy values of adhesives and Scots pine and Norway spruce wood depending on the moisture content are summarized in Table 2.

Table 2

Results of contact angle and surface energy values

Surface	Contact angle (±*SD*), °			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* = 14.1 %)[1]	40.90±7.49	27.37±3.73	33.80±1.68	44.32±0.70	38.74
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* = 17.9 %)[1]	44.50±1.51	23.35±0.07	31.90±1.46	45.34±0.26	36.29
Scots pine (un-coated) (*MC* = 18.9 %)[1]	38.45±5.44	29.00±4.10	32.90±5.02	37.78±0.04	51.55
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* = 14.7 %)[1]	43.35±2.62	26.50±12.16	34.62±7.03	43.80±1.20	42.94
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
Norway spruce (un-coated) (*MC* = 19.4 %)[1]	74.60±9.97	33.95±6.29	58.57±15.04	41.64±2.02	17.92

[1]Note: Samples were stored at desiccators more than month.

The determined contact angle values are higher for Glue I in comparison with the values of Glue II. Also the calculated Lifshitz–van der Waals component is higher due to the difference in the glue chemical composition. Glue I has twice lower viscosity than Glue II. Specific chemical composition is unknown for both adhesives. The difference between the obtained contact angle and surface energy values for Scots pine and Norway spruce is due to different wood species, chemical composition, moisture content, wood extractives and artifact of surface roughness. In several cases the calculated base component is very high (symbol [1] in Table 2) caused by the changes of wood extractives and aged sample surface. This indicates that extractives migrate from the wood to its surface during the aging.

At the surface reorientation of functional groups and oxidation at the wood/extractives-air interface occur [12]. The contact angle and surface energy also are affected by the moisture content into wood, the Lifshitz–van der Waals component decreases with increasing of the wood moisture content. The higher Lifshitz–van der Waals component values were obtained in the Norway spruce case. From the results close correlation between the moisture content and Lifshitz–van der Waals component is obtained. For Norway spruce $R = 0.94153$, but for Scots pine $R = 0.77721$.

The surface roughness of wood depends on the surface processing technique, used tools and speed, grain orientation, and type of wood and may vary over a wide range (40-33000 nm) [13; 14]. Surface roughness (R_a) values were calculated as the difference between the knob height and the mean value of surface roughness relative to the centre plane. Using AFM the obtained average surface roughness of Scots pine samples was 624.03±35.98 nm – in average 426.80±95.51 nm (for late wood) and 789.48±160.01 nm (for early wood). The obtained R_a of Norway spruce samples (for late wood) using AFM was 1.22±0.80 µm, using profilometer R_a value (for late wood) was 2.46±0.58 µm. Surface roughness of Norway spruce early wood was too high for both ranges of gauges. Comparing R_a values of late wood, Scots pine shows almost 3 times lower value as Norway spruce. Generally also for other wood species it was found by the authors that surface roughness of late woods presented lower values than early woods [13].

Surface roughness is one of the factors that are affecting the values of the contact angle, thereby affecting also calculation of surface energy. If the measured contact angle is less than 90º the contact angle decreases with surface roughness increasing and the calculated surface energy will increase [7]. From the obtained results this statement is confirmed. The determined surface roughness of Norway spruce wood is higher than Scots pine and the calculated surface energy has the same trend.

To achieve good images in microscopy the slides from samples should be as thin as possible, but the cutting process in the glued sample case is negatively influenced by the fact that the PU is a thermoreactive adhesive and cannot be plasticized with ethylene glycol. It was observed that Glue II fluoresces better than Glue I. If we cut off a thinner sample, we get better quality of sample image in fluorescence microscopy to measure tracheids size/shape and bond-line thickness. The microscopic images indicated four-sided tracheid shapes for early and late wood for both wood species. Average results of wood anatomical structure are summarized in Table 3.

Table 3

Anatomical structure of Scots pine and Norway spruce wood

Parameter	Scots pine		Norway spruce	
	Early wood	Late wood	Early wood	Late wood
Tracheid width, µm	27.33±2.20	17.90±1.33	35.10±13.79	18.74±4.50
Tracheid thickness, µm	29.98±4.76	12.50±2.21	33.06±4.94	12.65±1.37
Tracheid length, µm	529.32±189.58	-	763.27±298.89	-
Bandwidth, µm	1325.00±63.64	-	617.49±143.02	736.14±112.40

From the results it can be concluded that the tracheids size is dependent on the wood species and wood type (early or late wood). Norway spruce width, thickness and length of tracheids are greater than in the Scots pine case. There is very close correlation between the tracheid width and early or late wood width ($R = 0.999$). Coherence between average values of Scots pine late wood width and traheid width is represented in Figure 1. The same trend was concluded by Mäkinen et al. that fast growing Norway spruce trees with wider annual rings had wider tracheids [4]. The obtained results for Norway spruce are in close relationship with the results that are expressed by Trtik et al. – measured tracheids diameter was 35.3±5.9 µm and tracheids length was greater than 700 µm [2]. While Gindl et al. and Mäkinen et al. defined the Norway spruce length of tracheids between 2 and 4 mm and diameter between 20 and 50 µm [1; 4]. In the Scots pine case the length of tracheids is defined in a range from 1-3 mm and the width of tracheids in a range from 29-36 µm [3] that are similar to the obtained results.

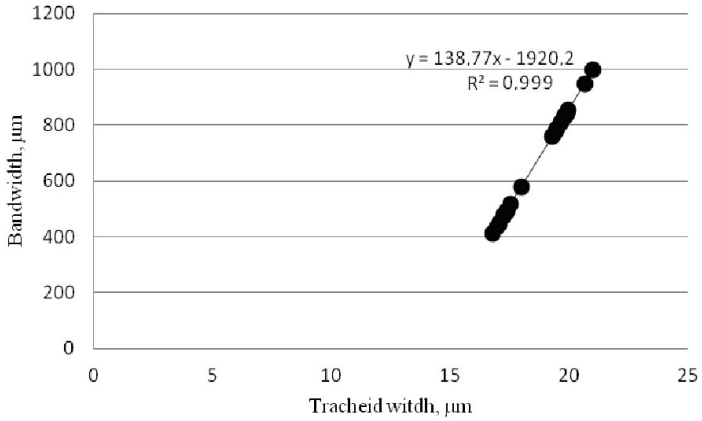

Fig. 1. **Coherence between average values of Scots pine late wood width and tracheid width**

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 the wood structure. Bond-line thickness depending on the wood species, adhesive type and wood moisture content is represented in Figure 2.

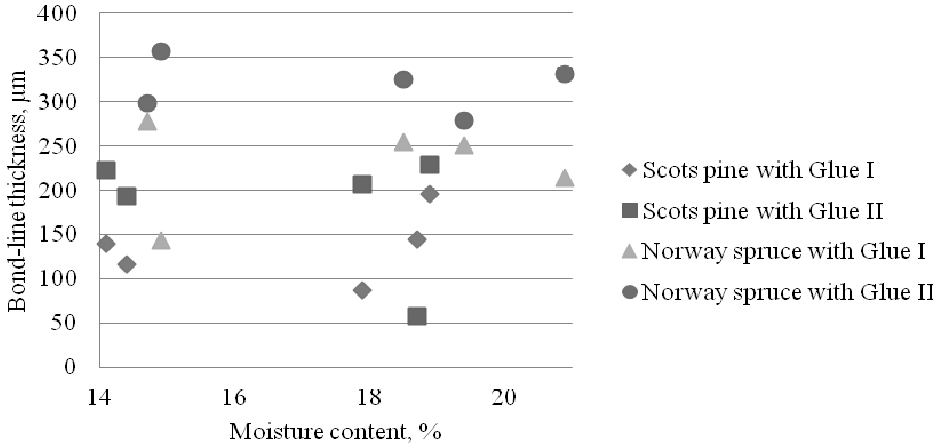

Fig. 2. **Bond-line thickness with 295 kPa working pressure depending on wood species, glue type and wood moisture content**

The obtained results do not show close correlation between the bond-line thickness and moisture content. Nevertheless, these results indicate that bond-line thickness is dependent on the wood species, adhesive type and moisture content. In the Norway spruce with Glue II case we obtained deeper glue penetration into wood. It can be concluded that the penetration depth of adhesive is deeper in Norway spruce wood due to longer average tracheids width, thickness and length than in the case of Scots pine. Bond-line thickness dispersion was large and its values varied from 56.95±27.60 µm (Scots pine with Glue II (MC = 18.7 %)) to 356.82±47.41 µm (Norway spruce with Glue II (MC = 14.9 %)). These results are close to the results of other authors [15], and the average penetration depth of polyurethane glue was about 55 µm (sum of the both sides from glue-line) and the glue bulk layer had average thickness about 100 µm, therefore the overall bond-line thickness was about 155 µm. To achieve good mechanical bond stability, it is necessary that the adhesive penetrates deep enough to bond with undamaged cell walls [15].

Conclusions

1. Bond-line thickness is dependent on different factors such as wood species, adhesive type and wood moisture content and can vary more than six times from 56.95±27.60 µm (Scots pine with Glue II (MC = 18.7 %)) to 356.82±47.41 µm (Norway spruce with Glue II (MC = 14.9 %)).
2. The obtained results show very close correlation (R = 0.999) between both wood species tracheid width and early or late wood width due to better growing conditions.

3. Surface roughness is one of the factors that are affecting the values of the contact angle, thereby affecting also the calculated surface energy. The calculated surface energy of Norway spruce is higher than of Scots pine wood due to higher measured surface roughness. Good correlation was obtained between the Lifshitz–van der Waals component of surface free energy and the wood moisture content.
4. Deeper glue penetration into wood was obtained with Glue II on the Norway spruce surface due to greater average tracheids width, thickness and length than in the case of Scots pine.

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