

TECHNICAL NOTE: HARDWOOD SPECIES BONDING PROPERTIES AFTER CYCLIC CHANGES OF ENVIRONMENTAL CONDITIONS: FIRST TRIALS

Tomáš Pipíška*†

Research Assistant
E-mail: tpipiska@gmail.com

Pavel Král†

Associate Professor
Department of Wood Science
Faculty of Forestry and Wood Technology
Mendel University in Brno
Zemědělská 3, 613 00 Brno, Czech Republic
E-mail: pavel.kral@mendelu.cz

(Received October 2019)

Abstract. The focus of this research was to investigate the bonding quality of the adhesives types after exposure to various moisture and temperature changes. This exposure was to simulate the changes in the interior application, which can cause problems in furniture manufacturing, especially with the usage of different adhesives. Different types of adhesives (Polyvinyl acetate [PVAc], Polyurethane [PUR], urea–formaldehyde [UF], and melamine–urea–formaldehyde [MUF]) were tested for bonding quality on two hardwood species: European beech (*Fagus sylvatica* L.) and English oak (*Quercus robur* L.). Bonding quality was tested based on EN 13354 standard for control specimens, after 8 and 16 cycles of moisture and temperature changes. There was no statistical difference in bonding quality for the PVAc adhesive on beech and oak wood. PUR adhesive showed a significant decrease in the bonding quality of beech wood and no change in oak wood after exposure. The highest reduction in bonding quality after the exposure cycles was found in the UF adhesive due to hydrolysis of the adhesive, regardless of the wood species. There was only reduction in the bonding quality for the MUF adhesive for beech wood after 16 cycles of the exposure condition.

Keywords: Beech, oak, cyclic changes, PVAc, UF, MUF, PUR.

INTRODUCTION

Changes in environmental conditions during the day and part of the year in our houses cause a change in the MC of the wood because of wood hygroscopicity. The amount and direction of the exchange (gain or loss) depend on the RH and temperature of the air and the current amount of water in the wood. Changes in the moisture conditions influence the mechanical properties of composite wood adhesive materials (Simpson and Tenwolde 1999; Vick 1999; Aydin et al 2006; Ren and Frazier 2012).

Durability of the adhesive joints against environmental stress is important for long-term

presentation of the final product. The increase in the MC can significantly decrease the bonding properties. Irrespective of the resin type used, different causes are considered to be responsible for this phenomenon. First, the intermolecular forces between the wood and adhesive may be disturbed if water replaces the reactive groups of the adhesive (Pizzi 1994). Second, moisture-induced dimensional changes of the wood constituents may disrupt the bondline (River et al 1994). Finally, hydrolytic degradation of the resin can cause cleavage of linkages within the adhesive network (Troughton 1969; Ginzel 1973; Robitschek and Christensen 1976; Myers 1982).

Polyvinyl acetate (PVAc) adhesives are well known to be susceptible to MC and heat. These deficiencies are primarily due to the inherent

* Corresponding author
† SWST member

mechanical properties of the PVAc polymer and plasticization of PVAc bondlines by moisture (Goulding 1984; Jaffe et al 1990). The glass transition temperature (T_g ; 35°C) of PVAc homopolymers is relatively low (Goulding 1984). This leads to softening of the PVAc matrix at temperatures above ambient and an associated weakening of bonded joints. Similarly, water exposure has a plasticizing effect and will also reduce the strength of any PVAc bondline (Grigsby et al 2005).

Polyurethane (PUR) adhesives have good strength, are ductile, are resistant to moisture, and cause little emission (Ionescu 2005). PUR exhibits a lower cross-linking density and lower thermal resistance, which was repeatedly reported in the literature for PUR-bonded wood joints, eg (George et al 2003; Richter et al 2006). Recent research demonstrates that thermal softening depends on the formulation of PUR adhesives and could also be reduced by adding filler material (Clauß et al 2011a; Clauß et al 2011) because of the possibility to vary the chemical composition of PUR adhesives to a comparably high degree, a wide range of properties could also be achieved and explained (Stoeckel et al 2013).

Urea–formaldehyde (UF) is one of the most important adhesive in furniture production because of its good performance at a comparably low cost (Dunky and Niemz 2002). However, lack of resistance to weather and water and its susceptibility to emission of formaldehyde vapors are two main disadvantages of UF resin (Pizzi 1994). In the presence of moisture or water, UF resin is slightly hydrolyzed, and the hydrolysis is enhanced under elevated temperatures (Marutzky 1994). Hydrolysis causes degradation of adhesive bonds, leading to loss of strength in UF adhesive

joints (Dunky and Niemz 2002; Ringena et al 2006; Park et al 2009; Paiva et al 2012).

Melamine–urea–formaldehyde (MUF) is used for its higher water resistance. Several improvements can be achieved by increasing the melamine content, such as better bond strength, decreased formaldehyde emission, decreased water absorption, and decreased thickness swelling (No and Kim 2007; Hse et al 2008). The better performance of MUF-bonded boards is usually attributed to its higher chemical resistance to hydrolysis (Park et al 2009).

It is an important conclusion that humidity not only significantly affects the performance of the adherent wood but also affects adhesives typically used in wood bonding themselves. Most wood adhesives are significantly softened by water contact or elevated humidity (Stoeckel et al 2013). After redrying, most of the adhesives (PVAc, MUF, and PUR) regained their original performance (Konnerth et al 2010).

Wood has the ability to absorb and evaporate water, which causes changes in the wood and the bondline. This study shows the results of bonding quality changes caused by changes in wood MC, due to the elevated temperature and changes in humidity. Elevated temperatures and humidity changes in the interior of buildings correspond with the different time of the year. These changes are important for the furniture made from solid wood. This study compares two hardwood species with different wood structures typical for furniture manufacturing. The main comparison was between different PVAc adhesives and other typical adhesives in furniture manufacturing, such as PUR, UF, and MUF.

Table 1. Adhesive spread and pressing parameters for each adhesive.

Adhesive	Manufacturer identification	Adhesive spread ($\text{g} \times \text{m}^{-2}$)	Press time (s)	Pressure (MPa)	Temperature (°C)
PVAc-1	Technobond D3	180	1800	1	20
PVAc-2	Technobond 1KD4	180	1800	1	20
PVAc-3	Multibond SK-8	200	1800	1	20
PVAc-4	Nexo D3	200	1800	1	20
PUR	Neopur 2238	200	5400	1	20
UF	Protodur 303.0	150	480	1	105
MUF	Danafix 458 (hardener H-5108)	150	480	1	105

Table 2. Humidity and temperature changes in time for one cycle of exposure.

	Temperature (°C)	Humidity (%)	Time (min)
1	40	95	240
2	20	65	480
3	30	30	240
4	20	65	480

MATERIALS AND METHODS

Specimen Preparation

The boards were prepared from two wood species: European beech (*Fagus sylvatica* L.) and English oak (*Quercus robur* L.), with a final dimension of 11 × 45 × 600 mm (thickness × width × length). The angle between the annual ring and surface to be bonded was from 30 to 60°. The boards were conditioned in an environmental test chamber (Memmert CTC 256; Memmert GmbH + Co.KG, Schwabach, Germany) at a temperature of 20°C and 65% RH until the EMC is reached.

Adhesives Used

The boards were bonded with seven different adhesives: PVAc (Technobond D3, Technobond 1KD4, Multibond SK-8, and Nexo D3), PUR (Neopur 2238), UF (Protodur 303.0), and MUF (Danafix 458 with hardener H-5108). The UF adhesive was prepared by mixing the adhesive and water in a ratio of 2:1. Meanwhile, the MUF adhesive with the hardener was prepared at a ratio of 10:2.

Manufacturing

After conditioning, the surface of the boards was planed to a final thickness of 10 mm. The

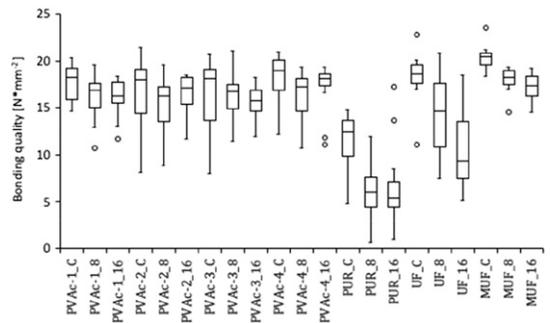


Figure 1. Bonding quality results of beech wood (C-control, eight cycles of exposure, 16 cycles of exposure).

adhesive was applied onto one surface of the paired specimen with the recommended amount by the adhesive manufacturer within 12 h after the planing to avoid oxidation of the bonding surface. The pressing was carried out using a laboratory press (HL 400 MENDELU; Strozatech, Brno, Czech Republic), and the pressing parameters (pressure, press time, and temperature) were maintained as per the recommendation of the adhesive manufacturer (Table 1). Finally, the bonded specimens were conditioned at a temperature of 20°C and 65% RH before testing. Three pairs of bonded specimens were prepared for each adhesive and wood species combination, resulting in a total of 42 bonded pairs.

Testing

Each bonded specimen was further cut into two smaller specimens with a dimension of 20 × 20 × 600 mm (thickness × width × length) by having a bondline in the middle of the specimen. Therefore, there were a total of six test specimens

Table 3. Bonding quality of beech wood for control and exposed specimens.

	Bonding quality ($N \times mm^{-2}$)		
	Control	8 Cycles	16 Cycles
PVAc-1	17.7 (1.9) E, F, G	16.4 (2.0) D, E, F	16.3 (1.7) D, E, F
PVAc-2	16.6 (3.6) D, E, F	15.4 (2.9) D, E	16.7 (1.8) D, E, F
PVAc-3	16.4 (3.9) D, E, F	16.2 (2.1) D, E, F	15.7 (1.7) D, E, F
PVAc-4	18.3 (2.5) F, G	16.2 (2.6) D, E, F	17.4 (2.3) D, E, F, G
PUR	11.8 (2.5) B, C	6.1 (3.0) A	6.3 (3.4) A
UF	18.2 (2.6) E, F, G	14.5 (4.0) C, D	10.3 (3.6) B
MUF	20.3 (1.0) G	18.1 (1.1) E, F, G	17.3 (1.3) E, F

Means with the same letter in columns do not differ statistically by Tukey’s test ($\alpha = 0.05$). Numbers in parentheses represent standard deviation.

Table 4. Bonding quality of oak wood for control and exposed specimens.

	Bonding quality ($N \times mm^{-2}$)		
	Control	8 Cycles	16 Cycles
PVAc-1	14.2 (1.1) H	13.0 (1.0) D, E, F, G, H	13.1 (1.5) D, E, F, G, H
PVAc-2	13.5 (0.9) F, G, H	13.2 (1.0) D, E, F, G, H	13.4 (1.0) E, F, G, H
PVAc-3	13.2 (1.0) D, E, F, G, H	12.9 (0.7) D, E, F, G, H	12.8 (0.7) D, E, F, G, H
PVAc-4	13.0 (1.6) D, E, F, G, H	12.2 (2.0) C, D, E, F	12.6 (0.8) D, E, F, G
PUR	11.8 (2.6) B, C, D	12.0 (1.4) B, C, D, E	10.7 (1.7) B, C
UF	12.6 (1.2) D, E, F, G	10.6 (2.2) B	8.5 (2.3) A
MUF	12.9 (1.0) D, E, F, G, H	13.2 (0.5) D, E, F, G, H	14.1 (1.3) G, H

Means with the same letter in columns do not differ statistically by Tukey's test ($\alpha = 0.05$). Numbers in parentheses represent standard deviation.

for each adhesive and wooden species combination. These six small specimens were divided into three groups based on the exposure cycle period (control, and eight and 16 cycles of condition changing). Control group specimens were conditioned at a temperature of 20°C and RH 65% after bonding. The other two groups were placed in the environmental test chamber Memmert CTC 256 (Mettler Toledo GmbH + Co. kg). One cycle of exposure condition had four steps of environmental exposures: high humidity and low humidity conditions with a relaxation time at a standard condition (Table 2). These high and low humidity conditions relate to the processes in the house during a day and time of the year. One group was exposed to eight cycles and another to 16 cycles. Then, the specimens were conditioned at a temperature of 20°C and RH 65% to reach the EMC.

All specimens were further cut to test according to the EN 13354 standard for a bonding quality test, 22 specimens for each variation. The test was carried out on a Zwick®Z050 universal testing machine equipped with a 50-kN load cell (Zwick GmbH & Co. kg, Ulm, Germany) with a testXpert v11.02 software. Following the EN standard, the crosshead speed was set for specimens to fail within 60 ± 30 s.

Statistical Analysis

The data were processed in STATISTICA 10 software (StatSoft Inc., Tulsa, OK) and evaluated using a one-factor analysis of variance and completed with Tukey's honest significance test (HSD test).

RESULTS AND DISCUSSION

There was no statistically significant difference for beech wood samples in the average bonding quality among all PVAc adhesives (Table 3); however, a slight reduction in bonding quality was observed within the group (Fig 1). The lowest bonding quality for the control group was observed with the PUR adhesive at $11.8 N \times mm^{-2}$, and this value decreased after 8 and 16 cycles with a mean value of $6.1 N \times mm^{-2}$ and $6.3 N \times mm^{-2}$, respectively. A similar trend of reduction in the bonding quality of the PUR adhesive was observed for the UF adhesive with 20% and 43%, respectively. Hydrolysis of the bonds in the UF adhesive occurred when exposed to water/humidity, which caused reduction in the bonding quality (Ringena et al 2006; Park et al 2009; Paiva et al 2012). Meanwhile, there was only statistical significant decrease in the bonding quality of the MUF adhesive after 16 cycles of moisture and temperature changes. The reduction can be caused by a similar mechanism, as in the

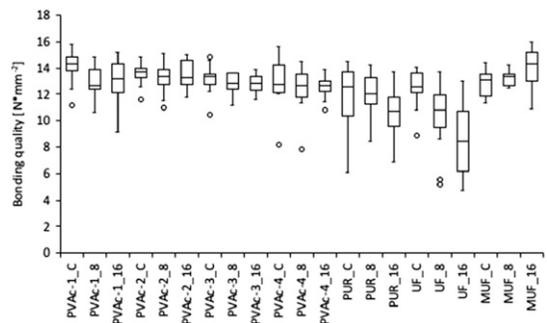


Figure 2. Bonding quality results of oak wood (C-control, eight cycles of exposure, 16 cycles of exposure).

UF adhesive, but in a smaller range because of the addition of melamine in the adhesive, which is difficult to hydrolyze (Paiva et al 2012).

There was no statistical difference in the bonding quality of all PVAc and PUR adhesives for oak wood samples (Table 4). The largest reduction in bonding quality was found in the UF adhesive bonds with a mean value of $12.6 N \times mm^{-2}$, $10.6 N \times mm^{-2}$, and $8.5 N \times mm^{-2}$ for the control, eight cycles, and 16 cycles of exposure, respectively. This reduction trend was the same as that in the beech specimens because of the hydrolysis of urea in the adhesive (Fig 2). Meanwhile, there was no statistical difference in oak specimens bonded with the MUF adhesive.

Results from this study showed some differences, mainly decrease in bonding quality in beech and oak wood specimens. The PVAc adhesives showed the same results for control specimens and after 16 cycles of exposures, and because of these results, we need to increase the number of cycles. There is an assumption that more cycles with changes of high and low humidity with swelling and shrinkage of the wood will cause a higher difference in bonding quality.

CONCLUSIONS

Exposure of the adhesives to the cycles with high and low humidity showed that the UF adhesive is very sensitive, showing a significant decrease in the bonding quality in beech and oak. The influence of the humidity changes on beech wood showed a significant decrease for the PUR adhesive. PVAc adhesives as typical furniture manufacturer adhesives showed the most consistent results in bonding quality. PVAc adhesives, based on the solidification and not curing or reaction during manufacturing, are adhesives with the best reconstruction to the same bonding properties after humidity changes.

This experiment is the first trial experiment based on the comparison of nonstructural adhesives typical for furniture manufacturing. The authors will increase the number of exposure cycles for the same types of adhesives for the verification of the conclusions from this experiment.

ACKNOWLEDGMENTS

This work was supported by the Specific University Research Fund, Internal Grant Agency (IGA) of the Faculty of Forestry and Wood Technology, Mendel University in Brno (LDF_VP_2018023).

REFERENCES

- Aydin I, Colakoglu G, Colak S, Demirkir C (2006) Effects of moisture content on formaldehyde emission and mechanical properties of plywood. *Build Environ* 41(10): 1311-1316.
- Clauß S, Allenspach K, Gabriel J, Niemz P (2011a) Improving the thermal stability of one-component polyurethane adhesives by adding filler material. *Wood Sci Technol* 45(2):383-388.
- Clauß S, Dijkstra DJ, Gabriel J, Kläusler O, Matner M, Meckel W, Niemz P (2011b) Influence of the chemical structure of PUR prepolymers on thermal stability. *Int J Adhes Adhes* 31(6):513-523.
- Dunky, M, Niemz, P (2002) *Holzwerkstoffe und Leime*. Springer-Verlag, Berlin Heidelberg, Germany.
- George B, Simon C, Properzi M, Pizzi A, Elbez G (2003) Comparative creep characteristics of structural glulam wood adhesives. *Holz Roh Werkst* 61(1):79-80.
- Ginzel W (1973) Zur frage der hydrolyse harnstoffharzgebundener holzspanplatten. *Holz Roh Werkst* 31(1):18-24.
- Goulding T (1984) Wood adhesives: Chemistry and technology. Pages 320-350 *in* *British Polymer Journal*. Marcel Dekker, New York, NY.
- Grigsby WJ, Ferguson CJ, Franich RA, Russell GT (2005) Evaluation of latex adhesives containing hydrophobic cores and poly(vinyl acetate) shells: Potential to improve poly(vinyl acetate) performance. *Int J Adhes Adhes* 25(2): 127-137.
- Hse CY, Fu F, Pan H (2008) Melamine-modified urea formaldehyde resin for bonding particleboards. *For Prod J* 58:56-61.
- Ionescu M (2005) Chemistry and technology of polyols for polyurethanes. *Smithers Rapra Press*, Shrewsbury, England.
- Jaffe HL, Rosenblum FM, Daniels W (1990) *Handbook of adhesives*, in I Skeist, ed. Springer US, Boston, MA.
- Konnerth, J, Stöckel, F, Müller, U, Gindl, W (2010) Elastic properties of adhesive polymers. III. Adhesive polymer films under dry and wet conditions characterized by means of nanoindentation. *J Appl Polym Sci* 118:1331-1334. DOI: 10.1002/app.32342.
- Marutzky R (1994) Release of formaldehyde by wood products, Wood adhesives-chemistry and technology, in A Pizzi, ed. Marcel Dekker Inc., New York and Basel.
- Myers GE (1982) Hydrolytic stability of cured urea-formaldehyde resins. *J Wood Sci* 15:127-138.

- No YB, Kim MG (2007) Evaluation of melamine-modified urea-formaldehyde resins as particleboard binders. *J Appl Polym Sci* 106(6):4148-4156.
- Paiva NT, Henriques A, Cruz P, Ferra JM, Carvalho LH, Magalhães FD (2012) Production of melamine fortified urea-formaldehyde resins with low formaldehyde emission. *J Appl Polym Sci* 124(3):2311-2317.
- Park B-D, Lee S-M, Roh J-K (2009) Effects of formaldehyde/urea mole ratio and melamine content on the hydrolytic stability of cured urea-melamine-formaldehyde resin. *Eur J Wood Wood Prod* 67(1):121-123.
- Pizzi A (1994) *Advanced wood adhesive technology*. Marcel Dekker Inc., New York and Basel.
- Ren D, Frazier CE (2012) Wood/adhesive interactions and the phase morphology of moisture-cure polyurethane wood adhesives. *Int J Adhes Adhes* 34:55-61.
- Richter K, Pizzi A, Despres A (2006) Thermal stability of structural one-component polyurethane adhesives for wood—structure-property relationship. *J Appl Polym Sci* 102(6):5698-5707.
- Ringena O, Janzon R, Pfizenmayer G, Schulte M, Lehnen R (2006) Estimating the hydrolytic durability of cured wood adhesives by measuring formaldehyde liberation and structural stability. *Holz Roh Werkst* 64(4):321-326.
- River BH, Ebewele RO, Myers GE (1994) Failure mechanisms in wood joints bonded with urea-formaldehyde adhesives. *Holz Roh Werkst* 52(3):179-184.
- Robitschek P, Christensen RL (1976) Degradation phenomena in ureaformaldehyde resin-bonded particleboard. *For Prod J* 26:43-46.
- Simpson W, Tenwolde A (1999) Physical properties and moisture relations of wood, *in* *Wood handbook—wood as an engineering material*. Department of Agriculture, Forest Service. Forest Products Laboratory, Madison, WI.
- Stoeckel F, Konnerth J, Gindl-Altmatter W (2013) Mechanical properties of adhesives for bonding wood—A review. *Int J Adhes Adhes* 45:32-41.
- Troughton GE (1969) Accelerated aging of glue-wood bonds. *J Wood Sci* 1:172-176.
- Vick CB (1999) Adhesive bonding of wood materials, *in* *Wood handbook—wood as an engineering material*. US Department of Agriculture, Forest Service. Forest Products Laboratory, Madison, WI.