

BEHAVIOUR OF WOOD JOINTS GLUED BY ONE COMPONENT POLYURETHANE ADHESIVE MODIFIED BY COTTON FLAKES AND BASALT FIBERS

Petra LACIKOVÁ¹, Jan VANĚREK¹, Adam BĚŤÁK¹

¹ Institute of Technology of Building Materials and Components, Faculty of Civil Engineering, Brno University of Technology, Veveří 331/95, 602 00 Brno, Czech Republic

lacikova.p@fce.vutbr.cz, vanerek.j@fce.vutbr.cz, betak.a@fce.vutbr.cz

DOI: 10.35181/tces-2022-0018

Abstract. The main objective of this paper is to describe the influence of cotton flakes and basalt fibers on the resulting tensile properties of one-component polyurethane and the tensile shear strength of glued lap joints of beech wood. Fillers are an integral part of the modification of adhesives, especially because they influence rheological and mechanical properties. A one-component polyurethane adhesive for waterproof joints was used for bonding. The amount of filler was optimized by determining the tensile properties of adhesive as a material. The basalt fibers improved the tensile properties of one-component polyurethane. The average tensile strength value for basalt fibers in the amount of 5% was found to be 30% higher than for basalt fibers in the amount of 1%. The resulting properties of glued joints were determined by tensile shear strength according to standard ČSN EN 302-1. It was found that the shear strength of the bonded beech joints was higher for modified one-component polyurethane by cotton flakes than those modified by basalt fibers of 7 mm length. Significant effect was shown for cohesive failure of lap joint especially for modification by cotton flakes when the value increased by 10% to reference wood joint.

Keywords

Wood adhesive, one-component polyurethane, fillers.

1. Introduction

Wood is a renewable material. By processing wood and its subsequent modification into the required shapes and lengths, various wooden elements with modified properties for the given construction purposes are provided. The mechanical properties of wood depend on

humidity. As humidity increases, the strength and stiffness characteristics of the given wood decrease [1], [2].

Glued wooden structures are used quite often because joining structural elements using glue is considered the most perfect connection. From the point of view of stress on the structure, it is possible to choose wood based on the class, dimensions, and sections of the cross-section, thereby achieving the required load-bearing capacity. Currently, natural plant adhesives (starches, natural resins, pectins, alginates) and natural animal adhesives (glue, bone) are not widely used in construction. There is a wide range of adhesives on the market for glued joints. Epoxy and polyurethane adhesives are mainly used for solid structural joints. Despite the higher costs, polyurethane adhesives show a considerable number of advantages. There is no emission of formaldehyde, curing takes place in a short time, less pressing pressure is required during gluing and the joint shows higher elasticity [3].

However, polyurethane adhesives still show deficits when gluing different types of wood, especially when gluing some hardwoods and when exposed to high moisture content [4], [5].

At the beginning of joining the wooden elements, the glue must have a suitable viscosity and good workability. The glue penetrates the wood cells that are on the surface of the glued surface. During curing, the glue will bond with the surface of the glued part of the wood. This interaction of the glue with the wood's surface affects the joint's bearing capacity. In addition to humidity, environmental temperature is a very important factor that affects the bonding process. External glued wooden structures are exposed to various climatic conditions throughout their lifetime, which affect the mechanical properties of the wood, the adhesive system, and their interaction [6], [7].

When modified with additives and fillers, it is possible to significantly influence the properties of the adhesive as a material and the properties of the resulting glued joint. Just as there are many types of adhesives to produce wood-based products, the market also offers a wide range of fillers, the choice of which depends on the requirements for the material and the application of the final product [8], [9].

This work aimed to assess the possibility of modifying a moisture-curing one-component polyurethane adhesive with fibrous filler and to monitor its effect on the strength and durability parameters of the glued joint. By using these fillers, a more load-bearing joint is expected, better resistance to volume changes, as well as saving glue or reducing the penetration of glue into the wood structure.

2. Materials and methods

2.1. Materials

For this experiment, a one-component polyurethane glue (PUR system 2010, AkzoNobel, Sweden) was used. This PUR adhesive is used for load-bearing structures. The characteristic of the PUR system 2010, which was used is shown in Tab. 1.

The following types of fillers were used: cut basalt fibers with a length of up to 7 mm and cotton flakes. The photomicrographs of used fillers from the optical microscope are shown in Fig. 1. The basalt fibers were obtained by cutting chopped roving with a surface treatment in the form of silane groups. The average fiber thickness is 5 μm . Other parameters are listed in the technical sheets of the producer (Basaltex, a.s., Czech Republic).

Cotton flakes R+G (Havel composites CZ s.r.o., Czech Republic) are used to increase the viscosity of adhesives, sealants, and coating with an effect already during the addition of a small amount. This type of filler shows good adhesion to polyurethane resins. The cotton flakes are white in colour and the average length of fibers ranges from 150 to 500 μm .

Tab. 1: Characteristics of one-component polyurethane adhesive AkzoNobel PUR system 2010.

Characteristics	AkzoNobel 2010
Product	isocyanate MDI
Form	liquid
Colour	white
Viscosity	6,000 – 19,000 mPa.s
Solid content	100 %
Density	1,160 kg/m ³

The initial amount of filler in weight percent was chosen based on scientific research related to the modification of wood adhesives with particulate or fibrous fillers [8], [10] and [11]. All investigated

parameters of the modified adhesive were compared with the parameters of the pure one-component polyurethane adhesive without filler. Tab.2 shows the mixture composition used to prepare the dogbone samples.

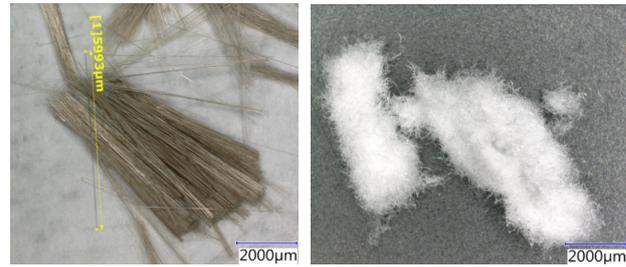


Fig. 1: Photomicrographs (magnification 200x): a bundle of basalt fibers with a length up to 7 mm (left), cotton flakes (right).

Based on the determination of the properties of the one-component polyurethane glue as a material, the optimal values of the filling were selected for wood joints. For this experiment, beech wood with a density of $680 \pm 30 \text{ kg/m}^3$ at equilibrium humidity ($12 \pm 1 \%$) was used. The durability of the glued joints was verified by determining the shear strength of the lap joints after exposure stress in selected treatments.

Tab. 2: Mixture composition of dogbone samples to determine tensile properties.

Designation	Type of filler	Amount of 1C-PUR [%]	Amount of filler [%]
R	-	100.0	-
		99.0	1.0
		98.0	2.0
		97.5	2.5
		97.0	3.0
		96.5	3.5
B ₇	Basalt fibers lengths of 7 mm	96.0	4.0
		95.0	5.0
		99.0	1.0
		98.0	2.0
		97.5	2.5
C	Cotton flakes	95.0	5.0
		90.0	10.0

2.2. Preparation of test samples

The modified adhesive was homogenized by manual mixing. The prepared admixture was poured into a silicone form. The samples were cured in a vacuum oven covered with foil to reduce polyurethane foaming due to the release of CO₂. The samples were unformed and adjusted to the desired shape. To determine the tensile properties of the adhesive as a material, ten samples for each mixture with dimensions (Fig. 2) according to the ČSN EN ISO 527-1 were prepared [12].

After optimizing the amount of filler, three amounts of filler (1.0%, 3.0%, and 5.0%) were selected for glued joints. The adhesive in the amount of 300 g/m² was applied to the surfaces of the glued beech wood. Two wood plates with a thickness of 5 mm were glued to a

panel assembly with a final thickness of 10 mm by applying a pressure of approximately 0.6 N/mm^2 for twenty minutes. Five strips with a width of $20 \pm 0.1 \text{ mm}$, a length of $150 \pm 5 \text{ mm}$, and a thickness of $10 \pm 0.1 \text{ mm}$ were cut from one prepared panel assembly in the direction of the wood fibers. Grooves were cut into individual strips transversely to the wood fibers. The cut separated the layers of wood and did not touch the glue layer. The samples were prepared according to the ČSN EN 302-1 [13]. The samples were subjected to selected exposure treatment according to Tab. 3.

Tab. 3: Type and duration of treatment before tensile shear testing.

Designation	Treatment
A1	No treatment other than conditioning in standard climate [20/65]
A3	4 days soaking in cold water at $(20 \pm 5) \text{ }^\circ\text{C}$ Recondition in standard climate [20/65] to original mass
A5	6 h of soaking in boiling water 2 h of soaking in cold water at $(20 \pm 5) \text{ }^\circ\text{C}$ Recondition in standard climate [20/65] to original mass

2.3. Experimental procedures

The tensile properties of dogbone samples were determined according to the ČSN EN ISO 527-100 [12]. The TIRA test 2850 S device was used for the determination (Fig. 2). A strain gauge was placed on the working part of the samples, the crosshead constant rate of 1 mm/min was set until the test specimen broke. The maximum applied force was recorded. The tensile strength of the material was calculated, and the modulus of elasticity was determined.

The tensile shear strength of lap joints of beech wood was determined according to the standard ČSN EN 302-1 [13]. The testing machine TIRA test 2850 S was used (Fig. 2). The crosshead constant rate of 1 mm/min was set until the test specimen broke. After the failure of the test specimen, the maximum applied force was recorded. The tensile shear strength determinations were performed for each set of specimens until ten valid measurements were obtained. For modified adhesives, the clusters of filler were analysed with the digital optical microscope Keyence VHX-950F.

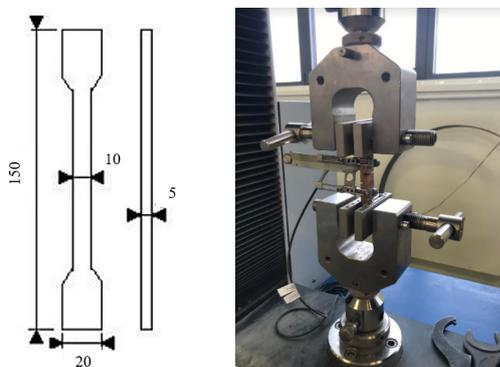


Fig. 2: The shape of the test sample “dogbone” (left image), a tensile testing machine with the clamped test specimen of lap joint (right image).

3. Results and discussion

3.1. Tensile strength of dogbone samples

The optimal amount of filler was chosen based on the trend of tensile strengths of modified polyurethane for a certain type of filler. The addition of the filler allowed a better release of CO_2 from the glue matrix during the curing reactions. Thanks to the basalt fibers, a more compact structure of the sample with better tensile properties were obtained. The values of the tensile properties are summarized in Fig. 3 and Fig. 4.

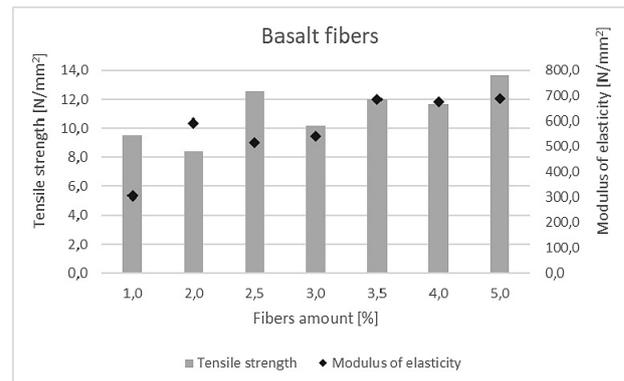


Fig. 3: The effect of the addition of basalt fibers (7 mm) on tensile strength and modulus of elasticity.

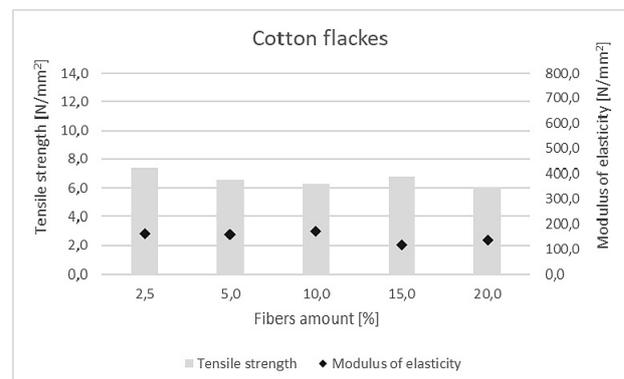


Fig. 4: The effect of the addition of cotton flakes on tensile strength and modulus of elasticity.

Higher average values of tensile strength and modulus of elasticity were obtained by filling with basalt fibers. By increasing the amount of basalt fibers, the tensile properties also increased. The average tensile strength value for basalt fibers in the amount of 5% was found to be 30% higher than for basalt fibers in the amount of 1%. The average value of tensile modulus of elasticity for basalt fibers in the amount of 5% was found to be 56% higher than for basalt fibers in the amount of 1%.

When cotton flakes were used, the samples were less porous than the reference, but compared to the filler in the form of basalt fibers, the samples showed a significantly lower tensile modulus of elasticity. The average tensile strength values for all amounts of filler fluctuated around the value of 6 N/mm^2 . The addition of

filler affected the tensile properties of the polyurethane, but further increasing the amount did not show an effect. Due to the workability of the modified adhesive, the limiting amounts of filler of 5% were selected.

3.2. Shear strength of glued joints

The summary evaluation of the results for the use of basalt fibers in the bonded joint is shown on the bar chart in Fig. 5. and Fig. 6.

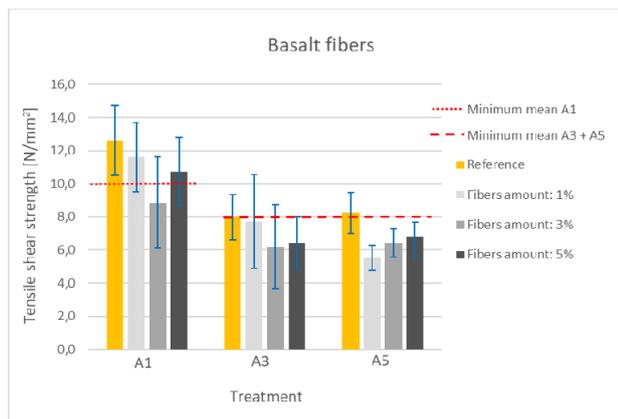


Fig. 5: The effect of the addition of basalt fibers (7 mm) on the shear strength of the glued joint.

The highest shear strength values were achieved in standard climate conditions without treatment (A1). In the case of using basalt fibers, based on the previous stage, a significant effect after treatment was expected. The shear strength for filling with basalt fibers with lengths up to 7 mm after treatment A3 and A5 did not meet the requirements of standard ČSN EN 15425 [14]. The average shear strength values for basalt fibers in all used amounts after treatment A3, and A5 were lower than for the reference. The use of basalt fibers with lengths up to 7 mm in the glued joint appears to be ineffective. After the shear test the samples glued modified one-component polyurethane with basalt fibers had a higher adhesion failure, on average 60%.

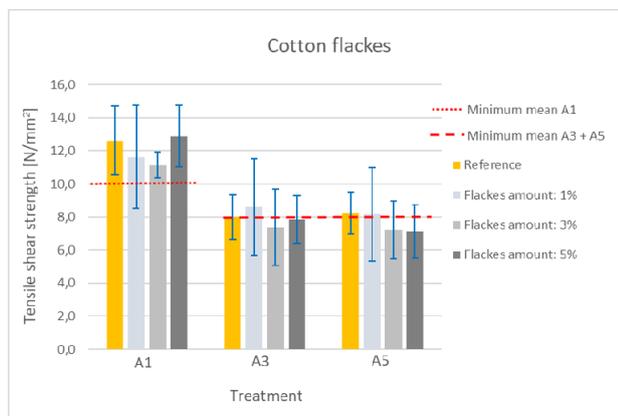


Fig. 6: The effect of the addition of cotton flakes on the shear strength of the glued joint.

The shear strength values for pure one-component

polyurethane (reference samples) and filling with cotton flakes of 1% after all treatments meet the requirements of standard ČSN EN 15425 [14], which sets out a minimum bonded joint shear strength of more than 10 N/mm² for treatment A1 and 8 N/mm² for treatments A3, A5. The average shear strength value for cotton flakes in the amount of 1% after treatment in the cold water (A3) was found to be 7.5% higher than for the reference. After hydrothermal treatment in A5, no positive effect of cotton flakes was noted. The samples bonded with one-component polyurethane adhesive with 1% cotton flakes had a significantly higher failure in the wood, on average of 70%.

3.3. Microscopic analysis of the fractured joints

The shear surface of the bonded joint after the tensile shear test is shown in Fig. 7 and Fig. 8 from an optical microscope.

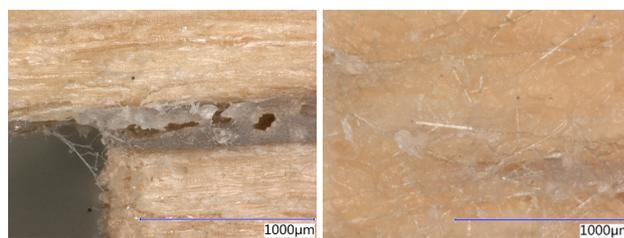


Fig. 7: The shear surface of a glued joint is modified with an adhesive with basalt fibers.

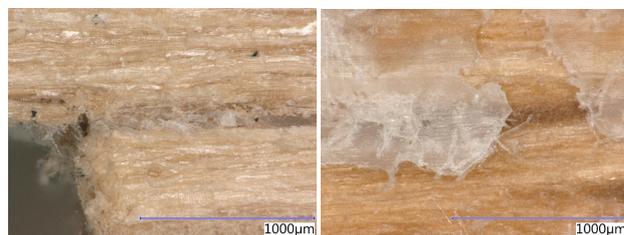


Fig. 8: The shear surface of the joint is glued with modified glue with cotton flakes.

From the photomicrographs of the glued joint after the tensile shear test, it is obvious that the bonded line contains a relatively large number of cavities and there was no formation of close contact. This could be caused by the low pressing pressure or the long-time of applying the glue to the surface of the wood. The fibers of both fillers were not oriented, but some places of the shear surface contained clusters after optical microscopy. These places could be the reason for the adhesion failure at the interface between the glue and the wood in the glued joint.

4. Conclusion

The determined values confirmed the different effect of fillers on the properties of 1C-PUR as a material, and the effect of fillers on the properties of 1C-PUR in interaction

at the interface of glue and wood. The addition of both fillers to the polyurethane matrix helped release CO₂ as a product of the curing reactions, consequently, it reduced the foaming of the polyurethane samples. The determination of tensile properties of polyurethane samples were a suitable optimization process for choosing the optimal amount of fillers. The effect of adhesive modification to improve the flexibility of adhesive which could ensure to withstand the volume changes of the wood were not proven by tensile shear strength properties. Only the amount of 1% of cotton flakes appeared to be the optimal amount to fulfil the strength limit of bonded joints. On the other hand, improvement of cohesive failure of lap joint was determined for modified polyurethane by cotton flakes (amount of 1%) used compared to reference polyurethane samples. The basalt fibers did not fulfil the standard strength requirements therefore a shorter length (than the used length of 7 mm) could have a greater benefit in the hydrothermal stressing of the glued joint. Further research will therefore be focused on improving the length of basalt fibers, reducing the time of application of adhesive on wood surfaces, and increasing the pressing pressure to make close contact without a large number of cavities.

Acknowledgements

This paper was supported under the Internal Grant Agency of Brno University of Technology, Faculty of Civil Engineering, specific junior research grant number FAST-J-22-8034 with project name: *Optimization of the amount of filler in the wood adhesive for the finger joints and assessment of its effect on the bonded joint after hygro-thermal stress*, and the Czech Science Foundation, under project No. 21-20645S with project name: *Characterization of modified isocyanate-based adhesives for engineered wood products*.

References

- [1] RŮŽIČKA, Martin. *Moderní dřevostavba*. Praha: Grada, 2014. ISBN 978-80-247-3298-5.
- [2] *Dřevěné konstrukce podle Eurokódu 5*. Zlín: KODR, 1998. ISBN 80-238-2620-4.
- [3] SVOBODA, Luboš. *Stavební hmoty. 2. přeprac. a dopl. vyd.* Bratislava: Jaga, 2007. ISBN 978-80-8076-057-1.
- [4] KONNERTH, Johannes, Marcel KLUGE, Georg SCHWEIZER, Milica MILJKOVIĆ a Wolfgang GINDL-ALTMUTTER. Survey of selected adhesive bonding properties of nine European softwood and hardwood species. *European Journal of Wood and Wood Products* [online]. 2016, 74(6), 809-819 [cit. 2022-11-06]. ISSN 0018-3768. DOI: 10.1007/s00107-016-1087-1.
- [5] MOHD YUSOF, Norwahyuni, Paridah MD TAHIR, Seng Hua LEE, Mohammad Asim KHAN a Redzuan MOHAMMAD SUFFIAN JAMES. Mechanical and physical properties of Cross-Laminated Timber made from Acacia mangium wood as function of adhesive types. *Journal of Wood Science* [online]. 2019, 65(1) [cit. 2022-11-06]. ISSN 1435-0211. DOI:10.1186/s10086-019-1799-z.
- [6] ROWELL, Roger M., ed. *Handbook of wood chemistry and wood composites*. 2nd ed. Boca Raton: CRC Press, c2013. ISBN 978-1-4398-5380-1.
- [7] REN, Dakai a Charles E. FRAZIER. Wood/adhesive interactions and the phase morphology of moisture-cure polyurethane wood adhesives. *International Journal of Adhesion and Adhesives* [online]. 2012, 34, 55-61 [cit. 2022-11-06]. ISSN 01437496. DOI: 10.1016/j.ijadhadh.2011.12.009.
- [8] CLAUB, Sebastian, Karin ALLENSPACH, Joseph GABRIEL a Peter NIEMZ. Improving the thermal stability of one-component polyurethane adhesives by adding filler material. *Wood Science and Technology* [online]. 2011, 45(2), 383-388 [cit. 2022-11-06]. ISSN 0043-7719. DOI: 10.1007/s00226-010-0321-y.
- [9] CAO, Long, Xiaojian ZHOU a Guanben DU. Wood Adhesive Fillers Used during the Manufacture of Wood Panel Products. In: FLORES HUICOCHEA, Emmanuel, ed. *Fillers* [online]. IntechOpen, 2021, 2021-2-3 [cit. 2022-11-06]. ISBN 978-1-83962-435-3. DOI: 10.5772/intechopen.91280.
- [10] HÝSEK, Štěpán, Přemysl ŠEDIVKA, Martin BÖHM, Ondřej SCHÖNFELDER a Rudolf BERAN. Influence of Using Recycled Polyurethane Particles as a Filler on Properties of Polyurethane Adhesives for Gluing of Wood. *BioResources* [online]. 2018, 13(2), 2592-2601 [cit. 2022-11-06]. ISSN 1930-2126. DOI: 10.15376/biores.13.2.2592-2601.
- [11] CHEN, Heyu a Ning YAN. Application of Western red cedar (*Thuja plicata*) tree bark as a functional filler in pMDI wood adhesives. *Industrial Crops and Products* [online]. 2018, 113, 1-9 [cit. 2022-11-06]. ISSN 09266690. DOI: 10.1016/j.indcrop.2018.01.005.
- [12] ČSN EN ISO 527-1, Czech Office for Standards, Metrology, and Testing (2020).
- [13] ČSN EN 302-1, Czech Office for Standards, Metrology, and Testing (2013).
- [14] ČSN EN 15425, Czech Office for Standards, Metrology, and Testing (2017).