

The quality of the wood bonding depending on the method of applying the selected thermoplastic biopolymers

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Abstract: *The quality of the wood bonding depending on the method of applying the selected thermoplastic biopolymers.* The aim of the research was to determine the effect of the method of applying the biopolymer on the surface of bonding solid wood elements on the quality of the obtained adhesive connection. The results of conducted mechanical research show that the highest average value of shear strength was observed for birch lamellas bonded with PLA, both with the first and second method of application. In case of estimating the quality of the bonding of wooden elements, better results were achieved for PLA and the second method of application the "green" adhesive.

Keywords: biopolymer, polylactic, polycaprolactone, bonding line, shear strength, density profile

INTRODUCTION

Currently, wood-polymer composites (WPC) are widely available on the world market and their production is constantly growing. Most of these materials are based on thermoplastic matrices, such as polyolefins: polyethylene (PE), polypropylene (PP) and polyvinylchloride (PVC), because the melting and processing process takes place at relatively low temperatures, while maintaining resistance to degradation under the influence of high temperatures (Madyan et al. 2020). WPC are widely used, i.a. in construction, automotive and engineering industries. The global vision of the market of plastics produced in the amount of 6.3 billion tonnes in the last 70 years has a significant impact on the development of new composites, the main advantage of which will be biodegradability (Okunola A et al. 2019). The mechanical properties of biodegradable polymer composites are on a similar level to that of petrochemical products (Kuciel et al. 2020). The ecology and the idea of sustainable development forces scientists around the world for research and experiments, the aim of which is to search for alternative materials that will reduce dependence on fuels and petroleum products, minimizing the negative impact on the environment throughout their life cycle (Tănase et al. 2015). Due to the current environmental problems, such as: depletion of fossil resources, environmental pollution, greenhouse gas emissions (Bugnicourt et al. 2014; Yu et al. 2006), there is a growing interest of wood-polymer composites (WPC), produced in a thermoplastic matrix derived from renewable raw materials. Commonly used substitutes for petropolymers are: polyhydroxyalkanoates (PHA), which include, among others polyhydroxybutyrate (PHB), polylactic acid (PLA) and polycaprolactone (PCL) (Amache et al. 2013; Chan et al. 2019; Nagarajan et al. 2016). Polyhydroxyalkanoate matrix composites become real competition to conventional WPC technology with polyolefins (Markarian 2008). PHAs, except melting, have a lower viscosity than commonly used polyolefins, which features can be an advantage in the case of composite technology (Gatenholm et al. 1992).

WPCs are made by mixing the matrix (plastics, biopolymer) and filler (fibers, wood flour) and additives (coupler agents, pigments). The most used WPC mixing systems are co-rotating and counter-rotating twin-screw extruders (Singh and Mohanty 2007; Plackett et al. 2003), injection molding (Bledzki and Jaszkievicz 2010; Ozyhar et al. 2020), compression

molding (Torres-Giner et al. 2018). Biopolymers are most often used as thermoplastic matrix in wood-polymer composites (WPC), as described above. However, researchers also testing biopolymers, polyhydroxyalkanoates (PHA) or natural and renewable source adhesives as binders in the production of wood-based materials: layered composites (Bakken and Taleyarkhan 2020a), particleboards (Baskaran et al. 2019) or fibrous composites (Domínguez-Robles et al. 2018). Summary of the current state-of-art about biopolymers applications in the composites based on lignocellulosic raw materials has been described by Gumowska and Kowaluk (2020). Plackett et al. (2003) manufactured 2 mm thick layered composites using commercial PLA and jute fibers (*Corchorus capsularis* L.). The polylactide, that was used in these testing, was in the form of drops, while the jute fibers were in the form of a non-woven mat with a weight of 300 g/m². Using a single-screw extruder, the PLA drops were converted to 0.2 mm thick film, then layered composites with dimensions 300 mm × 120 mm have been prepared, in which sections of jute fibers mat were put into a set with several PLA film layers on both sides. The press temperature was 180-220°C and the pressing time 3 or 10 minutes. Reinforcement by 40% jute fibers almost doubled increase the tensile strength compared to pure PLA. The researchers proved that tensile strength increase is depend on temperature in the heating stage of the process.

According to Raghu et al. (2018), interfacial adhesion is an essential aspect of the mechanical properties of WPC or blends, and reactive interfacial coupling agents are often used to improve the adhesion properties. These researchers confirmed the improvement of mechanical properties of TPS (thermoplasticized starch)-filled PLA blends (30%, 50%) by reinforcing the blends with wood fibers (20%, 40%) with using maleic anhydride grafted PLA (10% MA-g-PLA) as the coupling agent. At 40% wood fibers and 10% MA-g-PLA load, tensile strength increased by 128%, and bending strength showed about 180% improvement over TPS/PLA blends. They proved the rightness of using MAg-PLA as a coupling agent for such blends and composites.

The production of WPC, especially by the injection molding and extrusion method, using biopolymers as a thermoplastic matrix, has been widely researched. The scope of research concerning use of polyhydroxyalkanoates in wood-based materials and methods of their reinforced / modification is limited. Therefore, the aim of the research was to characterize the effect of the method of applying the biopolymer on the surface of bonded solid wood elements, on the quality of the obtained adhesive connection.

MATERIALS AND METHODS

Bonding of the lamellas

Depending on the method of applying the biopolymer to wooden elements, the samples were produced in two variants:

1st method – according to the procedure described in the publication by Gumowska and Kowaluk 2020.

2nd method – not less than 10 overlap samples made of air dry birch (*Betula pendula* Roth) lamellas of dimension of 110x22x7 mm³ and with sawmill surface roughness ($R_a=13.66 \mu\text{m}$, $R_y=98.63 \mu\text{m}$, $R_q=18.13 \mu\text{m}$, which was measured by Gumowska and Kowaluk 2020) have been prepared for every thermoplastic biopolymer like as: industrially pure polylactide (PLA) and polycaprolactone (PCL) in the form of 3 mm diameter drops. The overlap (bonding line) nominal size was about 22x17 mm². The adhesive mass for individual types of binders (PLA and PCL) were produced by dissolving the granules of biopolymers in solvent to achieved the consistency of a thick liquid. The solvents which have been used to achieve the binders was methylene chloride for PLA and toluene for PCL. Solutions with a concentration of 65% were prepered. After preparing the solutions, the excessive amount of biopolymers was applied to the lamellas surfaces, the open time was 24 hours. After 24 hours, the biopolymers on the

lamellas were heated to a temperature of 180° so that the previously applied layer of adhesive was softened and became a thick liquid able to bonding. Then, the lamellas with the biopolymer spread out on both surface (overlap) of lamellas, have been connected and pressed with use of hand carpentry clamp to remove the surplus of biopolymer and cool down the bonding line to room temperature.

Shear strength testing and in-wood damage evaluation

The shear strength of the manufactured samples have been measured on standard universal testing machine, where the samples have been loaded by tension to be broken within 60±30 s, and the maximum load [N] has been registered. Prior to the loading, the real dimensions of the bonding line have been measured. The shear strength was calculated as a maximum load [N] referred to the bonding line area [mm²]. After the break of the sample, every destroyed zone was analyzed if the break occur in-wood structure, and the area of in-wood destruction was in organoleptic way estimated (in % of total bonding line area) with accuracy of ±10%.

Density profile

The density profile of composites samples were analyzed used a GRECON DA-X measuring unit. The measurement based on direct scanning X-rays was carried out with a speed of 0.05 mm/s and across the panel thickness with a sampling step of 0.02 mm. The density profile was determined using one sample of each variant for a method of application and type of binder. All the samples to tests were cut into nominal dimensions of 50x50 mm² x thickness of composites.

For a wider analysis of physical properties, the width of the bonding line was measured for samples of each type of binder and the method of application. The measurement was carried out using an optical microscope Nikon SMZ 1500.

All the tested samples were conditioned prior to the tests in 20°C/65% relative humidity (RH) to achieved constant weight.

On the basis of above mentioned measurements, the statistical significance of the average values between factors as well as levels, has been estimated by analysis of variance (ANOVA) and t-tests calculations ($\alpha = 0.05$).

RESULTS

Shear strength testing and in-wood damage evaluation

The results of measurement of shear strength of birch lamellas bonded with use of selected thermoplastic biopolymers, are visualized in Figure 1. The highest average value of shear strength is 5.88 N/mm² for 1st method and bonded with PLA. The highest average value of shear strength was observed for birch lamellas bonded with PLA, for both, the 1st and 2nd method of application. The lowest average strength value was obtained for the samples bonded 1st method with PCL as adhesive. Considering the values of standard deviations, it can be noted that the mean values of shear strength confirmed statistically significant differences between the average values of the shear strength and the types of thermoplastic biopolymer used for 1st method. In the case of 2nd method, there are no statistically significant differences between the strength average value and the type of biopolymer. The spread of particular results around the average (sd) is at a similar level for all types of composites. These investigation confirmed the possibility of obtaining interfacial adhesion between PLA and the wood surface for both methods of application the binder. Luedtke et al. (2019) conducted research on layered composites bonded with PLA in order to obtain information on the morphology of adhesive

line and mechanical properties of layered materials. The mechanical properties were influenced by the type of wood species from which the veneers were obtained (maple, beech) and the processing temperature of PLA. There were no differences in the use of amorphous or semi-crystalline biopolymer grades. Determination of quantification of interfacial strength showed composites prepared at more than 160 °C has higher mechanical properties. In their research Bakken and Taleyarkhan (2020b) developed and analyzed two methods of adhesive application: PLA in the form of dry powder and wet spraying (emulsion). They used of two PLA polymer-based adhesive formulations based on crystalline and amorphous resins to producing and testing 2 and 3 layered plywood. Researchers confirm the possibility of using PLA as a binder for laminated composites, taking into account different application methods. The highest shear strength values were achieved by emulsion spray using amorphous PLA.

The results of the in-wood damage evaluation for 2nd method at least half of the PLA samples - was 20%, while for the PCL samples, there was 0%, still damage was in the bonding line, like as 1st method. In the case of PLA - 2nd method, it cannot be concluded that the damage occurred to a large extent in the wood zone, but it should be added that the damage is greater than in the case of PLA lamellas bonded with 1st method.

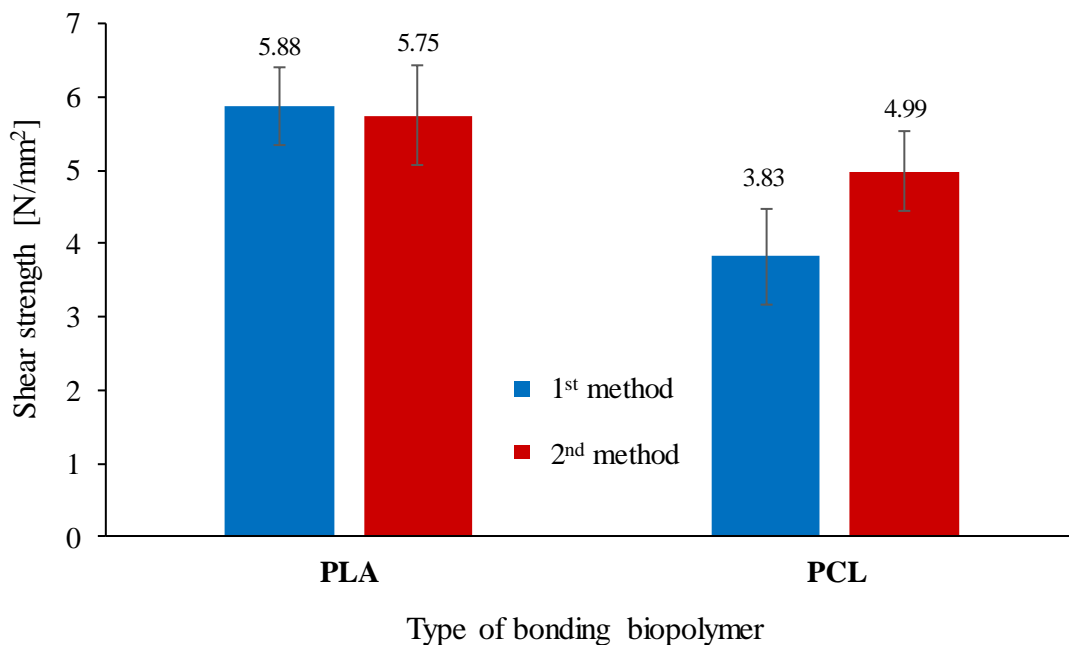


Figure 1. Shear strength of tested samples

Density profile

The results of measurement of density profiles of the produced composites are presented on Figure 2. The graph shows the density distribution on the cross-section of the composites for 4 representative samples of each type of binder and the method of application. Considering the graphs below, the significant increase in the density was noticed exactly in the middle of the composites thickness, in the bonding line. Above the main graph of density profiles was included the additional graph which showing a close-up of the density of bonding line itself on the cross-section of samples.

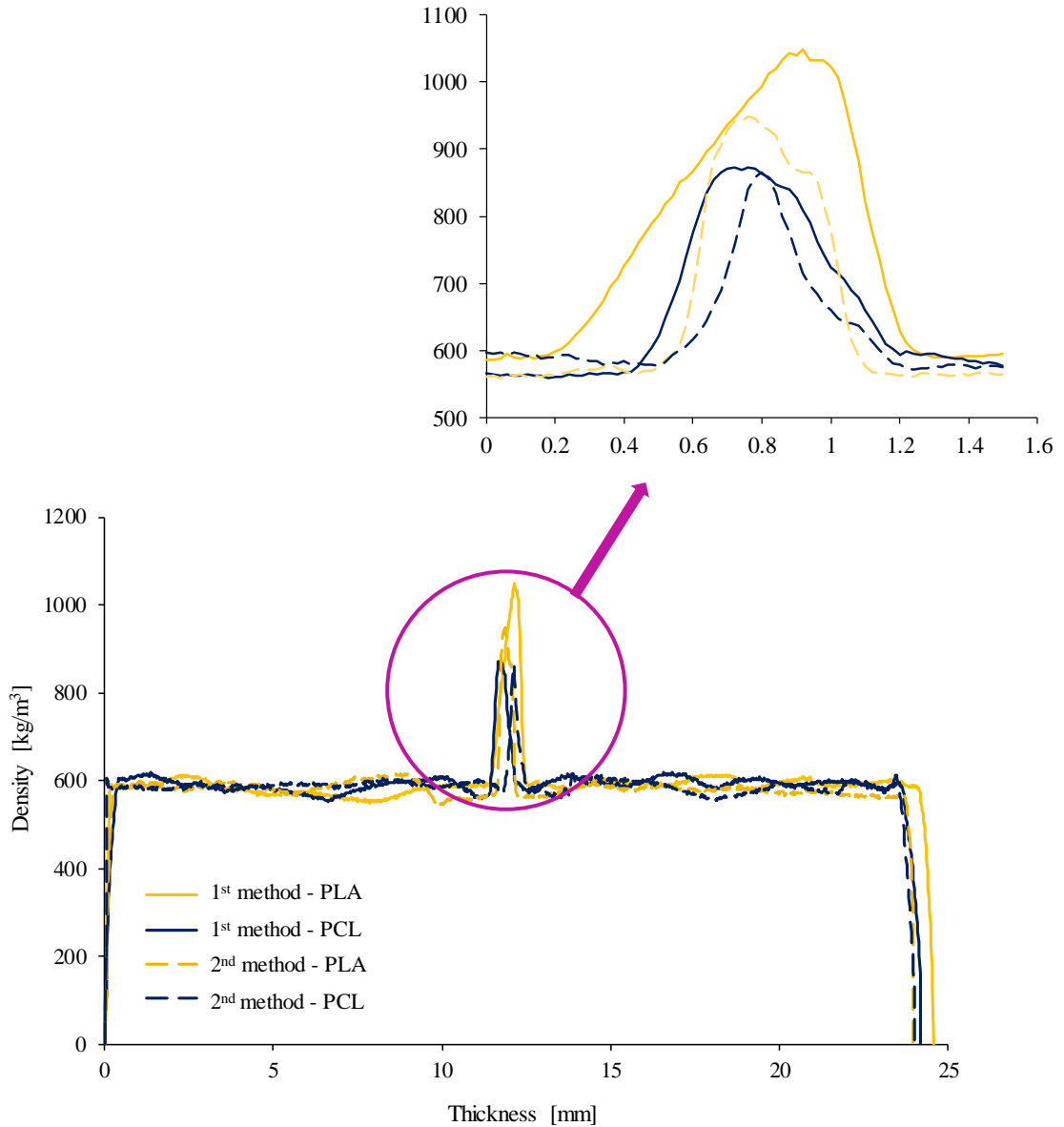


Figure 2. Density profiles of composites bonding under different method and adhesives

For better understanding and analysis the adhesion of biopolymer and wood surface, single graphs were prepared taking into account mainly the width of the bonding line (Figure 3). The width of bonding line was shown as a shaded area in the each graphs. The graphs were prepared in this way to check what penetration of the biopolymer into the wood structure looks like. For 2nd method, the width of the just adhesive line was in the range of approx. 0.05-0.1 mm, while for 1st method, the width was approx. 0.25-0.3 mm. This can mean that the biopolymers applied in liquid form (dissolved) better penetrate into the wood structure. But this does not sufficiently explain the poor tensile shear strength of the PCL samples for 2nd method. The explanation of the achieved results probably can be found in the mechanical properties of pure biopolymers, where the tensile strength for PLA is higher than for PCL (Chee et al. 2013). According to mentioned above research investigated by Luedtke et al. (2019) the higher tensile strength was related with a thinner bonding line. Penetration deep into the wood contributed to a better physical interlocking of the PLA matrix within the wood ultrastructure and related increases in interfacial strength.

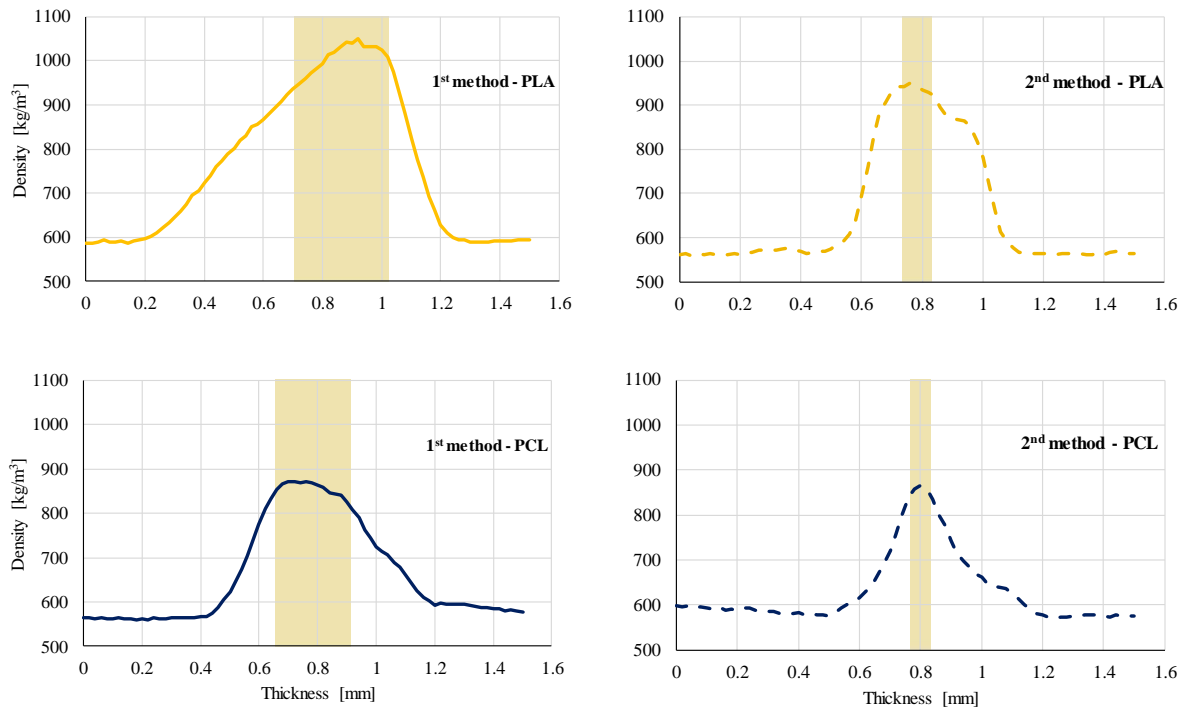


Figure 3. Single density profiles for each variant of composites

CONCLUSIONS

According to the conducted research and the analysis of the achieved results, the following conclusions and remarks can be drawn:

1. The shear strength was the highest for samples bonded with PLA, while the lower strength values have been obtained when PCL was used as a binder taking into account all variants of produced birch lamellas.
2. The share of in-wood damage for at least half of the samples bonding with PLA and produced using the second method was more than 20%.
3. The second method of application biopolymers was better in context of the quality of the bonding of wooden elements.

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Streszczenie: Jakość sklejania drewna w zależności od sposobu nanoszenia wybranych biopolimerów termoplastycznych. Celem badań było określenie wpływu sposobu nanoszenia biopolimeru na powierzchnię łączonych elementów drewnianych na jakość uzyskanych połączeń klejowych. Przeprowadzone badania wykazały, że najwyższą średnią wartość wytrzymałości na ścinanie przez rozciąganie otrzymano dla kompozytów lameli litego drewna brzozy klejonych PLA, zarówno pierwszym, jak i drugim sposobem nanoszenia biopolimerów. W przypadku oceny jakości sklejania elementów drewnianych lepsze wyniki uzyskano dla PLA przy drugim sposobie aplikacji „zielonego” kleju.

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