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Use of innovative environmentally friendly adhesives for wood veneer bonding

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Abstract. Hot-melts based on polyolefines (high density polyethylene, polypropylene) and thermoplastic textile fibre waste (polyurethane, polyamide-6) were used for wood veneer bonding. Investigation of their shear (adhesive) strength, bending strength, and water resistance showed that these adhesives could be used for birch wood veneer bonding and producing plywood for application in outdoor conditions.

Key words: birch wood veneer, hot-melts, polyolefines, textile waste, shear strength, bending strength, water resistance.

INTRODUCTION

Use of glues in the manufacturing of wood composite products has increased in recent years because large amounts of wood composite materials are needed for various purposes. One of the most important wood composites is plywoods laminated materials. Initially thermosetting glues based on phenol-formaldehyde, amino-formaldehyde, melamine-formaldehyde, as well as epoxy and urethane resins were used in the production of coniferous plywoods and other sheet materials [1,2].

Ecological claims on decreasing emissions of dangerous volatile products from wood composite materials manufacture have been increasing. Therefore many investigators are challenged to solve this problem. One of the possibilities is partial replacement of toxic products in glues containing formaldehyde with natural, ecologically friendly substances such as tannins, lignins, starch, etc. Complete elimination of the toxicity of glues is possible only if for wood bonding thermoplastic polymers (hot-melts) like polyolefines or biodegradable synthetic or natural polymers like polyvinyl alcohol, starch, polylactic acid, soy, benzilated lignocellulose products, etc. are used [3–6]. For example, polyolefines (polyethylenes, polypropylenes, ethylene vinyl acetate copolymers) were shown to serve as excellent glues (hot-melts) for wood veneer bonding in plywoods [3–5]. The durability of polypropylene and modified polypropylene glues is equal to that of melamine- and phenol-formaldehyde resins. Molten polyolefines make good contact with the veneer surface and penetrate into the lumina of wood cells and other spaces. As scanning electron microscopic investigations show, separated glue lines and casts of glue penetrate into the vessels and form mushroom-like projections. They fill the bordering pit cavities and it is indicated that polypropylene melt dominantly ensures good mechanical adhesion into various wood elements and spaces in the veneer. The depth of penetrated polyolefin glues can reach 30-40 microns in wood surface [5]. The moisture content (8-25%) of core veneer does not influence gluebility [4]. All adhesive joints show good water resistance [5,6].

The population growth in the world promotes increasing utilization of plastics, and therefore the amount of plastic waste has been growing considerably. In developed countries the plastic content in the domestic waste stream can reach 18–20 kg for one person per year. Recycling of these polymers would diminish the primary polymer consumption, helping to solve environmental protection problems. Recycled thermoplastic polymers are promising as hot-melts too [6].

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The goal of this work was to examine the possibility of using different thermoplastic textile waste products as hot-melts for wood veneers for manufacturing plywoods and to compare them with previously used hotmelts and thermosetting resin glues.

MATERIALS AND METHODS

In experiments a birch wood veneer with a thickness of 1.5 mm, made in A/S "Latvijas finieris" (Latvia), was used as substrate. High density polyethylene (HDPE) of grade 276-73 with melt flow index (MFI) 0.8 g/10 min, two kinds of polypropylene (PP) (Mosten GB (PPgb) with MFI 1.9 g/10 min and Mosten MA (PPma) with MFI 12.4 g/10 min), thermoplastic polyurethane textile fibre waste (PUW) with MFI 6.4 g/10 min (P = 2.16 kg, $T = 230 \,^{\circ}\text{C}$), and polyamide-6 textile fibre waste (PAW) were used as adhesives. Adhesive films (0.3-0.4 mm thick) from all polymer materials were made by compression moulding (temperature 140-220°C, pressure 1–2 MPa, time 1–3 min.) The gluing of single overlap joints of wood veneer-hot-melt adhesive-wood veneer (WAW) was performed in hydraulic press (time 1-8 min, pressure 2 MPa) at different temperatures (140-230°C). The geometry of specimens for shear strength tests was as in [5]. For the bending strength test 3-layer plywood samples with perpendicular direction of cellulose fibre in the middle veneer layer to the outer veneer layer of plywood were prepared. Shear strength of single overlap joints was examined as in ASTM D-3164-73 and water resistance according to EN 312-1:2 as in [5]. Bending test (three points method, distance between supports L = 100 mm) was made on universal dynamometer UTS-10 (deformation rate 2 mm/min, size of samples $130 \text{ mm} \times 25 \text{ mm} \times 5 \text{ mm}$) corresponding to ASTM D-790 and ASTM D-3043-87.

RESULTS AND DISCUSSION

Previous investigations showed that promising glues for wood veneer bonding are lignosulphonates modified with amines, used separately or together with 4.4¹diphenylmethanediisocionate (PMDI) [6] and hotmelts of polyolefines [5]. These glues allow forming strong adhesive joints with good water resistance and decreasing the toxicity of glues in comparison with those utilized in plywood production now.

The next step was to form hot-melts for wood veneer bonding from thermoplastic synthetic textile fibre waste. Textile waste usually contains various finishing products on the fibre surface, which could hinder adhesive interaction between the adhesive and wood veneer. Therefore the influence of dressing on the shear strength of adhesive joints (Fig. 1) was checked first of all. The results showed that the influence of textile fibre waste (PUW) treatment before its utilization as hot-melt was not significant. The best values were obtained in the case of acetone treated PUW, when the adhesive strength of specimens increased approximately by 15%. Considering that pretreatment of waste requires an additional technological procedure, untreated textile waste was used in the next experiments.

Results of the experiments with PAW and PUW used as hot-melts are presented in Fig. 2 and Fig. 3. Kinetic curves (Fig. 2) show that the optimal technological parameters for wood veneer bonding with PAW hot-melts at 2 MPa pressure are temperature 220–230 °C, time 4.5–5 min. The adhesive strength of glued veneer samples can be even as high as 10 MPa.



Fig. 1. Dependence of shear strength (σ_b) of single overlap veneer joints of laminated WAW systems on adhesive PUW pretreating methods. 1, untreated fibres; 2, compression moulded PUW film treated with acetone for 10 min; 3, fibres treated with cold water for 10 min; 4, fibres treated with acetone + water 5 + 5 min; 5, fibres treated with acetone for 10 min; 6, fibres treated in boiling water for 10 min.



Fig. 2. Relationship between shear strength (σ_b) of single overlap veneer joints of laminated WAW systems and contact time. Adhesive: PAW, contact temperatures 210–230 °C, and pressure 2 MPa.



Fig. 3. Relationship between shear strength (σ_b) of single overlap joints of laminated WAW systems and the kind of adhesive and contact pressure. Contact temperatures: 140 °C (HDPE), 200 °C (PUW), 220 °C (PAW). Contact time: 2 min.

Contact pressure influence on specimens' shear strength is presented in Fig. 3. The influence of pressure on the adhesive strength of the samples was not significant and 2 MPa contact pressure insured maximum values. The best result was obtained for PAW hot-melt (8.5 MPa), which was higher in comparison with HDPE (7 MPa) and PUW (5 MPa). A mixed adhesive-cohesive fracture mechanism was visually observed practically for all examined single overlap veneer joints. These results confirm that adhesive strength in laminated wood systems is determined not only by the interfacial interaction intensity between the wood surface and the glue layer, but also by the glue's own cohesive strength. Therefore it is important to investigate PAW adhesive, which has the best strength and modulus for the formation of strong adhesive layers during lamination. At the same time PAW hot-melts have low viscosity and can form a larger contact surface between hot-melt and wood. It is significant as mechanical adhesion theory of adhesive joints in wood laminated systems often observes [4,5].

Comparison of the efficiency of different hot-melts used for birch veneer gluing with the strength of an industrially produced plywood adhesive is presented in Fig. 4. All investigated hot-melts gave considerably higher adhesive strength than the industrial plywood glued with phenol-formaldehyde resins. The best adhesive joints were obtained when PAW and PP hotmelts were used. The shear strength of single overlap veneer joints was 3–5 times higher than for 4 mm industrial plywood. These results evidently confirm our previous supposition about a significant role of adhesive– cohesive strength in developing strong adhesive joints.

An important exploitation property of plywoods is not only their high shear strength, but also the stability of their adhesive strength during utilization under different conditions. Therefore the water resistance of adhesive joints was investigated. Tests results are presented in Table 1.



Fig. 4. Comparison of shear strength (σ_b) of industrially produced 4 mm plywood (FF) and single overlap veneer joints of WAW laminated systems with different adhesives.

 Table 1. Comparison of the water resistance of industrially produced 4 mm plywood (FF) and single overlap veneer joints of specimens glued with different adhesives

Sample	Shear strength, MPa				Applica-
	Dry samples	After 24 h exposure in water	After 4 h boiling +16 h drying +4 h boiling in water	After 72 h boiling in water	tion in outdoor condi- tions
FF (industrial plywood)	2.50	2.22	2.03	2.02	+
WAW with PPgb adhesive	8.72	8.40	7.78	6.85	+
WAW with PPma adhesive	6.90	-	-	4.42	+
HDPE	7.62	_	2.34	2.10	+
PUW	5.64	_	2.18	1.35	+

- No data.

It is difficult to expect the formation of water-stable covalent chemical bonds in the investigated systems. Water resistance tests are passed if after 4 + 4 h boiling the shear strength of samples is still over 1.8 MPa (1stclass plywood water stability) and after 72 h boiling more than 1.0 MPa (3rd-class plywood). All investigated hotmelts (Table 1) can be used for the production of plywoods with 3rd-class water stability, which is excellent water resistance, and may be used for exploitation in outdoor conditions. Good water stability results can be explained by the swelling of the wood surface under exposure to water and creation of additional friction forces, which do not let the adhesive to be pulled out from the wood surface. Similarly, increasing penetration of hot-melt glues in the lumina of wood cells and promotion of water resistance in laminated wood systems was observed for other systems [7,8].

If plywoods are envisaged for use in different bended constructions, it is necessary to predict the material's bending properties (strength, modulus, deformation ability). The relevant results are given in Figs 5–7.

Plywoods' bending strength depends on the used hot-melt (Fig. 5). The highest strength was achieved in samples glued with HDPE and PP grade Mosten MA



Fig. 5. Comparison of bending strength (σ_t) of industrially produced 4 mm plywood (FF) and 3-layer plywood with different adhesives.



Fig. 6. Comparison of flexural modulus (E_f) of industrially produced 4 mm plywood (FF) and 3-layer plywood with different adhesives.



Fig. 7. Comparison of bending deformation (ε_f) of industrially produced 4 mm plywood (FF) and 3-layer plywood with different adhesives.

hot-melts. In these samples bending strength values were approximately 20% higher than for industrial 4 mm plywood. It is known that an increase of adhesive strength leads to the growth of bending strength, but in our investigations this correlation was not observed (Figs 4 and 5). This suggests that plywood's bending strength does not depend only on the intensity of adhesive interaction on the surface between the substrate and adhesive and boundary layers, but to a great deal on the adhesive material's own strength and modulus of elasticity.

During bending plywood veneer and hot-melt material layers deform similarly. The bending stresses concentrate on the weaker border surface and plywood fracture takes place through delamination of layers. This can diminish the bending strength of plywood.

The kind of adhesive used is not significant for bending modulus (11.5-12 GPa) (Fig. 6). According to [9], plywood modulus is defined mainly by the material of its outer layers. In our experiments it was always birch wood veneer. One exception was observed during tests with polyurethane textile waste (PUW) used as hot-melt. In this case low values of bending modulus (about 1.5 GPa) were registered. It can be explained by the fact that the used polyurethane is an elastomeric, damping material [10], which can absorb and dissipate energy in the plywood structure. Therefore for plywood containing PUW adhesive the highest deformation ability before fracture was observed. It was up to 65% higher than for the industrially produced plywood control samples (see Fig. 7). Higher deformation ability in comparison with the control specimen was gained with other hot-melts too. Obviously all investigated hot-melt glues make plywoods more flexible without simultaneously decreasing the modulus of elasticity (Fig. 6). Furthermore, for PP, HDPE, and PAW hot-melt adhesives a direct relationship was observed between shear strength and bending deformation ability. When adhesive strength was higher, the deformation ability before fracture was lower (Figs 4 and 7). Increase of flexibility of plywoods glued with the investigated hot-melts simplifies the production of different bended constructions from plywoods.

CONCLUSIONS

In this work the use of various thermoplastic textile waste products as hot-melts for gluing birch wood veneers for manufacturing plywoods was studied. Comparison with our previously investigated polyolefine hot-melts and industrial plywood made on the basis of phenol-formaldehyde adhesive was made. The following conclusions were drawn:

• Use of high-density polyethylene, polyurethane, and polyamide-6 textile fibre waste hot-melts for wood veneer gluing guarantees the shear strength (5.6–

10 MPa) of the material that considerably exceeds the adhesive strength of industrial plywood (2.5-3.7 MPa) glued with phenol-formaldehyde resins.

- Water resistance tests showed water stability of all samples glued with the investigated hot-melts, and thus the possibility of their use under outdoor conditions.
- Bending strength depended on the used hot-melt. The highest values were observed for specimens glued with polypropylene Mosten MA and highdensity polyethylene hot-melts. These samples had a good deformation ability too.
- The influence of the kind of the hot-melt on the bending modulus was not significant. An exception was polyurethane textile waste hot-melt. This confirms the prediction that plywood bending modulus is defined mainly by the modulus of the material of the outer layer.
- No direct relationship was observed between the bending strength and adhesive strength of plywoods. For glues based on hot-melts the determinant factor for getting a high adhesive strength was a high cohesive strength and hot-melt material's own modulus.
- As the used polyurethane is elastomeric, different properties in comparison with other investigated hotmelts were observed: low modulus of elasticity (1.5 GPa) and high deformation during bending. These plywoods are suitable for the production of various bended constructions.
- Our investigations showed that hot-melts of thermoplastic polymer materials based on primary and recycled waste could successfully be used for wood veneer gluing with high exploitation properties of overlap joints. Their use would eliminate glue toxicity and considerably improve environmental protection.

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REFERENCES

- Kinloch, A. J. Adhesion and Adhesives: Science and Technology. Chapman and Hall, London and New York, 1987.
- Beand, F., Niemz, P., and Pizzi, A. Structure properties relationships in one-component polyurethane adhesives for wood: sensitivity to low moisture content. J. Appl. Polym. Sci., 2006, 101, 4181–4192.
- Young-Jun Park and Hyun-Joong Kim. Hot-melt adhesive properties of EVA/aromatic hydrocarbon resin blend. *Int. J. Adhes. Adhes.*, 2003, 23(5), 383–392.
- Goto, T., Salki, H., and Onishi, H. Studies of wood gluing. XII. Glueability and scanning electron microscopic study of wood polypropylene bonding. *Wood Sci. Technol.*, 1982, 16(4), 293–303.
- Kajaks, J. A., Bakradze, G. G., Vīksne, A. V., Reihmane, S. A., and Kalnins, M. M. The use of polyolefines-based hot-melts for wood bonding. *Mech. Comp. Mat.*, 2009, 45(6), 643–650.
- Grinbergs, U., Kajaks, J., and Reihmane, S. Usage of ecologically perspective adhesives for wood bonding. *Scientific Journal of Riga Technical University: Material Science and Applied Chemistry*, 2010, 22(1), 114–118.
- Smith M., J., Heming, D., and Karthik, R. Wood-thermoplastic adhesive interface – method of characterization and results. *Int. J. Adhes. Adhes.*, 2002, 22(3), 197– 204.
- Burhanettin, S. Usual bonding strength and dimensional stability of laminated veneer lumbers manufactured by using different adhesives after the steam tests. *Int. J. Adhes. Adhes.*, 2005, 25, 395–403.
- 9. Rowell, R. M. Handbook of Wood Chemistry and Wood Composites. Tailor & Francis, London, 2005.
- Zia, K. M., Bhatti, H. N., and Bhatti, I. A. Methods for polyurethane and polyurethane composites recycling and recovery: a review. *Reactive and Functional Polymers*, 2007, 67(8), 675–692.

Uudsed loodussõbralikud liimid vineeri valmistamiseks

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Polüolefiinide (kõrgtihe polüetüleen, polüpropüleen) ja termoplastsete tekstiilikiudude (polüuretaan, polüamiid-6) jääkide kuuma sulasegu kasutati puidukihtide liitmiseks vineeris. Nihke- ja paindetugevuse ning veekindluse uuringud osutavad nimetatud liimi sobivusele välistingimustes kasutatava vineeri valmistamiseks.