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BONDING OF BIRCH VENEER WITH HIGH MOISTURE CONTENT USING PHENOL-FORMALDEHYDE RESIN MODIFIED BY SOY PROTEIN

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ABSTRACT

The possibilities of gluing veneer of birch (*Betula pubescens* Ehrh.) at moisture content of 15% with phenol-formaldehyde (PF) resins modified by soy protein isolate (SPI) 92% are examined in this paper. The amount of added modifying agent was 1, 3 and 5 mass units on 100 mass unit of phenol-formaldehyde resin and pressing temperature of plywood was 130 or 150 °C. The influence of the soy protein isolate on the properties of the developed adhesive compositions (viscosity, pH, solid content, gelation time, pot life) and prepared plywood (shear strength) were evaluated. Pressing conditions of plywood were: pressure 1.0 MPa; time 6 minutes and during the last 30 seconds the pressure was lowered to 0 MPa; glue spread – 150 g/m^2. It was found that the modification of phenol-formaldehyde resin by SPI allows to bond veneer at moisture content of 15%. Results indicated that the shear strength of plywood specimens glued at pressing temperature of 150 °C were higher than those glued at pressing temperature of 130 °C. However, the obtained values of the shear strength of the plywood were higher than the requirement (1.0 MPa) of the standard EN 314-2 for exterior bonding quality.

Key words: veneer, moisture content, PF resin, plywood, soy protein.

INTRODUCTION

Rotary cut veneer with moisture content (MC) of 6 ± 2% is usually used in plywood production. For such moisture content, the conventional thermo-reactive adhesives provide high quality bonding of plywood with physical and mechanical performances that meet European standard requirements. The use of over dried veneer (less than 6% moisture) reduces the quality of plywood by worsened wetting and insufficient transfer of adhesive to another surface, brittleness of veneers and increases energy consumption during drying process (near 60%) that affects the price of the finished products (BEKHTA 2003, ŠATANOVÁ *et al.* 2015, SEDLIAČIKOVÁ *et al.* 2015, HAJDÚCHOVÁ *et al*. 2016).

The quality of veneer directly affects the formation quality of plywood. Cracks that appear as a result of drying, sorting and transporting veneer sheets are the main reason of decreasing of veneer grade (MAHÚT *et al.* 2006). It can be saved by increasing the elasticity of wood by drying veneer to higher final moisture content. The increase of moisture leads to decrease of the number and size of cracks in veneer sheets. In this connection, it is necessary to dry veneer to higher moisture content (over 8%) for the decreasing energy consumption and the increasing quality of plywood.

However, there are some difficulties with gluing veneer with higher moisture content (W > 6%): increasing adhesive penetration by the flow inside the vessel network through the veneer thickness, high vapour pressure causing steam blisters or blows, decreasing the viscosity of the applied adhesive, and loss of the veneer thickness due to the compression (BEKHTA *et al*. 2014).

The early research studies were oriented towards benefits of gluing high moisture veneers (BESINOVA *et al*. 1997) and dealt with certain methods of using: glue foils and glue powders (URBANIK *et al*. 1997, ELBEZ 1997, JOZWIAK 2007); faster-curing adhesive system (CLARK *et al*. 1988); vacuum presses or change of veneer preparation before gluing (interchange of wet and dry veneer sheets in the assembly, application of hygroscopic substances on the surface of veneer) (BEKHTA 2003, ORTYNSKA *et al*. 2013). One of them involves mixing a high molecular weight resin with alkylene carbonates or phenol-resorcinol-formaldehyde resins (CLARKE *et al*. 1990). However, the application of these methods requires using of expensive adhesives or replacement of some equipment that is unaffordable from the economical point of view.

Recent developments and trends in the field of eco-efficient bonding technology contribute to both ecological and economical aspects. Several new trends are in the application of vegetable carbohydrates or proteins, namely as modifiers of existing adhesives. Addition of polysaccharides or soy proteins to traditional synthetic wood adhesives after partial hydrolysis and modifications was reported (PIZZI 2006). Some studies were performed on possibilities of bonding high moisture veneers using phenol-formaldehyde resin filled by hydrolysed soy protein, wheat starch, rye flour and tannin (GHAHRI *et al*. 2016, WU *et al*. 2016, VIJAYENDRAN *et al*. 2000, BEKHTA *et al.* 2008).

Soy protein polymers have been reconsidered as alternatives to petroleum polymers in single-use items to prevent environmental pollution recently (SUN *et al*. 1999). Soy protein is an abundant by-product of the soybean oil industry, and is commonly used as a nutritional additive in food. Being a natural biomaterial, soy protein isolate contains a higher proportion of protein than is present in any other soy protein products, which gives enhanced film forming ability (SU *et al*. 2012). The most commonly produced isolates have high soluble protein content, protein dispersibility indexes (PDI) of 80–90%, and as a consequence, they are highly-valued functional additive. Soy protein isolate obtained is a creamy-coloured powder and on analysis yields a protein content of 90–95% (on a dry weight basis) and a PDI in the range of 80–90% (KUMAR *et al*. 2002).

Blends of soy protein and phenolic resins are useful in finger jointing of green lumber (CLAY *et al.* 1999). Such blends cure rapidly at room temperature have excellent water resistance and reduced formaldehyde emission. Addition of soy protein, soy flour, soy hydrolysate and casein to PF resin resulted in a decrease in reactivity with increasing amount of protein without affecting the emission of formaldehyde (KUMAR *et al*. 2002). However, these adhesives were recommended for cold gluing of wood and hot gluing of veneer with MC $\leq$ 8% (LI *et al*. 2014) but using of soy proteins for hot gluing of veneer with high moisture content (over 8%) was not investigated.

Therefore, the main objective of this study was the development of new PF adhesives modified by soy protein for hot gluing of high moisture content veneer.

MATERIALS AND METHODS

Rotary cut veneer sheets of birch wood (*Betula pubescens* Ehrh.) with dimensions of 300 × 300 mm and thickness of 1.5 mm without visible defects were prepared for the experiments. The average density of birch veneer is 600 ± 20 kg/m^3. High moisture content level of 15 ± 2 % was achieved by conditioning of dry veneer sheets inside a chamber at constant temperature over sulphuric acid solution with concentration of 30 %, till achieving the desired level of moisture content. The moisture content was controlled using electronic moisture-meter and exactly stated by weight method.

Adhesive mixtures were prepared from commercial PF resin (Lignofen, LERG SA, Pustkow, Poland) which was modified with organic additive soy protein isolate with protein content of 92%. This additive is inexpensive and nontoxic. Soy protein isolate is a widespread and renewable agricultural resource. Soy protein was sourced from commercial supplier. Soy protein isolate is the creamy-coloured powder with protein content of 92 % and protein dispersibility index 92. The amount of added modifying agent was 1, 3, and 5 mass units on 100 mass unit of PF. The formulations of adhesive compositions are shown in the Table 1. Adding of soy protein isolate into PF resin allows taking excessive moisture from the adhesive because it absorbs and swells in water very well. In addition, it prevents the penetration of glue into wood and through thin layers of veneer on the front surface of the plywood.

Viscosity, hydrogen ions concentration (pH), solid content, gelation time, pot life of adhesive compositions and shear strength were evaluated.

The viscosity and pH value were determined using rotary viscometer RHEOTEST RV 2 and pH meter HANNA HI 221 accordingly. The pot life and gelation time of adhesives at 20 °C and 150 °C were determined. Solid content of adhesives was determined according to EN 827 standard.

Tab. 1 Formulations of adhesive compositions.

Formulation of adhesive	Additives, mass units	
	PF resin	Soy protein isolate (92%)
A (control)	100	-
B	100	1
C	100	3
D	100	5

Three layer plywood panels were pressed using hydraulic laboratory press FONTIJNE. Pressing conditions of plywood were: temperature 130 or 150 °C; pressure 1.0 MPa (that is 44.4% less than pressure used in traditional plywood manufacture); time 6 minutes and during the last 30 s the pressure was lowered to 0 MPa; glue spread 150 g/m^2. Such lower value of pressing pressure were chosen because previous studies found that the high moisture content veneer are compressed better than dry veneer (BEKHTA *et al.* 2011).

PF adhesive mixture was applied onto one side of every uneven ply. The plies were assembled perpendicularly to each other (veneer sheets were laid up tight/loose) to form plywood of three plies. Glue was applied on the veneer surface with a hand roller spreader.

Laboratory testing machine LABORTECH 4.050 was used for the evaluation of the quality of gluing and the shear strength of plywood was determined according to EN 314-2 after pre-treatment for intended use in exterior conditions. Testing samples were immersed in boiling water for 4 h, dried in the ventilated drying oven at 60 ± 3 °C for 16 h, immersed in boiling water for 4 h, followed by cooling in water at 20 ± 3 °C for at least 1 h to decrease the temperature of test pieces to 20 °C. Ten samples were used for each variant shear strength mechanical testing. During experiment, all plywood samples were conditioned before testing for two weeks at 20 ± 2 °C and 65 ± 5 % relative humidity.

RESULTS AND DISCUSSION

Homogeneous adhesives were obtained in the case of adding soy protein isolate to PF resins. Basic properties of obtained adhesives are presented in Table 2. Adding of soy protein isolate into PF resin allows taking excess water from the adhesive, because it swells and absorbs water. So, the viscosity of PF adhesive B increased in 1.6 times, adhesive C in 6 times and D in 52 times compared with reference PF resin, due to active adsorbing of water from the adhesive by the macromolecules of modifying extenders. The adhesive viscosity was 9,185 MPa·s at 5 mass units addition of soy protein for modifying PF resin, which is slightly higher than viscosity used for plywood manufacture. Modified phenol-formaldehyde adhesives might prevent from over-penetration into the wood surface and through thin layers of veneer on the front surface of the plywood. Therefore, the adhesive D leads to poor flow and it is not easy to spread on the surface for bonding.

Increasing amount of modifying agent from 1 to 5 mass units leads to decreasing pot life of all developed adhesives, which value ranges from 1 to 5 hours. The value of pot life is closely related to the viscosity of the glue composition: an increasing of the viscosity leads to the decreasing of pot life.

Tab. 2 Basic properties of developed adhesives.

Properties of adhesives	Formulations of adhesives			
	A	B	C	D
Viscosity, MPa.s	174	278	1,049	9,185
Pot life, h	>5	>5	>3	1
pH	12.0	11.9	11.8	11.8
Gelation time at 150 ºC, s	120.4	135.6	121.6	91.2
Solid content, %	46.5	46.8	48.9	50.2

The solid content and concentration of hydrogen ions (pH) of B, C, D adhesives remain virtually unchanged and are in the range from 46.8 to 50.2% and from 11.8 to 11.9, respectively.

The gelation time of PF adhesives (B, C, D) modified by soy protein isolate ranges from 91.2 to 135.6 seconds. It was established that gelation time decreases with the increase in SPI content in the adhesive composition. This pattern of changes in viscosity and gelation time of adhesive compositions are appropriate and can be explained by the strong chemical reaction between functional groups of soy protein and PF resin by forming cross-linked thermoset matrix (VIJAYENDRAN *et al*. 2000).

The effect of SPI content on shear strength of plywood is shown in Figure 1. The values of shear strength of samples were over the requirement (1 MPa) of EN 314-2 standard.

In the case of increasing of SPI amount from 1 to 3 mass units, the shear strength of plywood is increasing. The unfolded molecules of SPI increase the contact area and adhesion onto other surfaces, and they become entangled with each other during the curing process to retain bonding strength (YANG *et al*. 2006, SUN *et al*. 1999). However, the increase of SPI amount to 5 mass unit leads to the decrease of shear strength of plywood. In this case, worse glue wetting of the surface of veneer and thickening of the glue layer is observed and all of that leads to the reduction in shear strength of plywood.

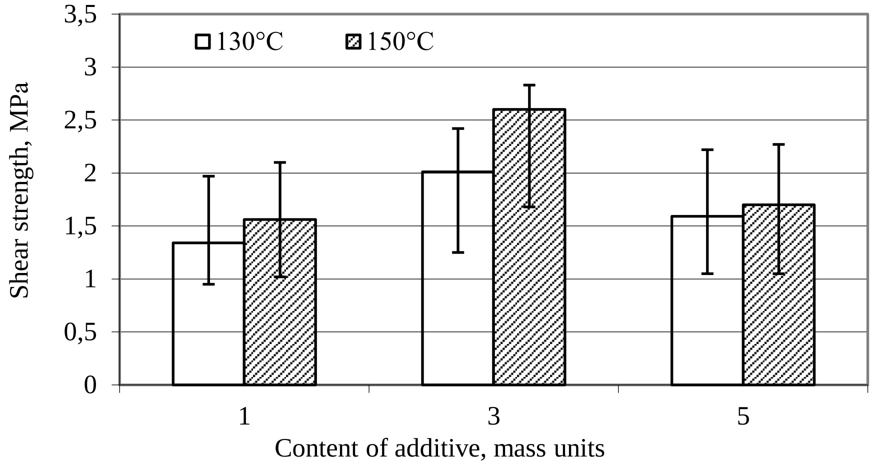

Fig. 1 Effect of SPI addition in PF resin and gluing temperature on shear strength of plywood.

The shear strength of plywood specimens glued at pressing temperature of 150 °C were higher than those glued at pressing temperature of 130 °C. High pressing temperature presents better heating of veneers assembly and faster curing of adhesive. So, for faster curing there is required to overcome the inhibiting effect of the moisture during the first stage of the curing process (LI *et al*. 2014, CLARKE *et al*. 1990).

The specimens made from veneer at moisture content of 15% and unmodified PF resin were delaminated after the boil test.

Adding of soy protein isolate to phenol-formaldehyde resin in the amount of 3 mass units is recommended because this adhesive has the most stable properties for plywood manufacture from the veneer with high moisture content of 15% and fully meets the requirements of the test according to EN 314-2 for exterior use.

CONCLUSIONS

The modification of PF resins by soy proteins isolate 92% creates the possibility of gluing veneer with high moisture content of 15% in the laboratory scale. The influence of SPI content on the properties of adhesives was studied. It was established that with increasing of the SPI amount, the viscosity of adhesives increases, the gelation time and pot life decrease, the solid content changes slightly, the pH value remains unchanged. The obtained values of the shear strength of the plywood were higher than the requirement (1.0 MPa) according to the standard EN 314-2 for exterior bonding quality.

Based on the obtained results, PF resin modified by soy protein isolates can be recommended for gluing veneer with 15% moisture content by hot-pressing (pressing temperature 130 or 150 °C). These positive results from the laboratory experiments give the base to plan experiments in industrial conditions. However, the application of some of them is limited due to their higher viscosity, mainly from the view of glue spread control.

REFERENCES

BEKHTA P. 2003.Vyrobnytstvo fanery. Kyiv : Osnova, 2003. c. 320.

BEKHTA P., BITS G., SEDLIAČIK J. 2008. Patent № 37808 Method of plywood fabrication from the veneer with high moisture content. National University of Forestry &Wood Technology of Ukraine,

Lviv, Ukraine, 2008. p.5.
BEKHTA P., BITS G., SEDLIAČIK J. 2008. Patent № 37809 Method of plywood fabrication from the veneer with high moisture content. National University of Forestry & Wood Technology of Ukraine, Lviv, Ukraine, 2008. p.5.
BEKHTA P., ORTYNSKA G., SEDLIAČIK J. 2011. Conditions of gluing high moisture content veneer. In Adhesives in Woodworking Industry: Proceedings of XX Symposium, Slovakia, 2011, p. 28–34.
BEKHTA P., ORTYNSKA G., SEDLIAČIK J. 2014. Properties of modified phenol-formaldehyde adhesive for plywood panels manufactured from high moisture content veneer. In Drvna Industrija, 2014, 65(4): 293–301.
BEŠINOVÁ O., KATUŠČÁK S., HESPODÁRIK A., TOKÁROVÁ L., LIPKA R. 1997. Advantages and benefits of gluing of veneers with high moisture content for plywood producers. In Adhesives in Woodworking Industry: Proceedings of XIII Symposium, Slovakia, 1997, p. 194–204.
CLARK R.J., KARCHESY J.J., KRAHMER R.L. 1988. Investigation of a new resin as an exterior adhesive to bond high moisture content veneers and glulam. In Forest Products Journal, 38: 71–75.
CLARKE M. E., STEINER P.R., ANDERSON A.W. 1990. Phenol-formaldehyde adhesive for bonding wood pieces of high moisture content and composite board and veneers bonded with such adhesive. U.S. Patent No. 4,897,314. U.S. Patent Off., Washington, D.C., 1990, p. 10.
CLAY J.D., VIJAYENDRAN B., MOON J. 1999. Rheological Study of Soy Protein-Based PRF Wood Adhesives. Annual Technical Conference Proceedings, Society of Plastics Engineers, Brookfield, CT, 1999, p. 1298–1301.
HAJDÚCHOVÁ I., SEDLIAČIKOVÁ M., HALAJ D., KRIŠTOFÍK P., MUSA H., VISZLAI I. 2016. Slovakian forest-based sector in the context of globalization. In BioResources, 11(2): 4808–4820.
JÓŹWIAK M. 2007. Possibility of gluing veneers with high moisture content with use modified MUF adhesives resin. Ann. Warsaw Agricult. Univ. SGGW, For. and Wood Technol. 2007, 61: 301–305.
ELBEZ G. 1997. Possibility of gluing of veneers with high moisture content. In Adhesives in woodworking industry: Proceedings of XIII Symposium, Slovakia, Vinne, 1997, p. 101–110.
GHAHRI S., PIZZI A., MOHEBBY B., MIRSHOKRAIE A., MANSOURI H.R. 2016. Soy-based, tannin-modified plywood adhesives. In Journal of Adhesion, Article in Press, pp. 1–20.
KUMAR R., CHOUDHARY V., SAROJ MISHRA S., VARMA I.K. 2002. Adhesives and plastics based on soy protein products. In Industrial Crops and Products, 2002, 16: 155–172.
LI H., LI C., CHEN H., ZHANG D., ZHANG S., LI J., 2014. Effects of hot-pressing parameters on shear strength of plywood bonded with modified soy protein adhesives. In BioResources, 9(4): 5858–5870.
MAHÚT J., RÉH R., VÍGLASKÝ J. 2006. Kompozitne drevne materialy. Cast I. Dyhy a preglejovane vyrobky. Zvolen : TU vo Zvolene, 2006. 292 s.
ORTYNSKA G., LYUTYY P. 2013. The possibility of using veneer with high moisture content in plywood production, KhNTUS, Kharkiv, 2013, 143: 26–31.
PIZZI A. 2006. Recent developments in eco-efficient biobased adhesives for wood bonding: opportunities and issues. In Journal of Adhesion Science Technology, 2006, 20(8): 829–846.
SEDLIAČIKOVÁ M., ŠATANOVÁ A., ZÁVADSKÝ J., ZÁVADSKÁ Z. 2015. Quality cost monitoring models in practice of woodworking company in Slovakia. In Procedia – Economics and Finance. 2015, 26: 77–81.
SU J. F., YUAN X. Y., HUANG Z., WANG X. Y., LU X. Z., ZHANG L. D., WANG S. B. 2012. Physicochemical properties of soy protein isolate/carboxymethyl cellulose blend films crosslinked by Maillard reactions: Color, transparency and heat-sealing ability. Materials Science and Engineering, 2012, 32: 40–46.
SUN X., BIAN K. 1999. Shear strength and water resistance of modified soy protein adhesives. In JAOCS, 1999, 76(8): 977–980.
ŠATANOVÁ A., ZÁVADSKÝ J., SEDLIAČIKOVÁ M., POTKÁNY M., ZÁVADSKÁ Z., HOLÍKOVÁ M. 2015. How Slovak small and medium manufacturing enterprises maintain quality costs: an empirical study and proposal for a suitable model. In Total Quality Management & Business Excellence. 2015, 26(11–12): 1146–1160.
URBANIK E., JABLOŃSKI W., JÓŹWIAK M. 1997. Gluing high moisture content veneer with adhesives based on RPF and PF. In Adhesives in woodworking industry: Proceedings of XIII Symposium, Slovakia, Vinne, 1997, p. 225–232.

VIJAYENDRAN B., CLAY J. 2000. Some recent studies on soy protein-based wood adhesives. In Wood Adhesives 2000 Extend Abstracts. Nevada : Forest Products Society, 2000 p. 4–5.
WU Z., DU G., LEI H., WANG H., XI X., CAO M., LIAO J. 2016. Modification of soy protein-based adhesives by a new phenol formaldehyde resin. In Chemistry and Industry of Forest Products, 36(1): 119–126.
YANG I., KUO M., MYERS D.J. 2006. Bond quality of soy-based phenolic adhesives in southern pine plywood. In JAOCS Journal of the American Oil Chemists' Society, 83(3): 231–237.
EN 314-2: 1998 Plywood. Bonding quality. Part 2: Requirements.
EN 827: 2005 Adhesives. Determination of conventional solids content and constant mass solids content.

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