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LIGNOCELLULOSIC WASTE OF FURFURAL PRODUCTION FROM BAGASSE AS NON-FOOD FILLER AND SUBSTITUTE FOR UF RESIN IN PLYWOOD MANUFACTURE

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ABSTRACT

The effect of lignocellulosic waste from furfural production of bagasse (LWFPB) as non-food filler for urea-formaldehyde resin (UF) and also substitute a part of UF resin in plywood manufacture was evaluated. LWFPB was used at four levels of 0, 10, 20 and 30% as filler and was replaced with UF resin at three levels of 0, 10 and 20%. Then the physical and mechanical properties of the plywood samples were measured. Results showed that the mechanical and physical properties of plywood were increased compared to the control (with wheat flour filler) when UF resin was used with LWFPB filler. Higher shear strength, MOR and MOE were associated with the use of 30% LWFPB. Besides that, the addition of LWFPB instead of UF resin reduced the mechanical and physical properties of plywood to some extent, but compared to control sample, the best results were obtained with the addition of 10% LWFPB.

KEYWORDS: Plywood, furfural, filler, lignocellulosic waste.

INTRODUCTION

Adhesives play a critical role in the production of wood-based materials and the quality of the connection, and therefore the characteristics of wood-based materials are often determined based on the type and quality of the adhesive (Carvalho et al. 2014). Currently, adhesives derived from fossil sources including urea formaldehyde, phenol formaldehyde and isocyanates, are mainly used for the production of wood-based materials, such as particleboard, plywood and MDF composites (Jang et al. 2011, Zhao and Umemura 2014). Plywood is a valuable wood panel that is widely used as a finishing and building material and has very favourable mechanical properties due to its layered structure (Bekhta et al. 2016). The most commonly used adhesives in plywood industry are urea-formaldehyde (UF) resin due to its colourless, fast curing, water solubility, and inexpensive (Jeong et al. 2019, Jovanovic et al. 2019). Despite many advantages, UF adhesives also have disadvantages, including the release of formaldehyde from manufactured wood-based materials and low water resistance (Gao et al. 2018, Kawalerczyk et al. 2019). Formaldehyde is a poisonous and dangerous compound that can cause dangerous diseases in humans such as cancer (Lin et al. 2012). In recent years, research and development on the use and application of natural adhesives in the manufacture of wood composites has become necessary due to scarcity of fossil resources (Gui et al. 2013) and environmental problems caused by the use of formaldehyde adhesives. Another disadvantage of UF resins is their low heat and humidity resistance, which limits its use in different conditions. As a result, many studies have been carried out to improve the performance of these resins, for example by chemical modifications or adding different fillers. One way to improve the parameters of UF resin is to add different fillers (Pawlak and Boruszewski 2018). Fillers are important components of UF resins, which are mostly added to adhesive mixtures in solid state (Kawalerczyk et al. 2019, Bekhta et al. 2021) in order to reduce production costs, limit unwanted adhesive flow or excessive penetration into veneers (Irle et al. 2012, Cao et al. 2020). Fillers improves the rheological properties of the adhesive mixture and regulates the viscosity or limiting the appearance of micro cracks in the joints (Kawalerczyk et al. 2019).

In the manufacture of plywood composites, 20-30% wheat flour (WF) is added to the UF resins as a filler, which increases their viscosity. With increased viscosity, the bond strength increases, this affects the fluidity of the UF resin and prevents resin penetration into wood veneers (Hong and Park 2017). Low viscosity adhesives penetrate into the surface of veneer during the application. It is difficult to spread the low-viscosity glue evenly on the surface of the veneer (Kawalerczyk et al. 2019). Flour and starch are fillers used to control the viscosity and rheological behaviour of the UF resin, which helps to increase the molecular weight and control the viscosity and rheological behaviour of wood glues, and also to prevent excessive flow and penetration of the resin into the wood (Aydin et al. 2017). To achieve good adhesion, the low molecular weight fraction of the resin penetrates through the wood cell walls, while the high molecular weight fraction remains at the bond line between the two woods (Nuryawan et al. 2014). Wheat flour particles are also broken by moisture and heat, and when adjusted, they create colloidal and gelatinous conditions that can also act as reinforcements and improve adhesion. But both flour and starch are food sources whose consumption in such industries causes food shortages (Vnucec et al. 2016). Approximately 2 million tons of wheat flour are used annually in plywood production (Li et al. 2019), therefore, the development of resins based on non-edible fillers is one of the new research objectives.

Use of wastes or organic substances can be a suitable alternative as a raw material in addition to reducing the production costs (Réh et al. 2021, Mirski et al. 2020, Bekhta et al. 2021, Aydin et al. 2017, Ružiak et al. 2017). As reported by Karimi et al. the use of 15% powder derived from alkali sulfite-anthraquinone (AS-AQ) black liquor instead of wheat flour as filler for urea formaldehyde resin does not adversely affect shear strength of plywood.

This publication is focused on filler obtained from waste from furfural production process of bagasse. Furfural is produced from agricultural or industrial wastes (corn, rice and oats, etc.)

by acid hydrolysis of polymeric pentoses fallowed by acid dehydration of aldopentoses (McKillip and Sherman 1980). Furfural is the organic compound that can replace the petroleum-based organics used in industry. The main source of commercial furfural is bagasse and agricultural wastes. The annual production of furfural waste is estimated at approximately 23 million tons (Sun et al. 2008). Some researchers consider the recycling of millions of tons of furfural waste justified for environmental and economic reasons (Dai et al. 2010). Thus, due to the abundant production of furfural from agricultural waste, a lot of waste is generated, while the residue (mainly cellulose and lignin) is usually burned (Sun et al. 2014). At the same time, these natural waste materials can be recycled in industry. Therefore, the purpose of this study is to investigate the effect of LWFPB instead of wheat flour as non-food filler in urea-formaldehyde resin and also as a part of UF resin in the production process of three-layer plywood composite and determine its effects on physical and mechanical properties.

MATERIAL AND METHODS

Materials

The adhesive used in this study is a commercial UF resin purchased from Samed Company whose properties are listed in Tab. 1. In the first step, the lignocellulosic residue from industrial furfural production from bagasse (LWFPB) was used as filler for UF resin at four levels of 0 (control sample), 10, 20 and 30% based on the dry weight of the resin (when the amount of LWFPB was 10 and 20%, wheat flour was used with up to 30% difference).

In the second step, LWFPB was also used as part of the urea-formaldehyde resin at 0 (control sample), 10, and 20% levels, and after calculating its amount, it was reduced by the urea-formaldehyde resin. Plywood bonded with UF resin filled with 30% wheat flour was used as control sample. Ammonium chloride (NH_4Cl) powder was also mixed as a hardener at a level of 2% based on the dry weight of UF resin.

Tab. 1: Specifications of urea formaldehyde resin.

Adhesive	Producer	Solids (%)	pH	Gel time (s)	Viscosity at 20℃ (s)	Density (g/cm^3)
Urea formaldehyde	Samed Co.	63.4	7.5	56	46	1.283

Plywood manufacture and physical and mechanical properties testing

Poplar veneers with dimensions of 400 mm × 400 mm × 2 mm and moisture content of 8% without visible defects were prepared for the production of three-layer plywood composite under laboratory conditions. The resin was applied to both sides of the veneers at a rate of 120 g/m^2. The resin-coated veneers were then stacked between two uncoated veneers so that the grain direction of the two adjacent veneers was perpendicular to each other. The stacked plywood was hot pressed at 1.0 MPa and 160℃ for 5 min. Three sheets of plywood composite were prepared for each group. Then the sheets were conditioned in a climate-controlled room for 15 days before physical and mechanical evaluation. After this period, plywood properties were determined as follow: thickness swelling after 2 h and 24 h soaking in water, bending strength (MOR) and modulus of elasticity (MOE) parallel and perpendicular to the surface grain according to EN 310 (1993) and shear strength according to EN 314-1 (Tab. 2).

Tab. 2: Dimensions and number of test samples per treatment.

Test type	Dimensions (mm)	Number of repetitions	Total
Thickness swelling	50×50×6	6	18
Bending strength parallel to fibers	170×50×6	3	9
Bending strength perpendicular to fibers	170×50×6	3	9
Shear strength	120×25×6	4	12

Statistical analysis

Statistical analysis was performed using a statistical software at a significance level of $\alpha = 0.05$ and significant differences between groups were determined by Duncan Multiple Comparison.

RESULTS AND DISCUSSION

Shear strength

One of the most important mechanical properties to evaluate the bonding quality of plywood composite joints is shear strength. Fig. 1 presents the results of bonding quality test data performed according to EN 314-1 (2004) on dry sample. As can be seen in Fig. 1, different levels of LWFPB powder added as filler and also as a substitute for UF resin affected the shear strength of plywood. Statistical analysis shows that this effect is not significant and all levels belong to the same group (a). The studies showed that plywood made with UF resin with LWFPB as filler achieved better shear strength results than the control (Fig. 1a). However, regardless of the amount of filler used, the bonding quality of all plywood reached values better than 1.0 N/mm^2, thus meeting the requirements of EN 314-2 (1993). The addition of LWFPB to UF resin up to 30% positively affected the bonding quality and increased the shear strength which improved 21.8% compared to the control sample. This means that the lowest shear strength is related to the control sample (1.74 MPa), without LWFPB, which is usually produced with UF resin and wheat flour filler. The highest shear strength is associated with plywood made with UF resin and 30% LWFPB powder filler (2.12 MPa). The consumption of 10% and 20% LWFPB powder increased by 13.79% and 6.89% compared to the control, which can be a positive result when using LWFPB as a waste, natural and renewable material instead of wheat flour. Hedjazi et al. (2018) used and compared lignin derived from three types of pulping processes, including alkaline sulfite-AQ, soda-AQ and Kraft, as a filler for urea-formaldehyde resin and as a substitute for wheat flour in plywood production. They indicated that the use of lignin from all three pulping processes as filler for UF resin had a positive effect on bonding performance and shear strength of the boards. Moreover, studies performed by Dukarska et al. (2022) confirmed that plywood composites containing 1 pbw of nanoclay and 7.3 pbw of the tannin as filler were characterized by improved bonding quality. Walkiewicz et al. (2023) who also recorded the addition of tea leaves as filler for UF resin in plywood production reported a reduction in formaldehyde emission from plywood compared to the reference sample. Whereas, the bonding quality test showed that this filler did not affect the strength properties of the tested bond lines.

The evaluation of the average strengths also shows that all levels related to the consumption of LWFPB powder are in the same homogeneous group (a) (Fig. 1b). The differences in strength are not statistically significant. Nevertheless, when the consumption of LWFPB increased to 10%, the shear strength was higher than the control sample. In other words, by replacing 10% of UF resin with LWFPB powder, the shear strength increased by 13.79%. However, even when replacing 20% UF resin with LWFPB, the shear strength decreases and is lower than that of the control sample. Guthwal et al. (2010) in their study on the synthesis of low-cost adhesives made from pulp industry waste, noted that the shear test properties of plywood obtained by replacing phenol (15%) with black liquor lignin had comparable physical bond properties and can be used to form an inexpensive modified PF resin for plywood sheets. Nasrinobandegani et al. (2017) used a phenol-formaldehyde-fish glue hybrid in the production of plywood and found that the addition of 20% fish glue with phenol formaldehyde resin improved the shear strength of the bonds. As well as, the addition of lignin to adhesive formulations, can improve the mechanical properties of phenolic adhesives (Belgacem and Gandini 2008).

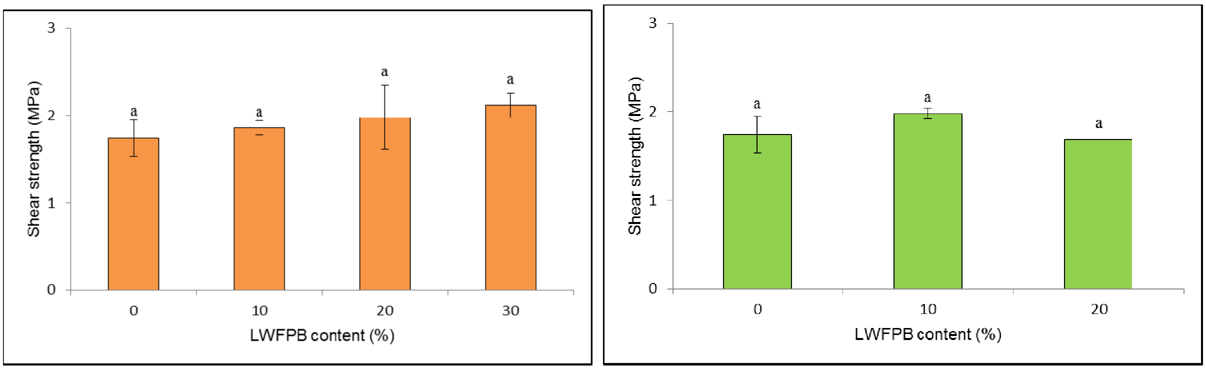

Fig. 1: Shear strength of plywood: a) according to the content of LWFPB as filler of UF resin, b) made by replacing UF resin with LWFPB.

Bending strength

The mechanical property such as bending quality (MOR) is presented in Figs. 2 and 3. The improvement of bending strength for both parallel and perpendicular to the surface grain, caused by adding different amounts of LWFPB as filler to the UF resin was found compared to the control sample. Indeed, when the LWFPB was increased up to 20%, the bending strength parallel to the surface grains was greater than the control and with an increase of 30%, the strength was lower than the control value (Fig. 2a). The bending strength perpendicular to the surface grain is greater than the control value for all LWFPB filler amounts (Fig. 2b). Statistical analysis showed that different levels of LWFPB powder as a filler in UF resin had no significant effect on the bending strength of plywood parallel and perpendicular to the surface grain, and all levels belonged to the same group (a). According to a study on replacing flour with beech bark as filler, the mechanical properties such as MOR and MOE increased (Ružiak et al. 2017).

But on the other hand, the results showed that the different amounts of LWFPB powder as a partial replacement of the UF resin had a significant effect on the bending strength parallel to the surface grains (Fig. 3a). Increasing the LWFPB consumption to 10% does not have a significant negative effect on the bending strength parallel to the surface grain. However, increasing it to 20%, its resistance is significantly lower than the value of the control sample. While by adding LWFPB as a partial substitute for UF resin, the bending strength perpendicular to the surface grain is not significant, and the values of all treatments were higher than the control value (Fig. 3b).

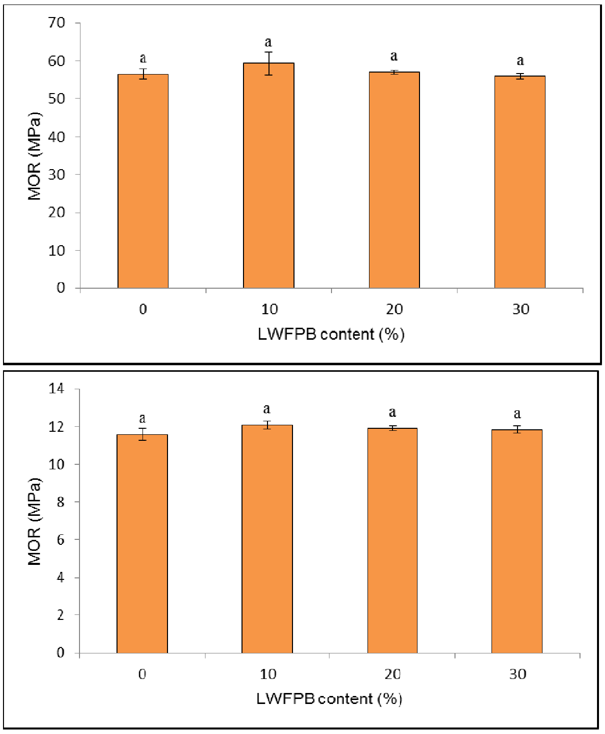

Fig. 2: Bending strength (MOR) of plywood parallel (a), and perpendicular (b) to the surface according to the content of LWFPB as filler of UF resin.

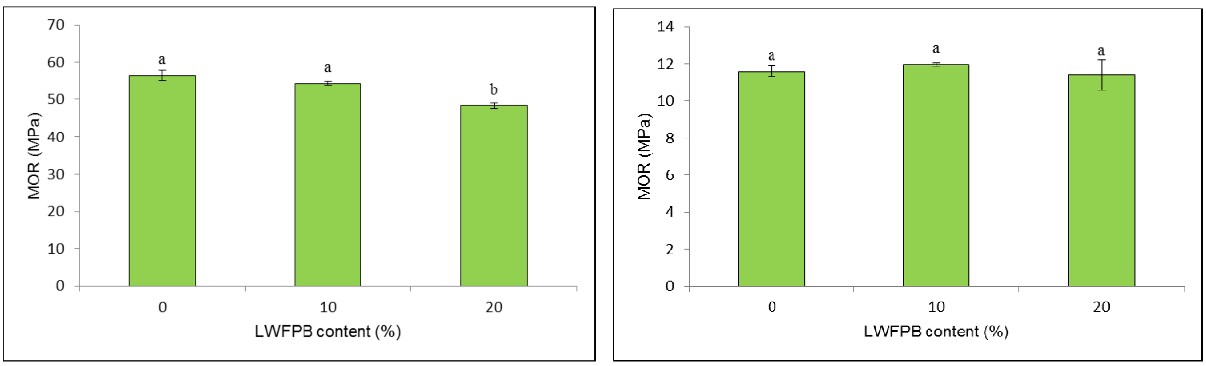

Fig. 3: Bending strength (MOR) of plywood parallel (a) and perpendicular (b) to the surface grain made by replacing UF resin with LWFPB.

Modulus of elasticity

The modulus elasticity (MOE) of plywood composite made by UF resin with different levels of LWFPB added as filler is shown in Fig. 4. Based on the data presented, it was found that as the amount of LWFPB increases, the modulus elasticity of boards parallel and perpendicular to the surface grain increases. The statistical analysis shows that different levels of LWFPB had a significant effect on the modulus elasticity parallel and perpendicular to the surface grain and Duncan revealed different levels of LWFPB in different groups. All MOE values of the plywood made with LWFPB as filler in UF resin were higher than the control sample (Figs. 4a and 4b).

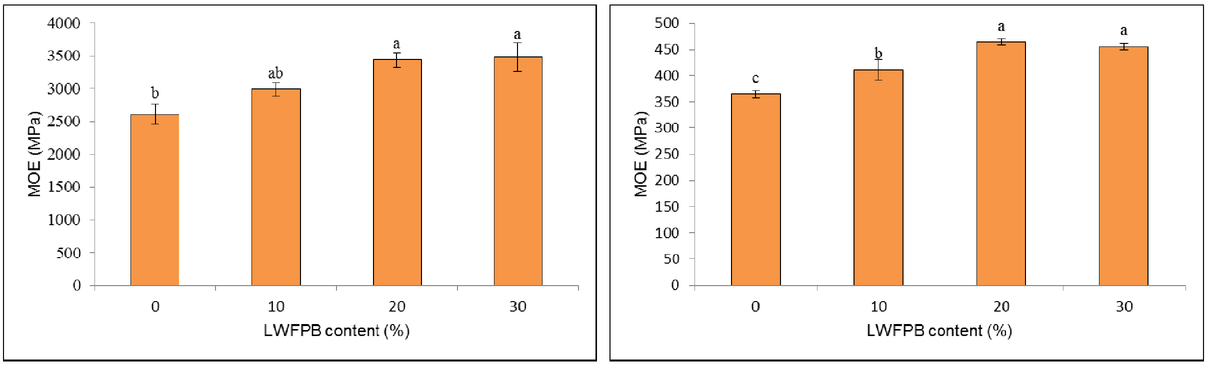

Fig. 4: Modulus elasticity (MOE) of plywood parallel (a) and perpendicular (b) to the surface grain according to the content of LWFPB as filler of UF resin.

However, the addition of LWFPB to replace a part of the UF resin up to 20% resulted in a slight decrease in MOE (Fig. 5). Duncan showed different levels of LWFPB in different groups (a, b). However, according to Figs. 5a,b, it is clear that when 10% of UF resin was replaced by LWFPB, the decrease in MOE was not significant, and both are in the same group. This means that 10% of the UF resin can be replaced by LWFPB without a significant reduction in MOE. But further increase in LWFPB amount caused the MOE values to decrease. In fact, when the amount of UF resin was replaced by 20% LWFPB, the MOE decreased significantly both parallel and perpendicular.

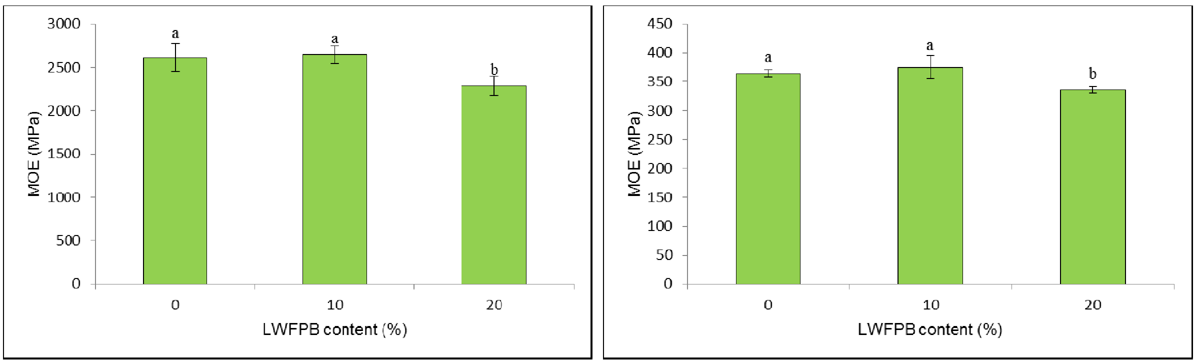

Fig. 5: Modulus elasticity (MOE) of plywood parallel (a) and perpendicular (b) to the surface grain made by replacing UF resin with LWFPB.

Thickness swelling

The results obtained for thickness swelling of plywood panels after 2 and 24 h bonded with UF resin and three levels of LWFPB as adhesive filler are presented in Fig. 6a. The results demonstrated that all levels related to the consumption of LWFPB powder are in the same homogeneous group (a) and the differences are not statistically significant. But as can be seen a decreasing trend in TS value of the produced plywood occurred when the amount of LWFPB as filler increased up to 20% compared to the control sample. In this way, the highest amount of TS is associated with the control sample. Furthermore, as can be seen in Fig. 6a, applying further this filler (LWFPB) at the level of 30 % caused an increase of TS value after 2 and 24 h soaking in water, but it is still lower than the TS value of the control sample. The highest thickness swelling is associated to the control sample made with UF resin and wheat flour (WF) as filler. The WF absorbs most of the water, which participates in forming a three-dimensional crosslinking network structure (Mirski et al. 2020). The lowest TS after 2 and 24 h soaking in water are related to 20% LWFPB, which are 22.54% and 14.55% reduction compared to the control sample. LWFPB has been reported to contain lignin (Sun et al. 2014). Lignin can react with formaldehyde in an acidic environment; the benzyl alcohols formed react with the lignin model compound, resulting in the formation of methylene-linked dimers (Van Der Klashorst and Strauss 1986). This problem can be effective in reducing the number of functional groups and reducing water absorption, which then reduces thickness swelling. Based on other studies, the addition of lignin powder derived from Kraft black liquor (jamalirad et al. 2007) and alkali sulfit-anthraquinone (AS-AQ) black liquor powder (Karimi et al. 2018) as a filler for urea-formaldehyde resin improved the physical properties of plywood.

The average TS value of samples made with UF resin (control) and the replacement of LWFPB with UF adhesive is shown in Fig. 6b. The results showed that at different levels, replacing LWFPB with UF resin did not significantly affect the thickness swelling after 2 h soaking in water, and all treatments were in the same homogeneous group (a). But the TS value after 24 h soaking in water has an increasing trend. The lowest thickness swelling after 2 and 24 h soaking in water is related to the control sample and the highest is related to 20%.

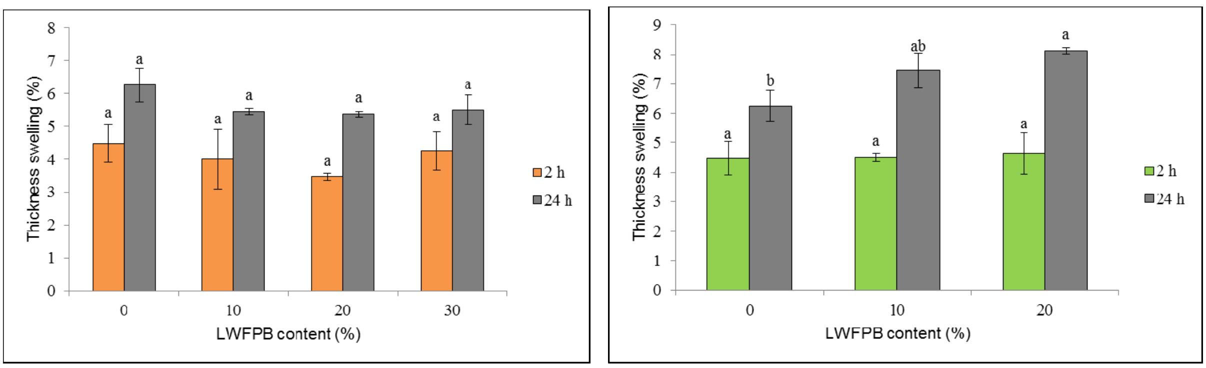

Fig. 6: Thickness swelling (TS) of plywood: a) according to the content of LWFPB as filler of UF resin, b) made by replacing UF resin with LWFPB.

CONCLUSIONS

The results of this study showed that when UF resin was used with LWFPB filler in plywood composite, the desired mechanical and physical properties were achieved compared to the control (with wheat flour filler) and the highest shear strength at the bonding surface, MOR and MOE was associated with the use of 30% LWFPB powder. Moreover, the presented results show that the addition of LWFPB instead of UF resin slightly weakened the mechanical and physical properties of the plywood, but certainly the best results compared to the control sample were obtained with the addition of 10% LWFPB.

Generally, the results showed that the use of LWFPB as a filler in UF resin instead of wheat flour in the plywood manufacturing process had good results. Since wheat flour is an important food source worldwide and its increasing shortage is also felt, finding a suitable substitute for wheat flour in this industry is important and inevitable. Whereas, LWFPB is a natural and renewable waste material that is produced from furfural production process from bagasse. Also, since chemical resins such as UF have negative and biodegradable effects, replacing even a small amount of UF resin and using disposable and biodegradable materials can be a positive step to reduce the negative effects of using this type of chemical adhesives.

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