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Surface characteristics and overlaying properties of flat-pressed wood plastic composites

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Abstract This study evaluated surface characteristics and overlaying properties of wood plastic composite (WPC) panels made from dry-blended rubber wood fiber-polypropylene (PP) powder formulations using a conventional flat-press process under laboratory conditions. Three levels of rubberwood fibers (Hevea brasiliensis), 40%, 50%, and 60%, based on the composition by weight, were mixed with the PP powder without and with 3% (based on weight) maleic anhydride grafted PP (MAPP) as a coupling agent. Contact angle measurements on the WPC samples were performed using a goniometer connected with a digital camera which collected one image per second. Roughness measurements, average roughness (R_a) , mean peak-to-valley height (R_z) , and maximum roughness (R_y) , were taken from the sanded samples along and across the sandmarks using a fine stylus tracing technique. With increasing polymer content, surface roughness of the WPC panels improved while their wettability and adhesive bonding strength decreased. The formulations without MAPP were found to have higher surface roughness but they also had better wettability. Wettability and surface roughness of the WPC panels can provide good information on their ability to bond.

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Oberflächen- und Beschichtungseigenschaften von im Flachpressverfahren hergestellten Holz-Polymer-Werkstoffen

Zusammenfassung In dieser Studie wurden die Oberflächen- und Beschichtungseigenschaften von Holz-Polymer-Werkstoffplatten (WPC) untersucht, die aus im Trockenverfahren hergestellten Mischungen aus Gummibaum-Holzfasern (Hevea brasiliensis) und Polypropylen (PP) im Flachpressverfahren unter Laborbedingungen hergestellt worden waren. Drei verschiedene Holzmehlgehalte, 40%, 50% und 60% (Masseprozent), wurden mit PP-Pulver gemischt, und zwar sowohl ohne als auch mit 3% (Masseprozent) mit Maleinsäureanhydrid gepfropftem PP (MAPP) als Haftvermittler. Die Kontaktwinkel auf den WPC-Proben wurden mittels eines Goniometers gemessen, der an eine Digitalkamera angeschlossen war, die eine Aufnahme pro Sekunde machte. Rauheitsmessungen, wurden durchgeführt. Dabei wurden die durchschnittliche Rauheit (R_a), die mittlere Rautiefe (R_z) und die maximale Rauheit (R_v) an den geschliffenen Prüfkörpern in und quer zur Schleifrichtung mittels des Tastschnittverfahrens bestimmt. Mit zunehmendem Polymergehalt nahm die Oberflächenrauheit der WPC-Platten ab, während ihre Benetzbarkeit und die Klebefestigkeit abnahmen. Die Mischungen ohne MAPP wiesen eine größere Oberflächenrauheit, aber auch eine bessere Benetzbarkeit auf. Die Benetzbarkeit und die Oberflächenrauheit von WPC-Platten können zur Beurteilung von deren Verklebungsverhalten herangezogen werden.

1 Introduction

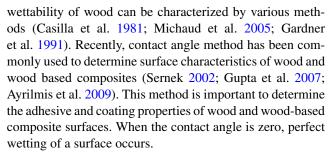
Wood plastic composites (WPCs) are a relatively new class of materials and one of the fastest growing sectors in the



wood composites industry. WPC consists of a mixture of wood, thermoplastics and some additives. Typically, the wood content of WPCs ranges between 50 and 80% wt. (Clemons 2002). Although commercially less important, the predominant technologies to produce WPCs are extrusion to obtain endless profiles and injection moulding leading to 3-dimensional forms. Another possibility which has only little been explored is to produce WPCs on a flat-press. The advantage of this technology is that only a relatively low pressure level is required, compared to extrusion and injection moulding. As a consequence, the naturally given wood structure is maintained, resulting in a considerably reduced material density. As the productivity of the pressing technology is much higher than that of extrusion and injection moulding, flat-pressed WPCs made by a dry-blending method have a clear cost advantage. WPC panels may be used for applications that require high moisture resistance, and that shall be easy to maintain. The use of conventional wood-based composites, such as particleboard and fiberboards, is quite limited for exterior and moist applications, due to the strong tendency of such materials to absorb water. By contrast, WPCs show a considerably reduced affinity towards water, compared to conventional wood-based composites such as particleboard and fiberboard, which is caused by their relatively high thermoplastic content.

When the panels are used as substrate for thin overlays their surface characteristics in terms of roughness play an important role in determining quality of final product. There are various methods to evaluate surface roughness of composite panels, including acoustic emission, pneumatic, laser, and stylus (Hiziroglu et al. 2004). Standard contact measuring devices employing a stylus tracer, such as used in the metal and plastic industry were successfully employed to evaluate roughness characteristics of various wood composites (Hiziroglu 1996). One of the main advantages of the stylus method is to have an actual profile of the surface and standard numerical roughness parameters which can be calculated from the profile. Any kind of irregularities and magnitude of show-trough on the overlaid substrate can be objectively quantified. Therefore, it is important to quantify surface roughness of the panel to have a better overlaying of the substrate. In this study, roughness properties of the WPC panels were evaluated using a stylus method to have a better understanding of the overlaying quality of the samples.

Wettability is defined as a condition of a surface that determines how fast a liquid will wet and spread on the surface or whether it will be repelled and not spread on the surface. Wettability is crucial for good adhesion in wood bonding. The adhesive has to wet, flow, and penetrate the cellular structure of wood in order to establish intimate contact between molecules of wood and adhesive. There is evidence about the positive relationship between wood wettability and adhesion (Ayrilmis and Winandy 2009). The



Overlaying of the panels with veneer sheets improves their appearance and properties resulting in value-added products. Currently, interior fitment and furniture manufacturers using wood based panels such as particleboard and medium density fiberboard (MDF) overlaid with decorative surfacing material do not commonly know the flat-pressed WPC panels. However, when the WPC panels are overlaid with decorative wood veneer sheets using a suitable adhesive, they can be a competitor to overlaid wood based panels in office furniture manufacture. Overlaid panels can also be used in the construction of cabinets, paneling, kitchen worktops, and work surfaces in offices, educational establishments, laboratories, and other industrial product applications.

As composites are used in more applications and capture additional market shares, the need to better understand the material's properties as they relate to secondary machining becomes more important. This study was conducted to obtain first indications of secondary processing properties of flat-pressed WPC panels made by a dry-blending method. An extensive literature search did not reveal any information about surface characteristics and overlaying properties of the flat-pressed WPC panels. For this aim, surface roughness, wettability, and adhesive bonding strength of the flat-pressed WPC panels made from various mixtures of the wood fiber and polypropylene (PP) powder were investigated, using a method currently used in the wood based panel industry. In addition, effect of the compatibilizer on the above mentioned properties of the WPC panels was evaluated. Table 1 shows the raw material formulations used for the WPC panels. The values chosen for the wood fiber/plastic content are within a range most commonly employed in the manufacture of wood plastic composites (Clemons 2002).

2 Materials and methods

2.1 Materials

Rubberwood fibers (*Hevea brasiliensis*) having average 1.5 mm length, were obtained from a commercial MDF plant in Thailand. Rubberwood is composed of fibers (58%), vessel elements (8.5%), axial parenchyma (11.5%), and rays (22%) and are distributed in different patterns and proportions as in other typical hardwood species. The fibers are



Table 1 Compositions of the evaluated WPC formulations

Tab. 1 Zusammensetzung der untersuchten WPC-Mischungen

WPC panel	WPC panel composition (by % weight)						
type	Wood fiber	Polypropylene	Maleic anhydride-grafted polypropylene				
A	40	60	_				
В	50	50	_				
C	60	40	_				
D	40	57	3				
E	50	47	3				
F	60	37	3				

non-septate, and belong to the medium group with a length of 0.8-1.8 mm. The width of the fibers ranges between 19-27 µm (Mathew 2004). The fibers were produced using a thermo-mechanical refining process without any chemical and resin. The moisture content of the fibers, as determined by oven-dry weight, was found to be 2-3% prior to treatment.

Polypropylene (PP) ($T_{\rm m}=160^{\circ}{\rm C}$, $\rho=0.9~{\rm g/cm^3}$, MFI/230°C/2.16 kg = 6.5 g/10 min) produced by *Petkim Petrochemical* Co., Turkey, was used as a polymeric material. Maleic anhydride-grafted PP (MAPP, OPTIM-415[®]; the reactive modifier maleic anhydride (MAH) content = 1% wt.) as a coupling agent was supplied by *Pluss Polymers* Pvt. Ltd. in India. The PP granules were then processed by a rotary grinder to pass through a US 40-mesh screen and retained on a US 80-mesh screen. The PP powder was then dried in a laboratory oven at 100°C for 24 hours to moisture content of 0–1% based on oven-dry PP weight.

2.2 Flat-pressed WPC panel manufacture

Flat-pressed WPC panels were manufactured using standardized procedures that simulated industrial production at the laboratory. After mixing wood fibers and the PP powder and placing the mixture into a rotary drum blender, the mixture was weighed and then formed into a mat on an aluminum caul plate, using a forming box. Wax paper was used to avoid direct contact of PP powder with the metal platens during heating and pressing. To reduce the mat height and to densify the mat, the mat was cold pressed. This procedure allowed for easy insertion of the mats into the hot-press. The mats were then subjected to hot-pressing, using a manually controlled electrical-heated press. The maximum press pressure, pressing temperature, and total press cycle were 3.5 N/mm², 170°C, and 6 min, respectively. Temperature was set to ensure that it is slightly above the melting point of the plastic component. At the end of the press cycle, the board was removed from the press for cooling. The nominal panel size was 250 mm \times 250 mm \times 10 mm after the cooling process. A total of 12 experimental panels, two for each type of panel, were manufactured. The density values of the WPC panels were 0.79 to 0.80 g/cm³.

2.3 Determination of the surface roughness

Surface roughness test samples with dimensions of 50 mm \times 50 mm \times 10 mm were conditioned in a climate chamber at 20°C and 65% relative humidity (RH). Ten samples were used from each type of the panel for surface roughness measurements. The samples were sanded with a sequence of 100- and 150-grit sand papers. A total of fourty roughness measurements (four from each of ten samples: two measurements parallel to the sand marks and two measurements perpendicular to the sand marks from each of the samples) were taken from each type of formulation.

A Mitutoyo SJ-301 surface roughness tester, stylus type profilometer, was employed for the surface roughness tests. Three roughness parameters characterized by ISO 4287 standard (1997), respectively, average roughness (R_a), mean peak-to-valley height (R_z) , and maximum peak-to-valley height (R_v) were considered to evaluate the surface characteristics of the panels. The surface roughness parameters can be calculated from the digital information. The vertical displacement of the stylus is converted into electrical signals by linear displacement detector before the signal is amplified and converted into digital information. R_a is the arithmetic mean of the absolute values of the profile deviations from the mean line and is by far the most commonly used parameter in surface finish measurement. Specification of this parameter is described in previous studies (Hiziroglu 1996; Hiziroglu and Graham 1998; Mummery 1993). Roughness values were measured with a sensitivity of 0.5 μm. Measuring speed, pin diameter and pin top angle of the tool were 10 mm/min, $4 \mu \text{m}$, and 90° , respectively. The length of tracing line (L_t) was 12.5 mm and the cut-off was $\lambda = 2.5$ mm. Measuring force of the scanning arm on the samples was 4 mN (0.4 gf). Measurements were done at room temperature and pin was calibrated before the tests.



2.4 Determination of the wettability

The wetting behavior of WPC samples conditioned at 65% RH and 20°C was characterized by the contact angle method (goniometer technique). Contact angle measurements were performed using a CAM 101 Optical Contact Angle Meter (KSV Instruments Ltd., Helsinki), equipped with a video camera which collected one image per second. Using the sessile drop method, which is the most widely used procedure, the contact angle was determined simply by aligning a tangent with the sessile drop profile at the point of contact with the solid surface. An imaging system was used to measure contact angle and droplet shape for the tested surfaces of the WPC samples. The drop image was stored by a video camera and the image analysis system calculated the contact angle (θ) from the shape of the distilled water drop at room temperature. After the 5 µL droplet of distilled water was placed on the sample surface, contact angles from the images were measured at 1 sec time intervals up to 90 sec total. Ten samples with a size of 50 mm \times 50 mm \times 10 mm were taken from each type of WPC formulation for contact angle measurements. A total of twenty contact angle measurements, two from each of ten samples were performed for each type of formulation.

2.5 Overlaying of WPC samples with veneer sheet

The top and bottom surfaces of the sanded WPC samples conditioned at 65% RH at 20°C were then overlaid with 0.60 mm thick sliced beech (*Fagus orientalis* L.) veneer having dimensions of 50 mm \times 50 mm \times 10 mm. Ureaformaldehyde (UF) resin was spread on the surface of the WPC samples at the rate of 180 g/m² using a roller prior to curing using a Carver bench-top press at a temperature of 130°C and a pressure of 65 bar for 4 min. Ten replicate overlaid samples were made from each type of formulation.

2.6 Delamination strength between WPC surface and wood veneer sheet (adhesive bonding strength)

Adhesive bonding strength between WPC surface and veneer sheet (delamination test) was evaluated on the veneer faced WPC samples according to DIN 68765 B1 (1987). On the surface of the samples, a circle with a 35.7 mm diameter was drilled through the veneer thickness. This veneer circle on the sample surface was separated from the surrounding veneer. A metal tension seal (pull-up seal) was glued with polyurethane adhesive and placed in the movable crosshead of the universal test machine to remove the veneer circle from the panel surface. The force was applied at an even rate and the rate of application was adjusted so the time from the initial application of the force until failure of the test sample was not less than 30 s and not more than 120 s. One measurement from each of ten replicate overlaid samples was performed for the adhesive bond strength.

2.7 Statistical analysis

An analysis of variance, ANOVA, was conducted (p < 0.01) to evaluate the effect of polymer/wood ratio and coupling agent on all the properties measured namely surface roughness, wettability, and delamination tests. Significant differences between the mean values of the WPC groups were determined using Duncan's multiple range test.

3 Results and discussion

3.1 Surface roughness and wettability

Table 2 shows the R_a , R_y , and R_z values of the WPC samples without veneer sheet. The surface roughness values of the WPC samples decreased with increasing PP content. Statistical analysis found some significant differences among the WPC means for the R_a , R_y , and R_z values. The results of Duncan's multiple range test are shown by letters in Table 2. Panel type D had the smoothest surface with an R_a value of 5.52 µm while the roughest surface was found for the panel type C having an R_a value of 9.67 µm. This can clearly be observed by inspection of raw data from the surface roughness profilometer that recorded noticeably shallower ridges and valleys when compared to WPC panels containing higher wood fiber content as it traversed the WPC surface at a constant speed (Fig. 1). With the addition of wood fiber, surface roughness of the WPC panels significantly increased. This was mainly attributed to the anatomical structure of the wood fibers such as caves inside (vessels and cell lumens). Particle size and geometry are major factors which play significant role in surface roughness wood based composites (Akbulut et al. 2000). The width, height, and shape of the irregularities on a surface establish surface quality of a product (Ozdemir et al. 2009). The lower surface roughness of the WPCs having higher plastic powder can be explained by their melting in the hot press. The PP may crystallize on the wood fibers and thereby wrapping wood fibers better and leaving less exposed wood on the WPC surface. This results in lower surface roughness on the WPC surface.

The WPC formulations without MAPP were found to have higher surface roughness than those of the WPC formulations with MAPP (Table 2). Similar results were found in a previous study carried out by Ozdemir and Mengeloglu (2008). They found that injection moulded WPC samples made without MAPP were found to have higher surface roughness (R_a : 14.91 µm) than those with MAPP (4% wt.) (R_a : 8.28 µm). Gupta et al. (2007) reported that in the absence of coupling agent between wood and plastic, the lower interfacial adhesion could result in removal of larger material chunks upon sanding and therefore higher surface roughness. It appears that as coupling agent was added into

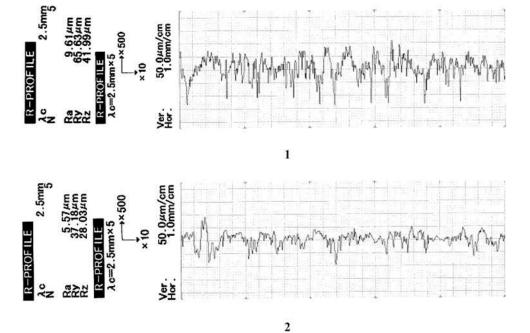


Table 2 Variations in average surface roughness, contact angle, and adhesive bonding strength values of the WPC panels **Tab. 2** Mittlere Oberflächenrauheit, Kontaktwinkel und Klebefestigkeit der WPC-Platten

WPC panel type	Panel density (g/cm ³)	Surface roughness parameters			Contact angle measuring intervals				Adhesive	
		R _a (μm)	R _y (μm)	R _z (μm)	5 s Degree (°)	10 s Degree (°)	30 s Degree (°)	60 s Degree (°)	90 s Degree (°)	bonding strength (N/mm ²)
A	0.79 (0.04)	6.10 A ^a (0.27)	50.73 A (3.12)	33.23 A (2.23)	112.8 A (3.22)	110.5 A (4.32)	107.5 A (4.65)	106.2 A (3.58)	103.3 A (2.94)	1.58 A (0.54)
В	0.80 (0.05)	7.88 B (0.35)	62.21 B (3.62)	44.12 B (2.12)	105.4 B (2.75)	103.7 B (3.92)	101.4 B (4.17)	99.5 B (3.24)	96.4 B (3.42)	1.73 B (0.32)
С	0.80 (0.06)	9.67 C (0.46)	71.25 C (5.12)	55.78 C (3.22)	98.3 C (2.66)	96.0 C (3.71)	93.3 C (3.45)	90.8 C (3.77)	88.5 C (2.66)	1.92 C (0.74)
D	0.79 (0.03)	5.52 D (0.22)	47.14 D (3.88)	30.86 A (2.16)	115.4 A (5.15)	113.1 A (3.22)	111.8 D (4.16)	108.3 A (4.54)	104.2 A (4.35)	1.53 A (0.61)
E	0.79 (0.02)	7.34 B (0.38)	58.24 E (4.32)	41.78 B (3.76)	107.2 B (5.12)	104.8 B (4.17)	102.7 B (3.72)	100.4 B (2.86)	97.9 B (2.78)	1.69 B (0.34)
F	0.80 (0.05)	9.14 C (0.55)	68.85 C (4.79)	51.42 D (3.45)	101.6 C (3.78)	99.3 C (3.45)	96.2 C (3.26)	94.1 C (3.12)	90.4 C (3.55)	1.88 C (0.72)

Values in parentheses are standard deviations

Fig. 1 Typical surface roughness profiles of panel type C (1) and panel type D (2) Abb. 1 Typische Oberflächenrauheitsprofile der Platte (1) vom Typ C und (2) vom Typ D



the panels their surface roughness decreased due to having well-developed contact between wood fiber and plastic on the surface layers. When the PP powder is melted by press platens in the hot press, it fills capillaries (micropores) in wood and this results in the smoother surface. Similar findings were also reported by Gupta et al. (2007).

The average contact angle values of each type of WPC formulation are presented in Table 2. The contact angle values of the WPC samples without veneer sheet were significantly affected by the increasing portion of wood fibers. For the contact angles values measured within 5 s, the lowest contact angle of 98.3° was obtained from the samples con-

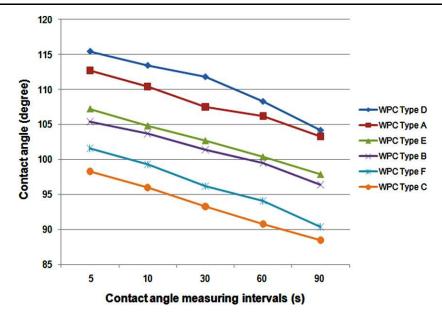


^aGroups with same letters in column indicate that there was no statistical difference (p < 0.01) between the samples according to Duncan's multiple range test

Fig. 2 Effects of rubberwood fiber/polypropylene ratio and coupling agent on the contact angle of the WPC panels

Abb. 2 Einfluss des Gummibaum/Polypropylen-Verhältnisses und des Haftvermittlers auf den Kontaktwinkel der

WPC-Platten



taining 40% wt. PP powder (panel type C) while the highest contact angle value of 115.4° was found for the samples containing 57% wt. PP powder and 3% wt. MAPP (panel type D). The increase in contact angle may be interpreted as a decrease in hydrophilicity (Sernek 2002). The WPC surfaces with higher plastic content (60% wt.) were less polar and thus repelled water, resulting in a lower wettability than in the case of the WPC samples having higher wood fiber content (60% wt.). This is expected because wood is a hydrophilic porous composite of cellulose, lignin and hemicelluloses polymers that are rich in functional groups such as hydroxyls readily interact with water molecules by hydrogen bonding, whereas thermoplastic is hydrophobic and non reactive (Clemons 2002). Wood also has a critical surface energy in the $40-60 \text{ mJ/m}^2$ range (Gupta et al. 2007). On the other hand, PP has very low surface energy $(20-25 \text{ mJ/m}^2)$, is hydrophobic, devoid of functional groups and develops smooth surfaces (Inagaki 1996). This large difference between PP and wood is what causes PP to be water repellent or hydrophobic. It should be noted that the lower wettability of rough surfaces may be due to the higher amount of peaks and valley points on the surface where liquid can be captured by capillary force. Surface roughness was proposed to enhance intrinsic adhesion by providing greater interfacial area and some mechanical interlocking mechanism. A low contact angle is very important to capillary flow in the complex porous structure of wood to achieve a strong bond between adhesive and material surface. Therefore, the lower contact angle on the surface should be analyzed as a function of the surface roughness.

MAPP compatibilizer has been extensively used in wood fiber and polymer composites to improve the filler/fiber bonding and in turn to enhance the water resistance (Clemons 2002). Wettability of the WPC panels was neg-

atively affected by the addition of the MAPP. The addition of MAPP in the WPC formulations decreased the adhesive bonding strength but this decrease was not significant as compared to the WPC without MAPP at the same wood fiber content (Table 2). The WPC samples without MAPP had better wettability (lower CA values) than those of the samples with MAPP (Fig. 2). This was attributed to polar interactions between MAPP and hydroxyl groups on the fiber surface. The anhydride groups in the MAPP enter into an esterification reaction with the surface hydroxyl groups of wood fibers and covalently bond to the hydroxyl groups (Adhikary et al. 2008). With the decreasing hydroxyl groups on the fiber surface, hydrogen-bonding sites for water molecules decreased on the WPC surface and this resulted in a higher contact angle value. Based on the findings obtained from the contact angle values, it can be concluded that decreasing hydroxy groups on the fibers resulted in a lower wettability on the panel surface.

3.2 Delamination strength between WPC surface and wood veneer sheet (adhesive bonding strength)

The adhesive bonding strength of the WPC panels decreased with increasing PP content from 40 to 60% wt. For example, the average bonding strength value of the panels containing 40 % wt. PP (panel type C) was 1.92 N/mm² as compared to panels containing 60% wt. PP (panel type A) which was 1.58 N/mm². All treatment groups without MAPP were significantly different from each other. The addition of MAPP in the WPC formulations decreased the adhesive bonding strength but this decrease was not significant as compared to WPC panels without MAPP at the same wood fiber content (Table 2). Adhesive bonding strength values of the WPC panels as compared to those of wood based panel



such as medium density fiberboard were lower. In a previous study, average adhesive bonding strength of the commercial MDF panels having a density of 0.81 g/cm³ was found as 2.30 N/mm² (Ayrilmis and Winandy 2009). The hydrophobic character of the WPC panels having higher PP content could diminish the ability of waterborne thermoset adhesives (aminoplasts) such as UF and melamine/ureaformaldehyde resins to adequately wet the surface and establish physical adhesion. As the UF resin used to adhere the veneer to the WPC sample was a polar adhesive, it needed to wet the fibers and PP to achieve adequate bonding and to then develop bonds. Studies on the surface treatments and adhesion properties of WPCs are scarce. In a previous study, the surface of WPCs can be modified using one or more surface treatments including chromic acid treatment, oxygen plasma treatment, Benzophenone/UV treatment, and flame treatment to improve adhesive bonding (Gramlich et al. 2006).

Micropore closure affects also adhesive penetration and wetting of the wood cell walls. The closure of larger micropores limits penetration by larger resin molecules, and thus, the bond strength and wood failure decreases (Wellons 1980). This applies particularly in those cases where mechanical interlocking plays an important part of the adhesion. The lower adhesive bonding strength of the samples with MAPP was attributed to the fact that the melted polymer filled capillaries in hot press and limited penetration of resin molecules into the wood. It is evident that the bonding (delamination) strength of the WPC panels was decreased with increasing contact angle values. The results of this study are consistent with previous studies on fiberboard and laminated wood (Ayrilmis and Winandy 2009; Korkut et al. 2008; Poncsak et al. 2007). In a previous study, a significant relationship ($R^2 = 0.92$) between the adhesive bonding strength and the contact angle value of MDF was found (Ayrilmis and Winandy 2009). This significant relationship indicated that contact angle could be an indicator for the degree of adhesive bonding strength of the MDF. Similar results were also observed for the WPC panels in the present study.

4 Conclusion and further work

Surface properties of the flat-pressed WPC panels were significantly affected by the wood fiber/polymer ratio. With increasing polymer content, the surface roughness of the WPC panels improved while their wettability and adhesive bonding strength were negatively influenced. The WPC formulations without MAPP were found to have higher surface roughness but they also had better wettability. The addition of the MAPP in the WPC formulations decreased the adhesive bonding strength but this decrease was not significant

as compared to the WPC panels without MAPP at the same wood fiber content. Wettability and surface roughness of the WPC panels can provide good information on their ability to bond. Further study will focus on new surface treatment applications to improve overlaying properties of the flat-pressed WPC panels.

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