Influence of Adhesive Type and Content on the Properties of Particleboard Made from Sunflower Husks

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The suitability of using milled sunflower husks as a wood substitute for producing medium-density particleboard was investigated. Additionally, the impact of the adhesive type and the amount used on the properties of the panels were evaluated. Urea-formaldehyde (UF) in three commercial variants (UCL, U96, and AG), phenol-formaldehyde (PF), modified melamine urea-formaldehyde (VM), and polymeric diphenylmethane diisocyanate (pMDI), as well as mixtures of VM/AG and of PF/pMDI, were used to manufacture the panels. The adhesive content was varied between 3% and 6% for pMDI, and from 9% and 12% for the other adhesives. Higher thickness swelling (TS) and water absorption (WA) values were observed with the UF panels compared with the PF and pMDI panels. The lowest mechanical strength properties were observed for the UF panels, with the commercial variants ranking (from highest to lowest): UCL > VM/AG > U96. Increasing the adhesive content level resulted in better dimensional stabilities and mechanical properties for the pMDI and PF panels, which met some of the performance requirements for interior uses prescribed by the relevant standard.

Keywords: Sunflower husks; Urea-formaldehyde; Phenol-formaldehyde; Melamine urea-formaldehyde; Polymeric diphenylmethane diisocyanate; Particleboard; Panels; Mechanical strength

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INTRODUCTION

Forests not only supply wood for the rapidly growing composite industry, but they also help to support a healthy ecosystem and a sustainable environment. Hence, it is important to preserve forest resources while at the same time to develop new building products.

Global wood panel production has increased rapidly, with a 123% increase in 2016 (416 million m^3) when compared with 2000 (FAO 2017). China, the USA, Russia, Canada, and Germany represented the five largest producers and consumers, which accounted for 69% of global output in 2016 (FAO 2016). In Europe, wood-based panel production grew after 1990, from approximately 30 million m^3 to 86 million m^3 because of the development of new products, such as medium-density fiberboard (MDF) and oriented strand board (OSB), as well as new investments made in Eastern Europe. These investments have contributed to a 70% increase in Eastern Europe’s panel production in 2017 when compared with 2000 (FAO 2017).

Romania is one of the Eastern European countries that have made significant investments in wood-based panels after 2008. In the last few years, new capacities were developed, which increased wood panel production to 6 million m^3 (2016), compared with 1.5 million m^3 in 2008 (FAO 2017). This increased production has placed some strain on the existing forest resources. In the near future, it is anticipated that available forest resources will be insufficient to satisfy all the wood demands while maintaining requirements for forest sustainability. This would require the promotion of new raw materials, such as agro-waste resources.

Short rotation wood could compensate for the forest resource deficits in Romania, but the areas harvested with these plantations at the national level represent less than 5% (Nicolescu and Hernea 2018). A more preferable alternative to wood is renewable agricultural residues. Currently, Romania has the potential for high-yield agricultural production; Romania is a typical agricultural country, with 62% of the arable land area (8.8 million ha) dedicated to agricultural activities (UNECE 2012). The most important agricultural crops are maize (corn), wheat, barley, sunflower, and soybean (soya). Agricultural residue wastes, such as straw, stalks, stems, and corn cobs, have the potential to be used as raw material for wood-based composites.

In Romania the annual crop residue amounts are between 10.2 and 27.0 dry Mt/year, of which approximately 4.7 to 12.6 dry Mt/year are collected (Scarlat *et al*. 2011). More than 50% of these unused residues are disposed into landfills, which contributes to environmental pollution. Another proportion of the unused residuals (approximately 46%) are used in animal feed and as fuel pellets for heat production. Sunflower (*Helianthus annuus* L.) has relatively a short growth cycle and it is easy to adapt at different soil conditions, thus is cultivated worldwide on a surface of 26.2 million hectares, reaching a production of 47.34 million tons in 2016 (Soare and Chiurciu 2018). Sunflower is the most important agricultural crop cultivated in Romania over 1 million hectares, followed by rapeseed and soybean. Sunflower production has increased in recent years, which has made Romania a leader in the European Union (EU); it accounted for approximately 24% of the total EU production in 2016 and 2017 (*i.e*., 1.95 to 2.25 million metric tons) (Dobrescu 2017). In 2018 Romania has maintained its first place in the harvested sunflower production, with 1.785 million tons from 7.906 million tons of EU 28 production. At the world level, Romania contributed with 4.29% to the world production, being on the 5th place among Ukraine, Russia, Argentina and China (Eurostat 2019). The sunflower is basically used for the oil production, in EU amounting about 7.6 million tons of the crushed seed (Nazlin *et al*. 2017). The sunflower oil produced in Romania is expected to increase slightly, reaching 0.338 million metric tons (2017/2018) (Dobrescu 2017) and 3.800 million tons in Europe, in 2019 (Krautgartner *et al*. 2019). The percentage of husks in sunflower seeds varies between 10% and 30%, which depends upon the dehulling process used (Wan *et al*. 1979; Isobe *et al*. 1992; Heinrich 2017; Kumar 2018). The density of the husks is very low (212 kg/m^3 at 10% moisture content) (Gamea 2013). Based on the husks ratio in the seeds it can be estimated that approximately 3.42 million tons of husks annually become available from the dehulling process in EU. As a consequence, a large area of storage is necessary for oil producers, therefore the wastes of sunflower husks could be degraded in time and pollute the environment. Generally, these husks are used for fuel pellets, briquettes, xylose extraction, fertilizer, and animal feed. The husks are high in fiber and low in protein, and therefore have a very low commercial feed value (Le Clef and Kemper 2015). Their use in the particleboard production is very scarce. Taking in consideration the pressure put on the forest resources, the intensifying of trees harvesting to meet the production demand, these husks (by-products) could be a potential new resources for the particleboard production. The principal constituents of the husks are cellulose (27.43%), lignin (24.23%), hemicellulose (29.04%), and extractives (9%) (Popescu *et al*. 2013), similar to those of hardwood species.

A literature review indicates that research has been conducted to make composites from sunflower residues (*e.g.*, stalk, husks, and by-products obtained after oil extraction from the seeds). These residuals have been combined with aspen wood particles (Gertjejansen *et al*. 1972), cement (Sisman and Gezer 2013), cotton waste (Binici and Aksogan 2014), Calabrian pine, poplar wood particles (Bektas *et al.* 2005; Guler *et al.* 2006), polypropylene (Kaymakci *et al*. 2013), and chitosan (Mati-Baouche *et al.* 2014) to form composites. Most of the developed composite panels that utilize sunflower wastes are combined with a variety of other raw materials, such as agricultural wastes (corn, rice, wheat), wood particles (poplar, pine, aspen), and inorganics (plaster and concrete). The use of these raw material mixtures in particleboard manufacturing involves the time and cost for collecting, storing, milling, defibration, sorting, heat-treating, and pressing operations.

According to recent data it could be estimated that the adhesives used in Europe for particleboard production are: ureo-formaldehidic (UF) (90 to 92%), melamino-ureo-formaldehidic (6 to 7%), and polymeric diphenylmethane diisocyanate (pMDI) (1 to 2%) (Kutnar and Burnard 2013). Phenolic resin (PF) is the second important bonding adhesive after UF, employed in the manufacture of wood based panels (Athanassiadou *et al*. 2015; Sandberg 2016). Their amount required by technology is between 9 and 12% (Ayrilmis and Nemli 2017; Laskowska and Mamiński 2018). The physical and mechanical properties of sunflower-based particleboard are lower than those of wood particleboard when UF and PF adhesives are used (Bektas *et al*. 2005; Kwon *et al*. 2014; Guler 2017). PMDI and emulsified pMDI in water (EMDI) have been found to be good substitutes for formaldehyde-based adhesives, leading to improved mechanical properties for the panels formed (Franke *et al*. 1994; Tongboon *et al.* 2002; Papadopoulos *et al.* 2002; Preechatiwong *et al*. 2007; Garay *et al*. 2009; Dukarska *et al*. 2017).

The objectives of this research were to produce particleboards made from 100% sunflower husks that possessed physical and mechanical properties approaching that of wood particleboards, as well as to evaluate how various adhesives and their usage levels affect the physical and mechanical properties of the resulting panels.

EXPERIMENTAL

Lignocellulosic Material

Sunflower husks (*Helianthus annuus* L.*)* were used as a raw lignocellulosic material to manufacture particleboards. The husks from the sunflower seed dehulling process were obtained from a Romanian sunflower oil manufacturer. The hammer-milled husks were sieved through 4- and 0.5-mm mesh screens to remove oversized and undersized particles. The accepted fraction had particles with lengths from 2.55 to 4.76 mm, widths from 1.05 to 2.3 mm, and thicknesses of 0.2 mm. The screened husks were dried to 4% moisture content.

Adhesives

The following adhesives were used to manufacture sunflower husk particleboards: urea-formaldehyde (UF), phenol formaldehyde (PF), modified melamine-formaldehyde (VM), and polymeric diphenylmethane diisocyanate (pMDI). Three commercial variants of UF were tested (UCL, U96, and AG); these variants differed from one another regarding the synthesis method and the formaldehyde/urea (F/U) molar ratio (1.15, 0.96, and 1.09, respectively). Mixtures of VM/AG (20:80 wt. ratio) and PF/pMDI (70:30 wt. ratio) were also used. PMDI today is generally applied in the European OSB industry (Stroobants and Grunwald 2014). The level applied is different depending on the product. In the OSB manufacturing the adhesive content ranges between 1.5% to 5%, and for particleboard an accelerator is added to the UF, and the combination in the core layer amounts to a percentage of 0.3% to 0.5% (Mantanis *et al.* 2017). Some research employed pMDI at rates of 1%, 2%, 3%, 4% and 6% for particleboard manufacturing (Papadopoulos *et al.* 2002; Korai and Ling 2011). Generally the hot pressing temperature varies from 180 °C to 240 °C (Papadopoulos *et al.* 2002) and the pressure time from 3 min to 6 min (Papadopoulos *et al* 2002; Korai and Ling 2011; Dukarska *et al.* 2017; Solt *et al.* 2019). A higher temperature was used for boards with pMDI, to reach sufficient temperature to allow the resin to cure. PMDI provides high bond strength, faster reaction time and superior resistance to water (Dunky 2003), thus it was employed beside UF adhesives. The choice of adhesives for the experimental tests was based on the data provided by the literature (Papadopoulos *et al.* 2002; Ressel 2008; Mendes *et al.* 2009; Korai and Ling 2011; Ayrilmis and Nemli 2017; Dukarska *et al.* 2017; Laskowska and Mamiński 2018; Solt *et al.* 2019). The adhesive types and the pressing schedule are presented in Table 1.

The solid resin content was based on the oven-dry weight of the husk particles. Ammonium chloride (NH_4Cl) was used a hardener for urea-formaldehyde resins and was added at 1.5% (based on the weight of the dry resin). All adhesives were obtained from Viromet SA (Victoria, Romania).

Table 1. Adhesive Content Level and Pressing Parameters

Board code	Adhesive content level (%)		Pressing parameters	
			Temperature (°C)	Pressing time (min)
UCL	9	12	150	7
U96	9	12	150	7
VM/AG	9	12	150	7
PF	9	12	160	7
pMDI	3	6	180	4
PF/pMDI	9	-	180	7

Panel Forming and Pressing

The milled husk particles were weighed and mixed with the selected adhesive in a blender. Panels with lateral dimensions of 420 mm x 420 mm were manually formed with a homogenous single-layer structure. Panels were hot-pressed at 2.45 N/mm^2 to obtain a target density of 600 kg/m^3. The pressing conditions are presented in Table 1. Two replicates were made for each panel type. After pressing, the panels were conditioned at 20 °C and 65% relative humidity until they reached equilibrium moisture content; the conditioned panels were trimmed to nominal lateral dimensions of 400 mm x 400 mm, with a thickness of 16 mm.

Physical and Mechanical Strength Characterization

Water absorption (WA) and thickness swelling (TS) tests were performed in accordance with the EN 317 (1993) standard. The density of the panels was measured in accordance with the EN 323 (1993) standard. The density profile was measured using a compact X-ray density profile analyzer (DPX300; Imal S.R.; Modena, Italy). The mechanical tests were performed using a Zwick/Roell Z010 universal-testing machine (Zwick/Roell; Kennesaw, GA) that was equipped with a ±10-kN load cell. The modulus of rupture (MOR), modulus of elasticity (MOE), and internal bond strength (IB) were evaluated in accordance with the EN 310 (1993) and EN 319 (1993) standards, respectively. Ten measurements were performed for each property tested; the reported values are the average of the measurements. One-way analysis of variance (ANOVA, using Microsoft Excel) was performed to evaluate the statistical effects of adhesive type and content level on the properties of the panels. A statistical significance level of $\alpha \leq 0.05$ was selected.

RESULTS AND DISCUSSION

Morphological Characterization

The panels produced with sunflower husk particles appeared rigid and strong. The particles showed good cohesion and were not easily detached (Fig. 1).

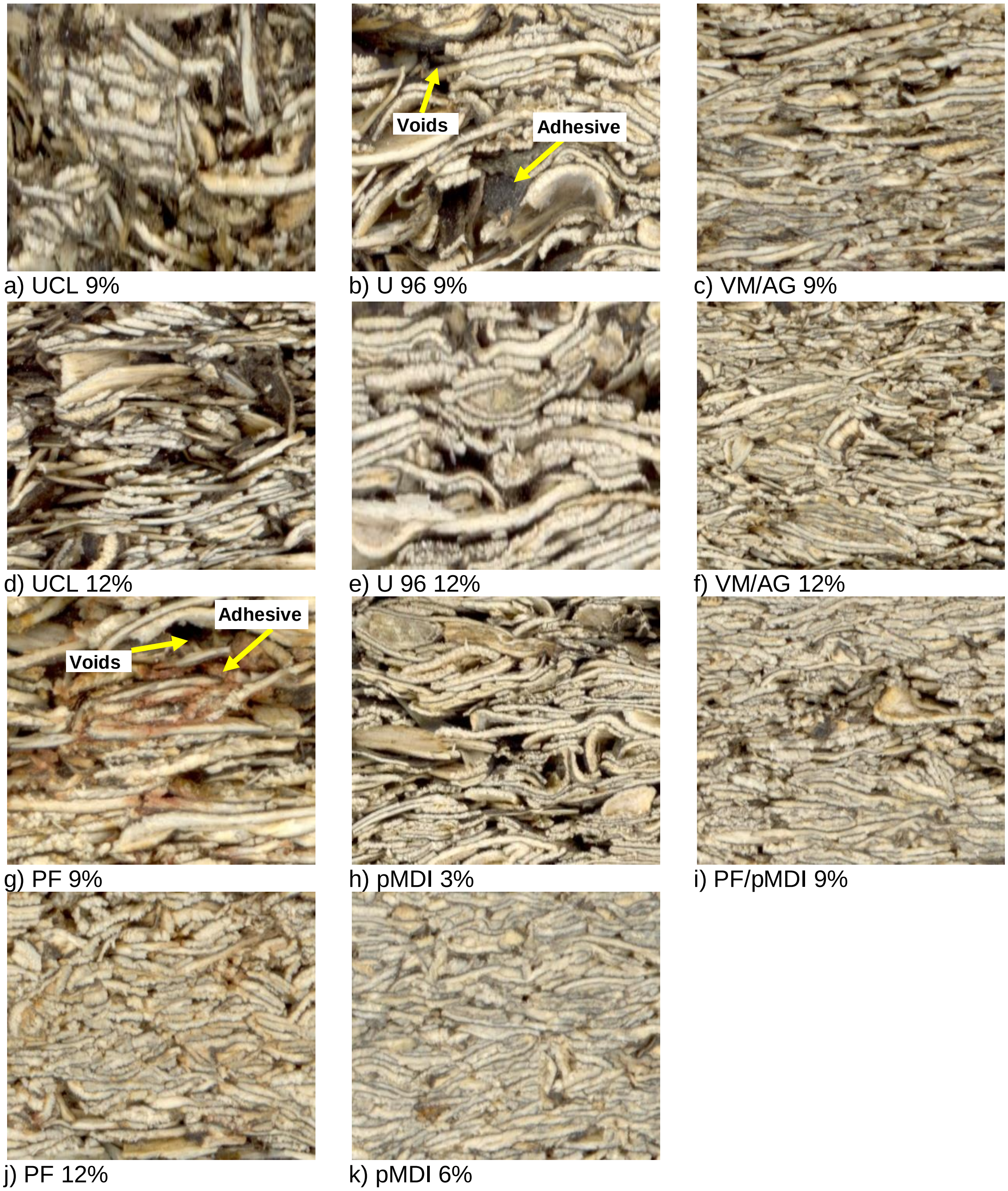
Fig. 1. Outside appearance of the experimental panels

More compact structures and uniform distributions of particles were observed for the panels made with 6% pMDI, 12% PF, and 12% VM/AG. It appeared that these adhesives were uniformly distributed over the surface of the particles and filled the voids between the particles to provide adequate adhesion among the particles. Moreover, pMDI penetrated into the amorphous components of the husk cell wall at the molecular level and led to plasticization, improving the thickness swelling resistance of panels (Frazier 2003). The internal morphologies of the various panels are more clearly observed in the high-definition photographs shown in Fig. 2 (originally 4800 dpi resolution).

For 3% pMDI, the panel structure was less compact and the adhesive partially adhered to some particles, resulting in the formation of localized agglomerations. The heterogeneous distribution of husk particles and adhesive led to more voids in the structure when urea-formaldehyde adhesives were used.

Physical and Mechanical Characterization

Figure 3 shows the vertical density profile (VDP) of the panels. These graphs exhibited a typical density profile caused by mat densification. A higher peak density was found 1 mm and 3 to 4 mm from the surface areas for the UF panels and PF/pMDI panels, respectively (Fig. 3; a through f, g through k). The core of the panels had a more pronounced U-shaped profile in the cases of UCL and U96 adhesives (Fig. 3; a, b, d, and e.).

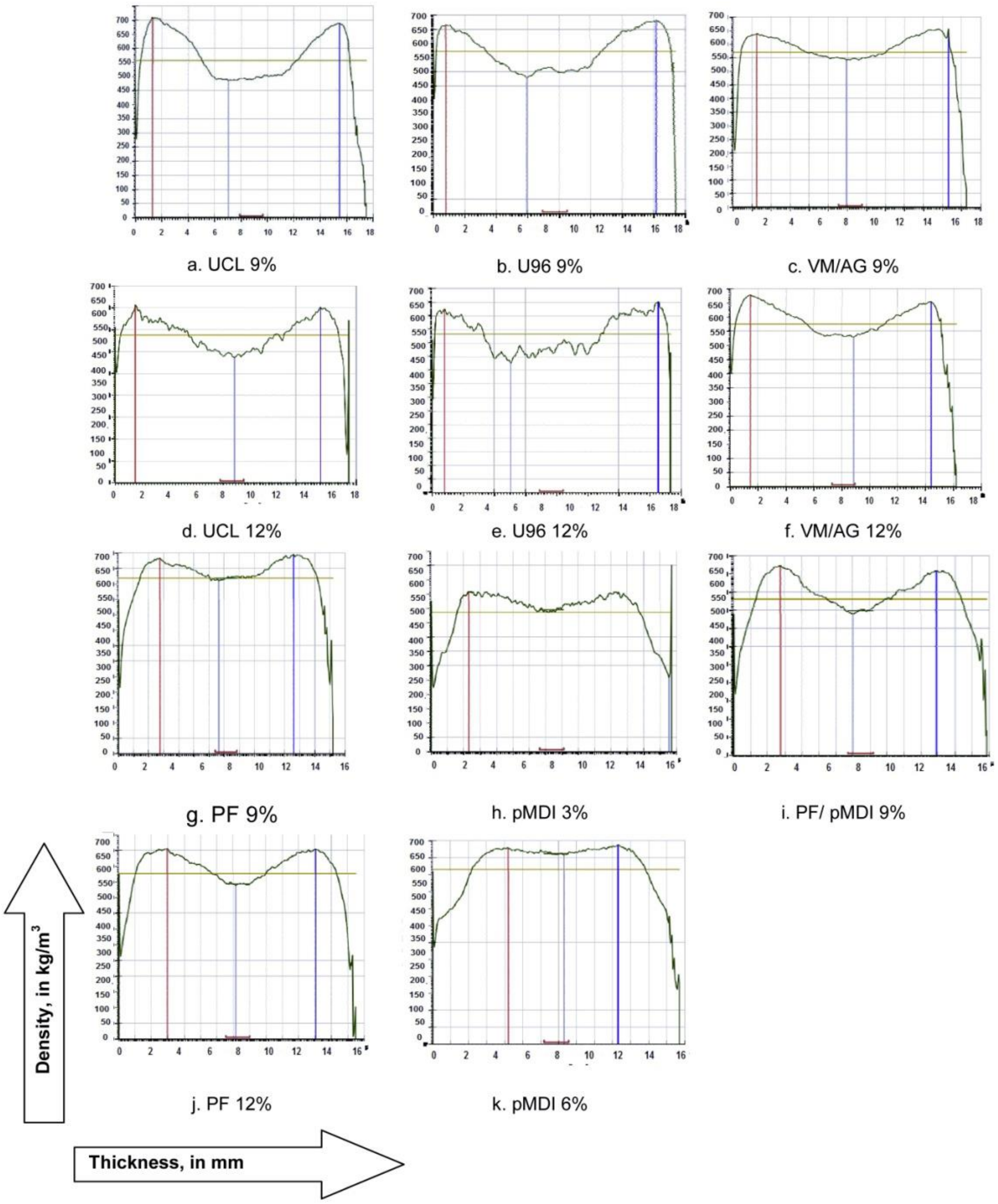


a) UCL 9%
b) U 96 9%
c) VM/AG 9%
d) UCL 12%
e) U 96 12%
f) VM/AG 12%
g) PF 9%
h) pMDI 3%
i) PF/pMDI 9%
j) PF 12%
k) pMDI 6%

Fig. 2. Morphological details of the experimental panels (originally 4800 dpi resolution)

The mean densities of the panels (approximately 550 kg/m^3) were similar to one another except for the pMDI panels, which exhibited the lowest density (490 kg/m^3) (Fig. 3; h and i).

Panels made with pMDI had a relatively flat-shaped density profile (Fig. 3; h and k). In these cases, the pressing time was shorter when compared with the other panels; this may have affected the surface density by minimizing the differences between the lateral edges and the core.



a. UCL 9%
b. U96 9%
c. VM/AG 9%
d. UCL 12%
e. U96 12%
f. VM/AG 12%
g. PF 9%
h. pMDI 3%
i. PF/ pMDI 9%
j. PF 12%
k. pMDI 6%

Fig. 3. Vertical density profiles of the experimental panels

The experimental panels can be classified as medium-density particleboard (below 600 kg/m^3). The lowest density was obtained for pMDI panels because of the low specific gravity of the adhesive (Rachtanapun *et al.* 2012). The average values of the physical properties are presented in Table 2. The moisture content of all panels was approximately 6.3%. The highest thickness swelling (TS) and water absorption (WA) values after 24 h of water immersion were observed for the panels made with UF adhesives; such adhesives are known to have low water resistance. Similar results in thickness swelling (TS 24h 49% ÷ 44%) were observed in the research made by Melo *et al.* (2014), who also observed a highest instability of particleboards that used only rice husk with urea-formaldehyde adhesive. The panels made with PF had the lowest TS and WA values (20% and 69%, respectively); these values were almost one-half the values observed for the UF panels. This behavior is attributed to the high water resistance of the methylene carbon-carbon bonds linking the aromatic nuclei of PF (Dunky 2003). A differential decrease in WA and

TS values was noted as the adhesive content increased for all panels, except for the PF panels. The lower density of the PF panels made with 12% adhesive could have induced more water penetration into the panel structure when compared with the 9% adhesive panel.

Table 2. Physical Properties of the Experimental Panels

Properties	Adhesive content (%)	Density (kg/m^3)	TS 24h (%)	WA 24h (%)
UCL	9	556 (27.8)	41.2 (6.1)	125.3 (11.6)
	12	573 (21.6)	38.4 (5.6)	110.5 (13.3)
U96	9	577 (39.0)	44.5 (1.5)	140.2 (3.1)
	12	580 (21.0)	41.2 (6.1)	126.4 (10.1)
VM/AG	9	570 (40.0)	40.3 (0.6)	124.9 (3.4)
	12	574 (23.8)	32.2 (1.2)	106.9 (15.4)
PF	9	570 (16.7)	19.9 (3.3)	69.5 (6.2)
	12	524 (29.3)	20.8 (3.7)	68.7 (6.0)
pMDI	3	495 (20.9)	56.0 (3.6)	131.7 (6.4)
	6	535 (19.3)	29.7 (1.6)	76.0 (14.4)
PF/pMDI	9	520 (16.5)	31.4 (0.6)	81.3 (4.9)
The numbers in parentheses are the standard deviations				

A slight decrease in TS and WA (of approximately 7% and 10%, respectively) was observed for the 12% UF adhesive panels (UCL and U96) *versus* the 9% UF adhesive panels. These results are consistent with those of sugar cane bagasse particleboards reported by Mendes *et al.* (2009). Garay *et al*. (2009), registered a higher level of TS 24h and WA 24h (over 60% and 100% respectively), even when UF was increased from 10% to 15%, by using rice and wheat husk in different proportion with wood, for particleboard manufacturing. Klimek *et al*. (2016) observed that TS 24h of sunflower stalk particleboards did not show significant differences when increases the dosage of UF from 8% to 10%. They reported similar values compared to the present data for UF and MDI adhesives. Better results were recorded for VM/AG and pMDI panels, for which TS and WA decreased by 24% to 47% and 24% to 42%, respectively. The melamine resin and polyisocyanate components of these adhesives form strong bonds, conferring stability to the formed panels. Similar results were reported by Hse and Choong (2002) and Klimek *et al*. (2016) for rice hull-wood and sunflower stalks particleboards with polyisocyanate adhesive. Unexpectedly, the 3% pMDI panels had the highest TS and WA values (56.6% and 131.7%, respectively). It seemed that the adhesive was heterogeneously distributed onto the surfaces of the husk particles, which also led to a decrease in internal cohesion. All panels had TS values that were higher than the 14% limit recommended for P3 type panels (EN 312 (2013)). The present results on TS and WA were relatively high but are consistent with literature (Hse and Chong 2002; Guntekin and Karakus 2008; Garay *et al.* 2009; Melo *et al*. 2014; Guler 2015; Kord *et al*. 2016) and can be explained by the fact that no paraffin or other hydrophobic substances were added in the boards. Additionally, the complex interrelation between chemical composition, the size of particles, and their interaction with adhesives influenced the performance of the boards.

As expected, the use of pMDI and PF adhesives resulted in better panel stability regarding water penetration (*i.e.*, lower TS and WA) and higher IB than did the use of UF adhesives (Fig. 4). In general, the IB value increased in a somewhat linear fashion as the TS and WA values decreased. The highest IB values were observed for the pMDI and PF panels.

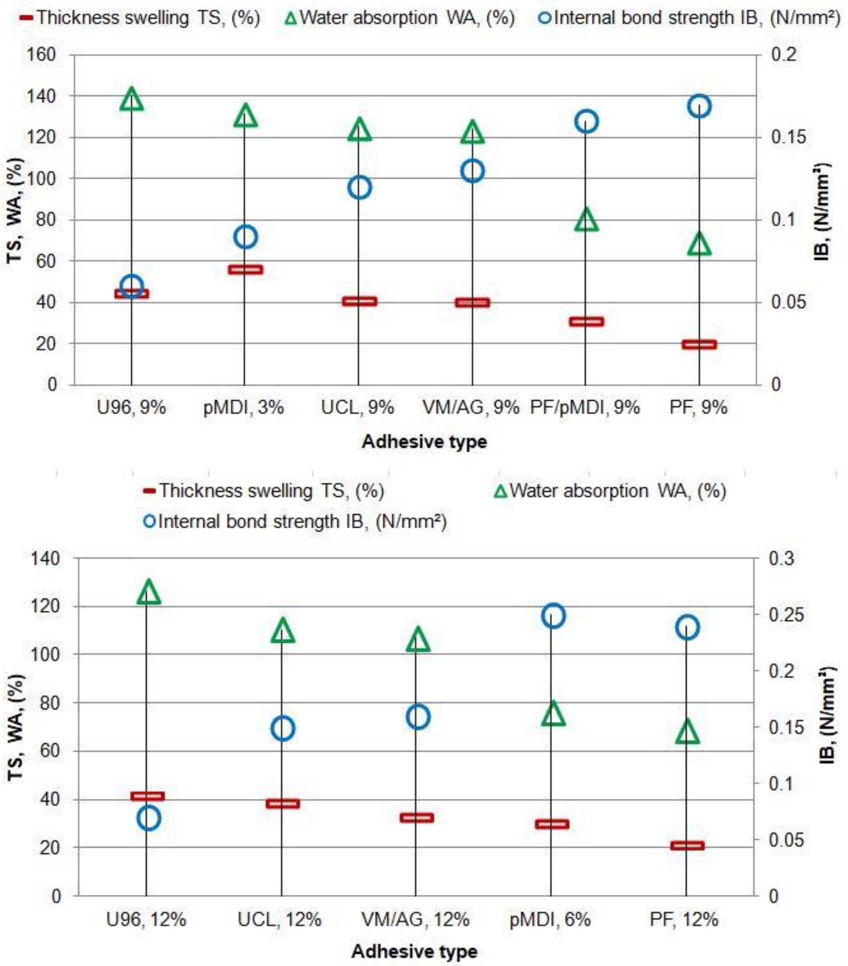


Fig. 4. Effect of adhesive type and its content level on the thickness swelling (TS), water absorption (WA), and internal bond strength (IB) of the panels

The adhesive type was found to have a significant influence on the measured TS and WA values at the 95% confidence level. The statistical impact of adhesive content level was more evident for pMDI (*p*-value less than 0.0001) than for UF (*p*-value of 0.004).

Mechanical Properties

The MOR and MOE values are presented in Fig. 5. The highest strength values were noted for PF and pMDI panels at the highest adhesive content level.

The UF panels had the lowest mechanical strength. Depending on the UF type, the average decrease in strength varied from 32% to 43% for MOR and 29% to 58% for MOE when compared with panels utilizing PF and pMDI at high adhesive content levels. Greater decreases, from 32% to 58% for MOR and 46% to 68% for MOE, were also observed at low adhesive content levels. The lowest strength results were noted for U96 panels and for 3% PMDI panels.

U96 adhesive had the lowest F/U ratio among the UF adhesives used; U96 is generally used to manufacture high-density fiberboards. This explained U96's poor performance with respect to the sunflower husk particles. An insufficient amount of adhesive in the case of 3% pMDI resulted in the lower mechanical properties of the resulting panel. The combination of UF with modified melamine-formaldehyde adhesive

(VM/AG) slightly improved the mechanical properties *versus* the other urea adhesives. Low MOR and MOE values were reported by Guler (2015) and Melo *et al.* (2014) when using other agricultural residues (hazelnut husk, peanut hull, and rice husk) combined with UF adhesive.

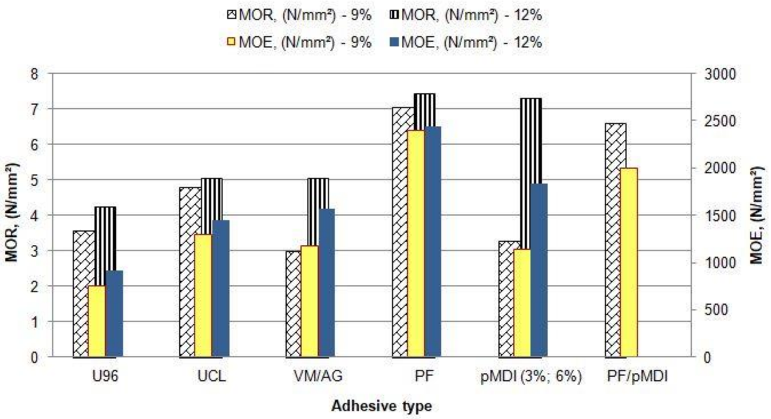


Fig. 5. Effect of adhesive type and its content level on MOR and MOE

All panels examined in this study failed to meet the minimum MOR requirements of EN 312 (2013). An increase in the percentage of agro wastes in the particleboard is expected to lead to a decrease in breaking strength and rigidity parameters (Ndazi *et al*. 2006; Melo *et al*. 2014; Klimek *et al*. 2016), and the boards generally failed to meet MOR standard P1 limits (Ndazi *et al*. 2006; Keskin *et al*. 2015; Klimek *et al.* 2016). The experimental boards had a density lower than 580 kg/m^3 and easily could be included in the low density category (Rowell 2014) where MOR, MOE and IB values are ranging from 3 N/mm^2 to 5.0 N/mm^2, 550 N/mm^2 to 1025 N/mm^2 and 0.1 N/mm^2 to 0.15 N/mm^2, respectively.

The effect of pMDI adhesive on MOR and MOE was obvious; therefore, the values obtained for 6% pMDI panels were comparable with those of 12% PF panels. This observation is similar to the results reported by Hse and Choong (2002) for 5.5% polyisocyanate adhesive used in particleboards made from rice hull mixed with wood particles. Only the PF and pMDI panels fulfilled the requirements for MOE (>1600 N/mm^2) in accordance with EN 312 (2013) for P2-type particleboard.

The average IB values ranged from 0.06 N/mm^2 (9% U96) to 0.25 N/mm^2 (12% PF and 6% pMDI). All panels bonded with UF adhesives had the lowest IB strengths at both adhesive content levels (Fig. 4). An increase in adhesive content from 9% to 12% improved the IB for all panels, which was more evident for PF (40% higher) than for UF panels (23% higher). The IB values of pMDI panels were significantly affected by adhesive content level. An IB increase of 177% was observed when the pMDI level increased from 3% to 6%; the IB values at 6% pMDI were comparable with those at 12% PF. The mixed resin system, pMDI/PF, was noted to have a lower IB than that of 6% pMDI panels, but it was comparable to those of 9% PF panels. A minimum IB value of 0.24 N/mm^2 is required for P1-type particleboards, according to EN 312 (2013); this requirement was met by 12% PF and 6% pMDI panels.

Statistical analyses showed that the mechanical properties of the experimental panels, in terms of MOR, MOE, and IB, were significantly influenced by the adhesive type (*p*-value of 0.0009). The statistical impact of adhesive content level on these values was more evident for pMDI (*p*-value less than 0.0001) than for UF and PF panels (*p*-value of 0.04).

CONCLUSIONS

1. Adhesive type had a significant influence on the physical and mechanical properties of experimental composite panels made from sunflower hulls.
2. The adhesive content level had a significant effect on the properties of the panels, with a more notable effect observed for pMDI panels than for UF and PF panels.
3. Higher TS and WA values were noted for UF panels at the two adhesive content levels, with the commercial variants ranking (from highest to lowest) U96 > UCL > VM/AG. Slightly improved properties were observed for the VM/AG panels as a result of the addition of melamine to the UF adhesive.
4. Panels with 3% pMDI adhesive content exhibited lower mechanical properties than UF panels; this was attributed to low density and low bonding efficiency when using 3% pMDI.
5. The best panel dimensional stabilities and mechanical properties were observed for PF panels at both adhesive content levels, followed by PF/pMDI (9%) and pMDI (6%). These results indicate stronger bonds between husk particles and adhesive. PF and pMDI panels were found to comply with the MOE and IB strength requirements for general use as prescribed by the EN 312 (2013) standard.
6. The results obtained revealed that sunflower husks are compatible with UF, PF and pMDI adhesives and represent an alternative material to wood in particleboard manufacturing. The performance of boards was influenced by the adhesive type and content and the low density (below 580 kg/m^3), which contributed to a decrease in strength. The experimental particleboard presents a potential for indoor use as light panels for paneling or other decorative products.

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