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WOOD POLYMER COMPOSITE BONDED VENEER BASED HYBRID COMPOSITES

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ABSTRACT

Wood veneer based composites have a great demand in present market as the material can utilize small diameter plantation timbers grown at short rotation cycle. This paper presents preparation and characterization of hybrid composites made of wood veneer and wood polymer composite. The study explored utilization of wood polymer composite as an adhesive for bonding veneers replacing formaldehyde-based adhesives. Wood polymer composite containing 40 % bamboo particles embedded in the matrix of polypropylene was used in sheet form to bind the veneers of *Melia dubia* wood. The composites were prepared in both laminated veneer lumber and plywood configurations. The assessment of physical and mechanical properties indicated that the properties of wood polymer composite contribute significantly to the properties of the hybrid composites. The density of the resultant composites was significantly higher (0,69 g/cm^3 – 0,75 g/cm^3) than conventional plywood or laminated veneer lumber. Among mechanical properties, there was no statistical difference in tensile and flexural strength of plywood and laminated veneer lumber configuration. Modulus of elasticity and compressive strength of laminated veneer lumber configuration were significantly higher than plywood. Glue shear strength and internal bond strength of the composites indicated acceptable bonding properties of wood polymer composite which suggests the potential application of these composites as a binding agent for wood veneers. These composites could be a special class of laminated composites with no formaldehyde emission hazards.

Keywords: Hybrid composite, laminated veneer lumber, *Melia dubia*, plywood, wood polymer composite,

INTRODUCTION

Wood based composites like plywood, particle boards, medium density fibre board (MDF), laminated veneer lumber (LVL) etc. have well established their niche in different applications. These composite materials facilitate optimum utilization of small diameter plantation timber species managed with lower rotation compared to traditionally used timber species (Uday *et al*. 2011). Among these composites, laminated products such as plywood and LVL offer many advantages like increased dimension stability, uniformity, improved stress distribution properties, cost effectiveness (Tenorio *et al*. 2011). Plywood has proved to be more promising compared to conventional wood as the perpendicular arrangement of the adjacent veneers provides uniformity in performance, properties in both directions with better dimension stability and also it is more resistant building material to lateral forces like earthquake and wind (Demirkır 2008).

For manufacturing these panel products, conventionally formaldehyde-based resins like urea formaldehyde (UF), phenol formaldehyde (PF), melamine urea formaldehyde (MUF), phenol resorcinol formaldehyde (PRF), etc. are used based on the ultimate end-product. However, formaldehyde emission is a major concerns in such panels during both production and utilization as long term exposure to formaldehyde is reported to be carcinogenic and can lead to various respiratory diseases (Raya *et al*. 2018; Jang *et al*. 2011; Makinen *et al*. 1999). Considerable research efforts are being made to eliminate formaldehyde emission completely or reduce it within permissible limits. New adhesives such as soya based adhesives (Raya *et al*. 2018), starch based adhesives and non-formaldehyde-based adhesives, are being explored for such panel products. (Imam *et al*. 1999; Li and Geng 2005).

The use of thermoplastic polymers as a binding agent for natural fibres in making unique composites avoiding use of any formaldehyde-based adhesive is a relatively new concept and has been attempted in recent times. Thermoplastic polymers like polyethylene (Chang *et al*. 2017, 2018, Fang *et al*. 2017, Hung *et al*. 2017, Arya and Chauhan 2022), PP (Kajaks *et al*. 2020, Song *et al*. 2017, Arya *et al*. 2022), polyvinyl chloride (PVC) (Matuana *et al*. 1998), etc. have been attempted as the binding agent. Lustosa *et al*. (2015) studied the properties of LVL prepared using high density polyethylene (HDPE) film as the binding agent and reported that the properties were comparable or even better compared to the LVL made with commercially preferred thermosetting formaldehyde-based adhesives. Chang *et al*. (2017) studied interfacial bonding mechanism of poplar plywood using on HDPE film as an adhesive, and reported that the thermoplastic was able to penetrate into vessel and xylem cells of the wood and resulting in a satisfactory bond strength which was in accordance to that of II-grade plywood.

Wood plastic composites (WPC) have emerged as a specific class of composite material utilizing lingo-cellulosic fibres as a reinforcing material to conventional thermoplastics mainly PP, HDPE and PVC. WPC is used for making injection moulded and profile extruded products. WPC are also extruded in thin sheets for thermoformed products. The technology for making WPC is already well established (Benthien and Thoemen 2012) and its market is expanding at a rapid rate globally. A recent market analysis report estimated the market size of WPC was 5, 3 billion USD in 2019 and expected to grow by 11,4 % by the year 2027 (https://www.grandviewresearch.com/industry-analysis/wood-plastic-composites-market). In North America, WPC are low priced as products are manufactured using recycled plastics and different natural fibres. Whereas in Germany and other European countries, WPCs have become an advanced material used in various speciality application (Carus *et al*. 2008). Since thermoplastic films have been successfully attempted as the binding agent for wood veneers to prepare specific class of plywood/LVL composites, it is hypothesized that the WPC can also be used as a binding agent in veneer-based composites. The present study aimed at evaluating WPC as the binding agent for making plywood and LVL creating novel hybrid composite materials which would be completely free from formaldehyde. Preparation of such hybrid

composite may also provide a strategy to recycle WPC products at the end of their life.

MATERIALS AND METHODOLOGY

Veneers - Rotary peeled veneers of *Melia dubia* (2,5 mm - 3 mm thickness) were used for this study. The species is one of the fast growing tree species extensively raised in several parts of India and prominently used for plywood manufacturing. The average moisture content of the veneers was in range of 9 % to 13 %.

Wood polymer composite sheet – Profile extruded 3 mm thick WPC sheets were used for the study. The sheets were provided by the Spectrus Sustainable Solutions Pvt. Ltd., Bengaluru, India having composition of polypropylene (55 % wt), bamboo flour (40 % wt) and maleic anhydride grafted PP coupling agent (orvac / P613, dupont make, 5 % wt). The mechanical properties of the WPC sheet material were determined in the laboratory and are given in table 1. The sheets were used as a bonding material instead of traditional formaldehyde based adhesives.

Table 1: Properties of WPC sheet prepared from PP-bamboo particles. Values in the parenthesis denote standard deviation.

Density (g/cm^3)	MoR (MPa)	Tensile strength (MPa)	MoE (GPa)	HDT (ºC)
1,01 (0,01)	58,42 (5,39)	27,47 (2,0)	3,98 (0,37)	137,8

2.1 Preparation of veneer-WPC hybrid composite

The hybrid composites were fabricated by sandwiching WPC sheets between wood veneers (Fig. 1). The orientation of the core veneer was varied to make two different types of hybrid composite namely plywood and laminated veneer lumber (LVL). In plywood configuration, the grain direction of core veneer was oriented perpendicular to the top and bottom veneers. In LVL configuration, the grain direction of all the veneers was parallel to each other. In this study, composites with three ply configuration were prepared. The assembly of veneers and WPC sheets was placed in a hydraulic press, preheated at 155 ºC - 160 ºC. The assembly was pressed at a specific pressure of 10 kg/cm^2 for 15 min. The pressed boards were allowed to cool down under pressure till the temperature reached to 65 ºC - 70 ºC to avoid warping in the board due to differential cooling. Three boards of each plywood and LVL configuration were manufactured with dimensions 300 mm X 300 mm X 10 mm. The prepared boards were conditioned at the 21 ºC - 25 ºC and 60 % - 70 % relative humidity for 24 h. Thereafter, test specimens were extracted from the boards.

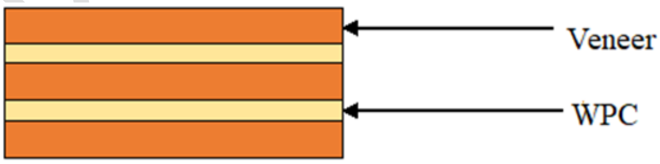

Figure 1: Schematic representation showing assembly of Veneer-WPC based boards.

Testing of Panels

The panels were tested for various physical properties namely density, volume fraction ratio, adhesion of plies (knife test), water absorption (WA), thickness swelling (TS), and volumetric swelling (VS) were measured after exposure to water for 2 h and 24 h. Mechanical properties such as modulus of rupture (MoR), modulus of elasticity (MoE), compressive strength (CS), tensile strength (TSS), glue shear strength (GSS), internal bond strength (IB), were evaluated using universal testing machine (UTM) (Shimadzu-autograph-AX) with 50 kN capacity. All the tests were carried as per recommendations given in Indian standards BIS 1734 (BIS 1983) and BIS 14616 (BIS 1999) and for each test, five replicates were taken. Statistical analysis was carried out using SPSS statistical software (IBM 2019) and t-statistics was used to determine the statistically significant differences in properties of WPC bonded ply and LVL configuration panels. The fractured surfaces of specimens used for testing tensile strengths of LVL and plywood were examined using scanning electron microscope (SEM) to understand the penetration and surface interface between WPC and wood veneers of the hybrid composites. Additionally, heat deflection temperature (HDT) measurements were carried out to observe the temperature at which the sample of ply/LVL and WPC sheet deflects. The test was carried out with loading stress of 455 KPa, heating rate of 3 ºC/min. Test was conducted for two replicates of each sample with span length of 100 mm and deflection of 0,25 mm.

RESULTS AND DISCUSSION

3.1 Physical test

The physical properties of plywood and LVL are given in table 2. The average value of moisture content of prepared plywood and LVL was observed to be 3,91 % and 4,08 %, respectively. Air dry density average value of the prepared hybrid composites was found to be 0,75 g/cm^3 and 0,72 g/cm^3 for plywood and LVL, respectively. Khali *et al*. (2017) prepared plywood using veneers taken from different progenies of *the same species* and conventional urea formaldehyde resin, pressed at 17,5 kg/cm^2. The density of the prepared plywood varied from 0,50 g/cm^3 to 0,52 g/cm^3. Similarly, Prakash *et al*. (2019) reported density of LVL prepared using *Melia dubia* and phenol formaldehyde, pressed at 16 kg/cm^2 to be 0,6 g/cm^3. The hybrid ply as well as LVL prepared in this study exhibited higher density as compared to conventional composites using the same species. The density of the *Melia dubia* wood ranges between 0,39 g/cm^3 to 0,46 g/cm^3 (Kumar *et al*. 2018; Sharma *et al*. 2019). The higher density of hybrid composite is attributed to the high density of WPC sheet (≈ 1,01 g/cm^3) instead of a thin layer of formaldehyde-based resins. There was no statistically significant difference in M.C. % and density of hybrid plywood and LVL (P > 0,05) which was on the expected lines.

Water absorption after 2 h and 24 h of soaking for hybrid WPC plywood was 7,34 % and 19,29 % respectively. Water absorption values for hybrid WPC-LVL composite after 2 h and 24 h of soaking were 7,89 % and 18,06 %, respectively. Water absorption was nearly the same in both types of composites irrespective of time of immersion. Lustosa *et al*. (2015) manufactured the LVL using HDPE and reported that the water absorption after 2 h and 24 h was ranged in between 17,78 % - 19,77 % and 43,82 % - 49,48 %, respectively, which was substantially higher as compared to hybrid WPC – LVL composite prepared in this study. Lower moisture absorption by the composites may be attributed to the effective encapsulation of core veneer by thick WPC layer and filling of pores/crevices present on wood veneer by wood fibres present in the WPC restricting free movement of water (Fang *et al*. 2014). WPC itself is reported to absorb negligible amount of moisture i.e. 3 % - 4 % even on long-term repeated cycle of wetting and drying (Gunjal *et al*. 2020). The ability of WPC's to absorb moisture drastically reduces as the wood fibres are entangled with polymers which are hydrophobic in nature. As a result the total moisture uptake capacity of the hybrid ply and LVL composites prepared by incorporating WPC reduces significantly.

Table 2: Physical properties of Plywood and LVL hybrid composites. Values in the parenthesis denote standard deviation.

Properties	Plywood	Laminated Veneer Lumber	P-values (t- test)
Moisture content (%)	3,91 (0,25)	4,08 (0,25)	0,20
Density (g/cm^3)	0,75 (0,02)	0,72 (0,02)	0,13
Water absorption (%) 2 h	7,34 (1,30)	7,89 (0,53)	0,28
Water absorption (%) 24 h	19,29 (1,54)	18,06 (0,78)	0,03
Thickness swelling (%) 2 h	3,68 (0,38)	4,33 (0,43)	0,01
Thickness swelling (%) 24 h	5,09 (0,46)	6,80 (0,40)	< 0,01

Thickness swelling on water absorption is an important parameter for veneer based panels products. Hybrid plywood exhibited thickness swelling of 3,68 % and LVL exhibited 4,33 %. After 24 h of water immersion, thickness swelling was 5,09 % in plywood panel and 6,80 % in LVL panel. The thickness swelling in plywood configuration was significantly lower ($P < 0,05$) than LVL panel after 2 h and 24 h of exposure to water though the water absorption was not significantly different. In principle, both water absorption and thickness swelling should have been similar in both types of composites. Slight difference in thickness swelling may be attributed to the natural variation in density of veneers used in fabricating the panels and it is expected that the prolonged exposure to water may lead to uniform swelling. Tenorio *et al*.

(2011) reported uniform thickness swelling in plywood and LVL made from *Gmelina arborea* wood.

Mechanical test

Mechanical properties of plywood and LVL are given in table 3. The average value of MoR in the case of plywood panel was 57,19 MPa and was in close agreement with the MoR of conventional plywood from the same species (Khali *et al*. 2017). However in case of LVL, MoR average values of hybrid composite were much lower (63,79 MPa) than PF bonded LVL of the same species (106,8 MPa) as reported by Prakash *et al*. (2019) and was not differing significantly from MoR of plywood configuration (Table 3). The low MoR of LVL is attributed to the poor flexural strength of WPC (58,42 MPa) as compared to MoR of *Melia dubia* wood along the grain (89,44 MPa) (Chauhan and Sethy, 2016). The thick layer of WPC is expected to contribute significantly to overall MoR of the composites as the strength of the composite laminate is expected to depend on the strength of the components of the composites, their relative volume fractions and orientation of each layer. The contribution of each layer of the laminate in the flexural strength of the composite is different with top and bottom layer influencing the most. The difference in plywood and LVL configuration panel was only the orientation of the core veneer which is on the neutral axis on bending influencing very little on overall strength of the composites. Therefore, the bending strength of both configurations were statistically similar despite of nearly six-fold difference in MoR of the wood along the grain (89,44 MPa) and across the grain (15 MPa). Menezzi *et al*. (2016) studied the mechanical properties of LVL bonded with expanded polystyrene (EPS) and reported that increase in amount of EPS had a negative effect on the MoR and MoE of the composites, also the increased amount of wood content resulted in improved flexural performance of EPS bonded LVL. Also, the amount of compression achieved during the fabrication of the veneer composites plays a crucial role in modelling the mechanical properties (Kurt and Cil 2012). In the current study, the pressure of 10 kg/cm^2 was used for only achieving uniform heat transfer through the material as well as improving surface bonding between polymer and the veneers and not for achieving higher compression of the assembly as the higher pressure may result in spilling of the melted WPC during the fabrication process.

Table 3: Mechanical properties of Plywood and LVL hybrid composites. Values in the parenthesis denote standard deviation.

Properties	Plywood	LVL	P values (t-test)
Modulus of rupture (MPa)	57,19 (2,77)	63,79 (7,64)	0,15
Modulus of elasticity (GPa)	6,89 (0,80)	8,73 (0,45)	<0,01
Compressive strength parallel to grain (MPa)	35,50 (6,54)	44,88 (1,63)	< 0,01
Tensile strength (MPa)	46,48 (7,43)	39,08 (1,36)	0,14
Glue shear strength (MPa)	1,25 (0,19)	1,15 (0,14)	0,28
Internal bond strength (MPa)	1,77 (0,15)	2,75 (0,27)	<0,01

MoE of the hybrid WPC- plywood was 6,89 GPa which was significantly lower than MoE of the hybrid WPC- LVL was 8,73 GPa. This is on expected lines as the orientation of core veneer is going to influence the overall MoE of the composites unlike flexural strength. MoE of *Melia dubia* along the grain is nearly 20 times higher than across the grain. However, MoE of LVL in this study was observed to be slightly lower that the MoE reported in conventional LVL from same species (Prakash *et al*. 2019). Low MoE of the hybrid LVL as compared to conventional composites is mainly attributed to the higher proportion of low modulus WPC present in the composites as the modulus of the composites depends on the modulus of the individual component of the composites and their relative proportions (Chauhan *et al*. 2005). The modulus of elasticity of WPC sheet was 3,98 GPa whereas MoE of *Melia dubia* veneer along the grain is 11 GPa and across the grain is 1,06 GPa. The MoE of the composites was theoretically estimated based on rule of mixture (eq. 1).

$$MOE_C = \frac{MOE_v \times Vol_v + MOE_{wpc} \times Vol_{wpc}}{Vol_C} \qquad (1)$$

Where, MOE_C = MOE of composite (GPa), MOE_V = MOE of veneer (GPa), MOE_{wpc} = MOE of WPC (GPa), Vol_v = volume of veneer (mm^3), Vol_{wpc} = volume of WPC (mm^3), Vol_C = volume of composite (mm^3).

The predicted MoE for plywood configuration was found to be 6,16 GPa and for LVL configuration it was 8,10 GPa which were in close agreement with the observed MoE of composite panels. Using WPC instead of pure polymer in binding veneers provides added advantages in terms of higher volume proportion of natural material in the overall composites formulations. The volume faction of polymer and wood can be estimated by the following equation (eq. 2) (Ashok 2015).

$$\text{Volume fraction \% of wood vj\%} = \frac{\frac{\mathrm{wv}}{\mathrm{\rho v}}+\frac{\mathrm{wb}}{\mathrm{\rho b}}}{\frac{wv}{\rho v}+\frac{wb}{\rho m}+\frac{wp}{\rho p}} \times 100 \qquad (2)$$

Where, wv = weight of veneer (g), wb = weight of bamboo (g), wp = weight of polymer (g), ρv = density of veneer (g/cm^3), ρb = density of bamboo (g/cm^3), ρp = density of polymer (g/cm^3). The volume fraction of overall wood component (wood veneers and wood in WPC) was estimated to be 68,39 % when WPC with 40 % wood content was used.

The average value of compressive strength parallel to grain ($CS_{\parallel}$) for hybrid plywood was 35,50 MPa whereas, the hybrid WPC–LVL showed significantly higher $CS_{\parallel}$ i.e. 44,88 MPa. Tensile strength ($TSS_{\parallel}$) of hybrid plywood was not significantly differing from hybrid LVL. Glue shear strength average value of hybrid WPC-plywood and LVL was 1,25 MPa and 1,15 MPa, respectively. GSS mainly reflects the strength of bond against slippage between WPC layer and veneer on tensile force. Chang *et al*. (2017) reported GSS of 1,50 MPa in plywood prepared with HDPE which is in close agreement with the current study.

Figure 2: Internal bond sample after delamination.

Interestingly, average value of IB for hybrid WPC - plywood (1,77 MPa) was significantly lower than average value of IB in hybrid WPC - LVL (2,75 MPa) and mostly wood failure was observed (as shown in Fig. 2) indicating effective bonding between wood and WPC layer which is attributed to the deeper penetration of WPC in veneers. Further in order to investigate the bond strength, knife test was carried as per specifications given in BIS 1734 (BIS 1983) on the dry and wet (boiled in water for 2 h) specimens by pushing a sharp knife with its cutting edge parallel to the grain of the face veneer. After insertion, the knife was pulled upwards. The specimens showed excellent bond as the penetration of knife was difficult and after prising upwards the veneer breaks off instead of completely pulling out (Fig. 3). Since, WPC melts at higher temperature i.e. 160 ºC - 170 ^{0}C, boiling in water does not have any detrimental effect on the bonding mechanism of these hybrid composites compared to the conventional plywood and LVL especially prepared using urea formaldehyde adhesives.

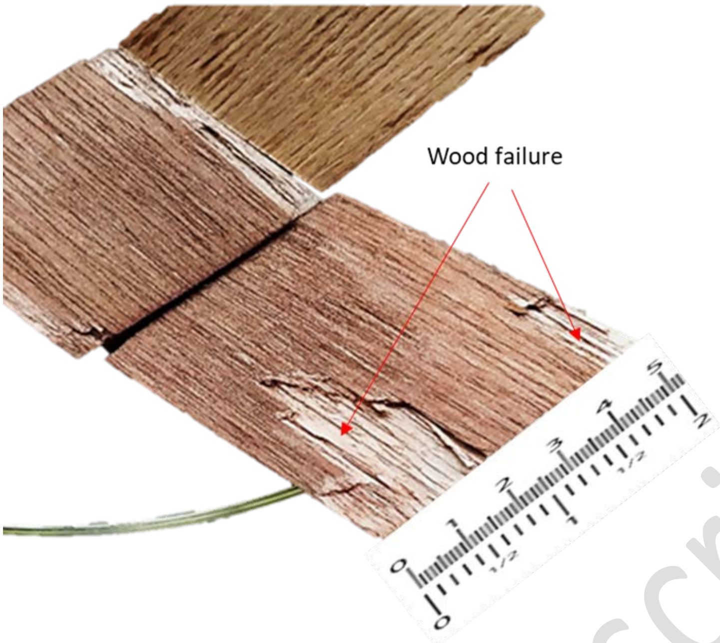

Figure 3: Knife delamination test.

Scanning Electron Microscopic Characterization

The SEM images of the fractured surfaces of the hybrid LVL and plywood are shown in Fig. 4. It was observed that the wood veneers were embedded/bonded in the matrix of WPC. The polymers penetrated deeply inside the voids. During the fabrication process the external heat causes the WPC to melt and flow into the voids present in the veneers created during the peeling, resulting in a strong mechanical interlocking between cells of the wood and the polymer. Images further confirmed that there was no de-bonding observed where veneer surfaces and WPC interacts. Chang *et al*. (2017) studied interfacial bonding mechanism of poplar plywood bonded with HDPE films and reported that there was a poor cohesion between the HDPE and the wood cells which was attributed to their poor compatibility. However, such phenomenon was not observed in the SEM images and there was no delamination at the interphase of WPC and veneer. This suggests that the coupling agent added in the WPC may enhance the adherence of the polymer matrix to the wood veneer substrate which results in an excellent bonding strength of the final product. Also, addition of coupling agent during the fabrication of WPC's plays a vital role in enhancing the wood – polymer interaction resulting in satisfactory adhesion between fibre and polymer matrix (Chauhan *et al*. 2016; Karmarkar *et al*. 2007; Nandi *et al*. 2013; Poletto 2017). The thick layer of the WPC imparts better performance due to presence of cross- linked/ polymerized structures, providing superior resisting against failures. The polymer flow in the pores and crevices present in the veneer would also result in mechanical locking/attachment of WPC with veneer. The overall volume of polymer and wood present in material can significantly affect physical and mechanical properties of the hybrid composites (Lustosa *et al.* 2015).

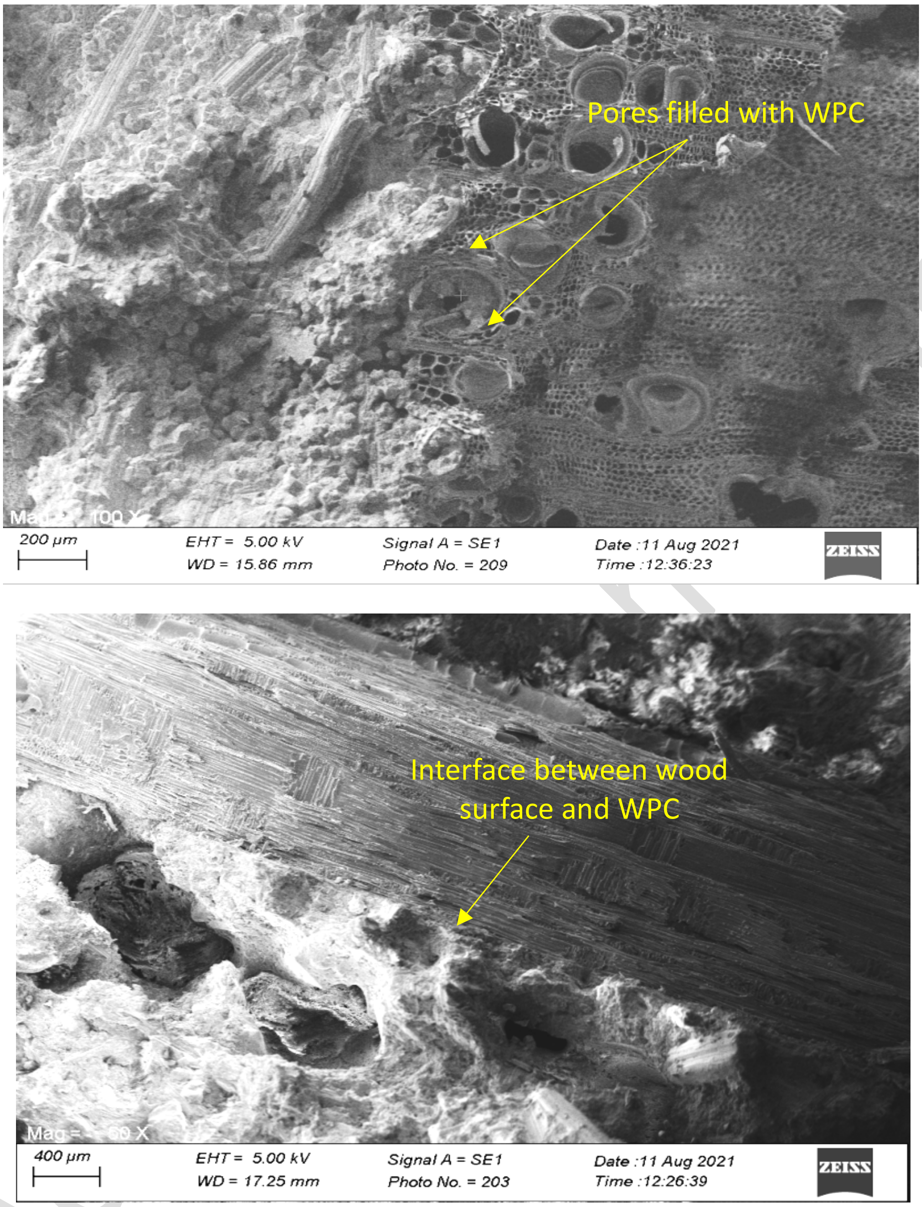

Figure 4: SEM micrographs showing bonding of thermoplastic polymer (up) penetration of WPC in pores of veneer (down) bonding interface between wood veneer and WPC.

Heat Deflection test

The heat deflection observed with respect to change in temperature for polypropylene, WPC sheet and hybrid composite is represented in Fig. 5. HDT test result revealed that a deflection of 0,25 mm was observed when temperature was 120 ºC, 137,8 ºC and 195,2 ºC for PP, WPC sheet and hybrid composite specimens, respectively. Reinforcing polymer with natural fibers is known to result in higher HDT indicating increased stiffness at higher temperatures. Rojanathavorn *et al*. (2014) studied HDT of WPC manufactured using PP and HDPE polymers, reinforced with Ironwood (*Xylia xylocarpa*) at different fibre content loading and reported that with increase in wood content resulted in improvement of thermal stability of WPC. HDT of both plywood and LVL configuration composites were similar. The higher heat deflection temperature of hybrid composites can be attributed to presence of wood veneers which restrict the conduction of heat through the material as compared to WPC specimens. WPC used as a binding agent along with presence of veneer can result in improved thermal stability when exposed to higher temperature as compared to composites bonded using pure polymers. The deflection of hybrid composite with increased temperature also suggest that such plywood or LVL can potentially be thermoformed at elevated temperatures which is not observed in conventional plywood/LVL.

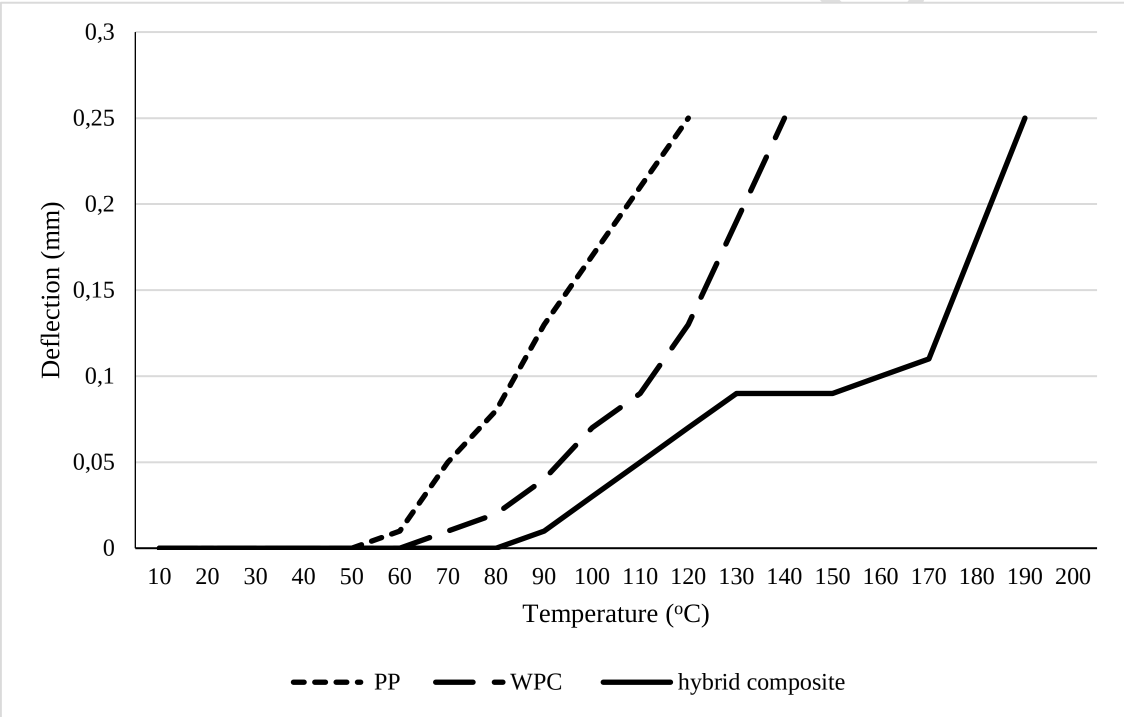

Figure 5: Heat deflection curve of WPC and hybrid composite.

CONCLUSIONS

The present study investigates physical, mechanical and bonding properties of two wood veneer based composites namely LVL and plywood fabricated using veneers of *Melia dubia* wood, bonded using WPC sheet containing 40 % bamboo particles embedded in the matrix of polypropylene. The results revealed that there was no statistically significant difference in physical properties namely density and water absorption, and mechanical properties namely flexural strength, tensile strength and glue shear strength in hybrid composites with LVL and plywood configurations which was attributed to the relative properties of WPC and its comparative proportion in the hybrid composites. The modulus of elasticity, compressive strength and internal bond strength of the LVL were found to be higher than plywood. The SEM micrographs revealed that the polymer penetrates deeply in the voids of wood veneers, resulting in a strong mechanical interlocking responsible for excellent mechanical and bonding performance of the composites. Overall the study indicated that WPC can effectively be used as a binding element for wood veneers resulting in novel hybrid composites.

AUTHORSHIP CONTRIBUTIONS

S. A.: Investigation, Methodology, Writing-Original Draft. **S. C.:** Conceptualization, Supervision, Writing- Review and Editing. **R. K.:** Supervision, resources, Writing-Review and Editing. **B. K.:** Investigation.

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CONFLICT OF INTEREST

On behalf of all authors, the corresponding author states that there is no conflict of interest.

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