

PREPARATION OF PLYWOOD PANELS USING WASTE MILK POUCHES AS AN ADHESIVE

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ABSTRACT

Recycling of single use plastic like milk pouches, oil pouches, etc. for high value products can have significant environmental, social and economic implications. The present study focuses on the utilization of waste milk pouches made out of low-density polyethylene as the bonding agent in preparation of plywood panels. Plywood panels with varying content of polymeric content (milk pouch layers) were prepared and evaluated for physical and mechanical properties. The results indicated that with increase in the dosage of milk-pouch content, both physical as well as mechanical properties of the plywood improved substantially. There was nearly three folds difference in modulus of rupture of panel prepared with 80 g/m² and 310 g/m² polymer content. Similarly, tensile strength, modulus of elasticity also increased significantly with increase in polymer content. The glue shear strength, which is very important for plywood, was found to be influenced by the quantity of polymer. Glue shear strength for panel with 80 g/m² polymer was 0,75 MPa which increased to 1,90 MPa in panel with 310 g/m² polymer content. It was observed that for strong bonding it is essential that the quantity of polymer should be sufficient enough to penetrate into the pores of wood creating a mechanical interlocking of veneers. The study opens up the scope of recycling of waste milk pouches in the preparation of formaldehyde free plywood.

Keywords: Composites, milk-pouches, plywood, polymer, recycling.

INTRODUCTION

Plywood is known to have several favourable features such as increased dimension stability, uniformity, large size with improved stress distribution properties and cost effectiveness. It is one of the most used wood based composite materials (Tenorio *et al.* 2011). Conventionally, plywood is manufactured using thermoset resins such as phenol formaldehyde (PF), urea formaldehyde (UF), melamine formaldehyde (MF), polyurethane (PU), etc. Among these, PF and MF adhesives are the most used in commercial plywood production. Although these adhesives provide good bonding in plywood but the chemicals particularly formaldehyde used in the preparation of these adhesives is known to be toxic. A high dosage of resin i.e. 20 % of total plywood mass may result in concerning formaldehyde emission (Bekhta and Sedliačik 2019). Formaldehyde emission is known to cause eye irritation, sickness syndrome among workers and strongly affects respiratory and nervous system (Jang *et al.* 2011, Mäkinen *et al.* 1999). It is one the main cancer-causing agents as reclassified as carcinogenic to human by the International Agency for Research on Cancer. To overcome this issue, many researchers have formulated formaldehyde free adhesive based on soya flour (Jang *et al.* 2011), starch, palm, etc. (Bono *et al.* 2010). Thermoplastic resins have also been in trend nowadays as a formaldehyde free adhesive, with different types of thermoplastic resins being used for laminated veneer lumber (LVL) and plywood production (Bekhta *et al.* 2020). Widely used thermoplastic resins include high density polyethylene (HDPE)

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(Chang *et al.* 2018, De Barros Lustosa *et al.* 2015, Fang *et al.* 2013, Fang *et al.* 2014, Fang *et al.* 2017, Tang *et al.* 2011) polystyrene (PS) (Borysiuk *et al.* 2010, Demirkir *et al.* 2017), low-density polyethylene (LDPE) (Follrich *et al.* 2006, Cui *et al.* 2010, Ibragimov *et al.* 2017), polypropylene (PP) (Kajaks *et al.* 2012, Song *et al.* 2016, Song *et al.* 2017) and polyvinyl chloride (PVC) (Matuana *et al.* 1998, Song *et al.* 2017).

Utilization of thermoplastic as a potential adhesive in plywood opens up an opportunity to recycle waste thermoplastic material. With the abundant usage, the disposal of plastic is one of the major concerns for the environment due to its slow degradation. Among different thermoplastics, polyethylene terephthalate (PET) and polyethylene (both HDPE and LDPE) are the major plastic waste generators, contributes to about 75 % of plastic waste in India (CPCB 2015). Used and disposed PET plastic is easy to collect, is relatively priced higher for the rag-picker and nearly 90 % is recycled. However, PE plastic specially materials like post consumption empty milk pouches, oil pouches, etc., which are daily household waste in most of the developing nations, are recycled little (Aryan *et al.* 2019). This particular type of waste tends to clog sewage, drainage, etc. resulting in urban problems during rainy seasons. Otherwise also, if left alone in landfills and open lands, these waste plastics gets disintegrated into micro-plastics over a long period contributing to major environmental issues by entering the food web (Chamas *et al.* 2020). Literature suggests that milk pouches only accounts to a total of 74 % of total PE waste while the rest comes from PE shopping bags (Aryan *et al.* 2019). Recycling and utilization of such waste for other products can have substantial implications in dealing with the problems associated with such wastes (Aranda-García *et al.* 2020). This waste material can effectively be used as an adhesive for mainly plywood which can also address the issues of formaldehyde emission from such panels along-with the recycling of waste material.

This study focuses on utilization of waste milk pouch as a binding material in preparation of plywood panels. Amid alarming environmental issues, PE may be utilised as binding agent in production of plywood thereby increasing its recyclability and helping in source reduction. In the present paper, one litre LDPE milk pouches waste collected from household were layered in between veneers and hot pressed to prepare formaldehyde free plastic bonded plywood (PBP). Besides removing major plastic waste, it will also contribute in safeguarding environment from formaldehyde emission.

MATERIALS AND METHODS

Rotary peeled 2 mm \pm 0,5 mm thick veneers of *Melia dubia* wood were sourced from Indian Plywood Industries Research and Training Institute, Bengaluru. The species is commonly used in manufacturing of plywood panels. The average moisture content of the veneers was 8 % \pm 2 % at the time of panel preparation. Post-consumer waste milk pouches of 40 and 60 micrometre thickness and density of 0,92 g/cm³ (LDPE based as specified on the pouches) were collected from the residential areas nearby the laboratory (Figure 1). The pouches were thoroughly cleaned with detergent and water in order to remove dirt, oil and fat content of milk and were finally dried in oven at 50 °C to remove the moisture.

Three-layer plywood panels of dimension 300 mm \times 300 mm \times 4,8 mm were prepared with varying proportion of LDPE films amounting to polymer content of 80 g/m², 105 g/m², 210 g/m² and 310 g/m² between the two veneers. The arrangement of veneer was such that the core layer grain direction was perpendicular to face veneer grain i.e. criss-cross arrangement. In between two adjacent layers of wood veneers, milk pouches in the form of film were placed without any chemical additives. The panel with 80 g/m² polymer content was prepared with half-litre milk pouches of dimensions 150 mm \times 120 mm. These pouches were having polymer film thick-ness of 40 micrometre and each pouch weighed about 1,45 g. Whereas, when one-litre capacity milk pouches (dimensions - 220 mm \times 150 mm; weight - 3,70 g) having plastic layer thickness of 60 micrometre were used in single layer, double layer and triple layer, it amounted to 105 g/m², 210 g/m² and 310 g/m² of plastic con-tent between the veneers for 300 mm \times 300 mm sized panels. The assembly of veneers and LDPE sheets was placed in a preheated hydraulic hot press of 500 tons capacity maintained at 150 °C - 155 °C (Figure 2). The composites were pressed at a specific pressure of 12 kg/cm² for 10 minutes. The panels were removed from the hot press and placed in a cold press at a pressure of 30 kg/cm² to avoid warping in the panel due to differential cooling of polymer and wood veneer. Though the melting point of LDPE ranges from 105 °C -110 °C (Alkan *et al.* 2018), for the better flow of polymer in the pores of wood veneers to achieve good bonding between the ve-neers, the temperature was kept much higher than its melting point. Higher temperature is also required due to low thermal conductivity of wood. Keeping hot press temperature much higher than the polymer melt tempera-ture has been suggested by other researchers for such panels (Bekhta and Sedliačik 2019, Fang *et al.* 2013).



Figure 1: Waste milk pouches.

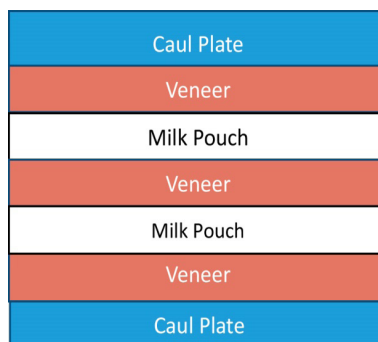


Figure 2: Assembly of polymer bonded plywood.

Physical and mechanical properties

Five panels of plywood were manufactured at each polymer content levels. The prepared panels were conditioned at $23\text{ }^{\circ}\text{C} \pm 2\text{ }^{\circ}\text{C}$ and $65\% \pm 5\%$ relative humidity for seven days. Thereafter, test specimens were extracted from the panels. The panels were tested for various physical properties namely density, adhesion of plies (Knife test), water absorption (WA), thickness swelling (TS) and volumetric swelling (VS). WA and swelling tests were carried out for 2 hours and 24 hours duration. Density was calculated by measuring the mass and volume (volume by displacement method) as per IS 1734 (1983). Percentage of TS and WA were calculated from the difference between initial and final reading after 2 hour and 24 hours (Parthiban *et al.* 2019) as per IS 2380 (1977) using Equation 1 and Equation 2.

$$TS \% = \left(\frac{T_f - T_i}{T_i} \right) \times 100 \quad (1)$$

$$WA \% = \left(\frac{W_f - W_i}{W_i} \right) \times 100 \quad (2)$$

Where T_i and T_f are the initial and final thickness (in mm) respectively, and W_i and W_f are the initial and final weight (in grams), respectively for the specific period of time. The adhesion strength between the veneers was tested with knife test or delamination test after 2 hours of boiling in water as per IS 1734 (1983).

Mechanical properties namely modulus of rupture (MOR) and modulus of elasticity (MOE) were tested as per ASTM D790-15 (2016). Tensile strength was evaluated as per ASTM D638-14 (2016) and glue shear strength (GSS) was evaluated using IS 1734 (1983). Shimadzu make universal testing machine (Model autograph-AG, 10 kN capacity) was used for testing mechanical properties. Since the dimensions of the prepared panels were only 300 mm × 300 mm, it was not possible to extract samples as per Indian standards for testing of plywood panels (IS 1734 (1983)) and therefore the composite samples were tested with procedure prescribed for reinforced thermoplastic composites. Sample dimension for MOE and MOR determination was 100 mm × 12 mm × 4,8 mm. The span length for three-point bending test was as at least 14 times the thickness of the sample and the loading rate was 0,6mm/min. For tensile strength, the span length was 100 mm and the loading rate was 1 mm/min.

For glue shear strength (GSS) test, samples were tested in dry condition for the bond class 1 (EN 314-2). The samples were prepared by making saw cut up to glue line as shown in Figure 3. The grain direction of the ply between the glue-line under test is perpendicular to the length of the test specimen. The loading rate was 0,6 mm/min. For each test, five replicates were tested. The adhesion phenomenon was also analysed by optical microscopy Nikon and model no. C-W 10 X B/22.

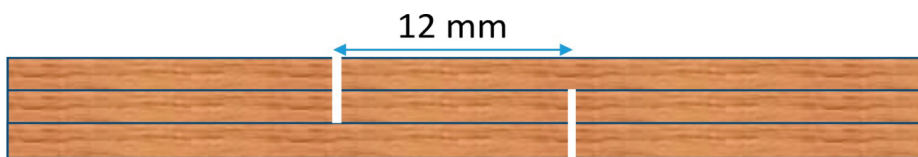


Figure 3: Schematic diagram for saw-cut in plywood panel for glue-shear strength.

The data was analysed using SPSS software for statistically significant differences in properties with respect to polymer content. Duncan test was performed to identify significantly different group for each property.

RESULTS AND DISCUSSION

Physical properties of the prepared plastic bonded plywood (PBP) at varying proportion of polymer content are given in Table 1. Density of the plywood panels was ranging from 0,53 g/cm³ to 0,64 g/cm³ which is much higher than the density of *Melia dubia* wood. The air dry density of *Melia dubia* is reported to be between the range of 0,42 g/cm³ to 0,52 g/cm³ (Khali *et al.* 2017, Saravanan *et al.* 2013). The higher density of the panel is due to densification of wood attributed to compression during hot-press and also due to presence of polymer. The density of PBP was found to increase with the polymer content in the panel. The density of panels with 80 g/m² polymer dosage was found to be 0,53 g/cm³ ± 0,003 g/cm³ and for 310 g/m² polymer dosage it was 0,64 g/cm³ ± 0,02 g/cm³. The increase in density of composite with increasing polymer dosage is attributed to the relatively higher density of LDPE (0,92 g/cm³) as compared to wood.

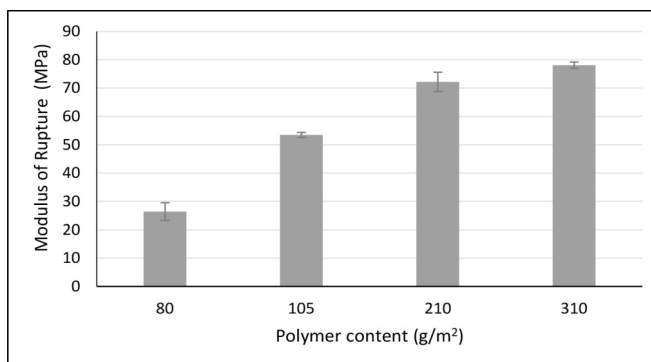
Table 1: Physical properties of plywood with different LDPE content. Values in parenthesis is standard deviation.

Properties	Polymer content			
	80 g/m ²	105 g/m ²	210 g/m ²	310 g/m ²
Density (g/cm ³)	0,53(0,003)	0,57(0,03)	0,61(0,07)	0,64(0,02)
Water absorption - 2 h (%)	21,54(1,36)	17,40(2,20)	15,72(2,90)	14,68(2,22)
Water absorption - 24 h (%)	55,33(2,27)	52,18(5,80)	43,39(5,70)	40,23(3,13)
Volumetric swelling - 2 h (%)	5,18(1,96)	4,40(0,61)	3,95(0,63)	2,94(2,13)
Volumetric swelling - 24 h (%)	7,61(1,12)	7,01(0,20)	6,26(0,67)	5,84(1,35)
Thickness swelling - 2 h (%)	4,48(1,61)	3,88(0,45)	3,19(0,69)	2,8(2,32)
Thickness swelling - 24 h (%)	6,09(0,84)	6,04(0,30)	5,52(0,86)	5,35(1,44)

WA (%) of PBP after 2 hours and 24 hours of soaking in water for 80 g/m² was recorded to be 21,54 % and 55,33 % respectively. Panels with higher polymer content absorbed relatively less water. After 24 hours of WA, PBP with 310 g/m² polymer content absorbed 15 % less water as compared to PBP with 80 g/m² polymer content. TS on WA of PBP after 2 hours of soaking was inversely proportion to the dose of polymer as the TS for PBP with 310 g/m² polymer content was 37,5 % lower than in PB with for 80 g/m² polymer content. The same trend was observed for 24 hours test however the swelling in boards with 80 g/m² was only 12,5 % higher than observed in boards with 310 g/m² polymer content. The reduced WA and TS on increasing polymer loading may be attributed to the slower water absorption rate by the core veneer due to its encapsulation by the polymer film which acts as a barrier. However, long term absorption may lead to similar TS irrespective of polymer loading. This is also evident from the large difference in WA and swelling properties after 2 hours test and the difference tends to reduce after 24 hours test in boards prepared with two extreme polymer loadings. De Barros Lustosa *et al.* (2015) reported 3,21 % TS in LVL panel prepared with *Trattinnickia burserifolia* veneer and HDPE at 150 g/m² polymer loading and 2,82 % in panel with 350 g/m² polymer loading however after 24 hours of soaking TS for both 150 g/m² and 350 g/m² polymer loading was similar. Similarly, Bekhta and Sedliačik (2019) prepared plywood from *Alnus gultinosa* veneer with HDPE and found the WA range from 38,86 % to 44,58 % for 2 hours which is much high compare to PBP prepared in this study. Knife test carried out after two hours of soaking in boiling water revealed complete delamination at 80 g/m² polymer content. However, at higher polymer levels bonding was intact without any delamination and wood failure was observed during the test indicating strong bonding between the veneers.

Mechanical properties

Mechanical properties of the composite material are very important as it determines their suitability for different applications. The prepared panels were tested for MOR, MOE, tensile strength and GSS. The mechanical properties of panels prepared with different polymer content are shown in Figure 4, Figure 5, Figure 6 and Figure 7.

**Figure 4:** MOR of plywood panels at varying polymer content.

There was an increasing trend in MOR as the dose of LDPE was increased from 80 g/m² to 310 g/m² (Figure 4). During the processing, some fractures are formed in veneers and on increasing the doses the molten HDPE is expected to penetrate inside the pores and repair the veneer contributing to the enhanced mechanical properties. MOR of panels with 80 g/m² polymer content was observed to be only 26,41 MPa, as the dose of polymer was not enough to form the bond between veneers. With the increase in polymer content, the strength value gradually increased and MOR value at 310 g/m² polymer content was 78,10 MPa which was nearly three folds higher than at 80 g/m² polymer content. The strength of the panels prepared in this study with higher polymer content (> 80 g/m²) were higher than the conventional plywood prepared using veneers of the same species and UF as the adhesive, which was reported to be in the range of 48,66 MPa to 63,9 MPa (Khali *et al.* 2017). MOR of the plywood depends on the strength of the individual layer and adhesion strength. At low polymer content, adhesion strength between the veneers is expected to be poor resulting in the failure of the core veneer as the MOR of *Melia dubia* wood across the grain was 14,86 MPa as against 81,95 MPa along the grain. With the improved adhesion at higher polymer content, the MOR of plywood was close to that of solid-wood MOR which suggests that the core veneer had little influence on flexural strength of the plywood. Fang *et al.* (2017) observed MOR for poplar plywood bonded with HDPE film of 0,06 mm thickness to be 73,34 MPa. In an another study, Fang *et al.* (2013) prepared poplar plywood bonded with varying doses of HDPE ranging from 61,6 to 246 g/m² and found that MOR of the panel increased with increasing film dosage up to 185 g/m² and thereafter stabilized which is in congruence with the results obtained in this study where after polymer loading of 210 g/m² there was very little change in MOR. MOR values at 210 and 310 g/m² polymer content were not significantly different.

MOE of the composites did not exhibit any significant variation with polymer content (Figure 5). For 80 g/m² doses of LDPE milk pouches, MOE was observed to be 8642 MPa ± 526 MPa and on increasing the doses to 105 g/m², 210 g/m² and 310 g/m² the value of MOE was 8874,9 MPa ± 870 MPa, 8523,6 MPa ± 247 MPa and 9022,95 MPa ± 891 MPa, respectively. The uniform MOE of composites irrespective of polymer content indicates that interfacial adhesion is not influencing MOE. This is mainly attributed to the fact that MOE of a laminate mainly depends on the corresponding properties of each of the plies and their relative thickness as it follows the law of mixture (Chauhan *et al.* 2005).

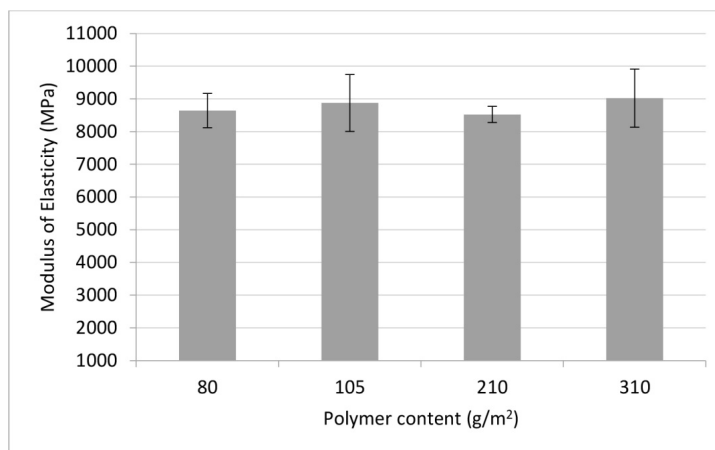


Figure 5: MOE of plywood panels at varying polymer content.

Tensile strength is generally considered to be an indicator of interfacial strength. In case of poor interfacial adhesion between the veneers, the core veneer is expected to fail very quickly as the tensile strength of wood across the grain is extremely poor. This was evident from the result as the tensile strength of the panel was 21,28 MPa at 80 g/m² polymer content (Figure 6). The polymer quantity was not enough to cover the complete surface of the veneers and to penetrate into the pores and vessels during hot pressing resulting in poor strength. As the polymer content was increased, the bonding between the veneers also improved and tensile strength was 48,90 MPa at 310 g/m² loading. However, it appears that 210 g/m² was optimal polymer content layer between the veneers as there was no significant difference (at $\alpha=0,05$; Duncan test) in tensile strength at 210 g/m² and 310 g/m² polymer loading.

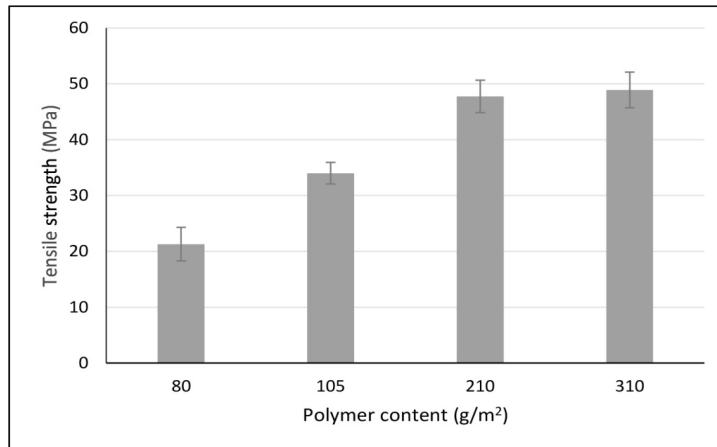


Figure 6: Tensile strength of plywood panels at varying polymer content.

Yue *et al.* (2019) studied the tensile strength of laminated wood plastic composites prepared from maleic anhydride treated poplar veneer and 0,3 mm thick thermoplastic sheet with a ratio of PP and PE at 1:1 and found it to vary from 20,49 MPa to 31,47 MPa with varying wood to plastic ratio (w/w basis) which is lower than the results obtained in this study. In this study untreated veneers were used and also no coupling agent was incorporated in the assembly.

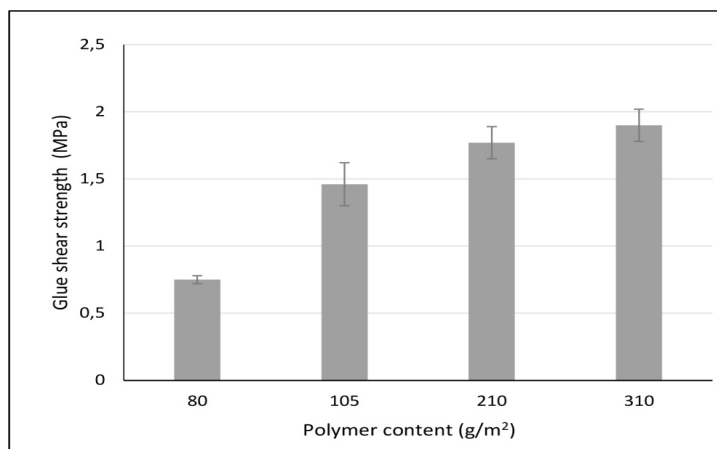


Figure 7: Glue shear strength of plywood panels at varying polymer content.

Further to analyse the adhesion strength, GSS was determined which suggests the adhesion strength between the veneers. GSS was found to be 0,75 MPa \pm 0,028 MPa in panel with 80 g/m² dose of polymer and increased by nearly 150 % at 310 g/m² polymer loading (Figure 7). This was attributed to filling up of larger proportion of voids and lumen space of the two adjacent veneers by polymer during hot pressing as the polymer content is increased forming improved mechanical bond. There was a sharp increase (~100 %) in GSS when polymer content increased from 80 g/m² to 105 g/m² but thereafter the strength increased gradually. The Duncan test revealed that the GSS was statistically differing significantly at each polymer dosage level. Chang *et al.* (2018) assessed the GSS of poplar plywood and found that on increasing the dosage there was increase in maximum penetration depth and effective penetration depth. The variation in other mechanical properties, particularly strength properties, with polymer content is associated with the adhesion strength between the veneers and with improved adhesion at higher polymer content the strength properties also exhibited the improvement on the expected lines.

Optical microscopy

The penetration of polymer was further investigated using optical microscopy for different doses in PBP. It was found that LDPE was able to penetrate and flow into the vessel and porous structure of wood.

Mechanical bond was formed which lead to mechanical interlocking and providing the strength to plywood. It was observed that when the LDPE was 80 g/m² (Figure 8a), polymer is sparsely spread over veneers and there were discontinuous phases of polymer over veneer surface. At lower polymer content, polymer did not penetrate into the vessels and pores resulting in poor interlocking and thus poor mechanical strength. This is further influenced by the poor chemical compatibility of wood with LDPE leading to poor interfacial adhesion between the two veneers. Whereas in panels with higher polymer loading (Figure 8b, Figure 8c, Figure 8d), there is a thick and uniform dispersion of polymer at the glue-line and also the molten polymer has penetrated inside the vessels of the veneer leading to improved mechanical bonding between the two veneers.

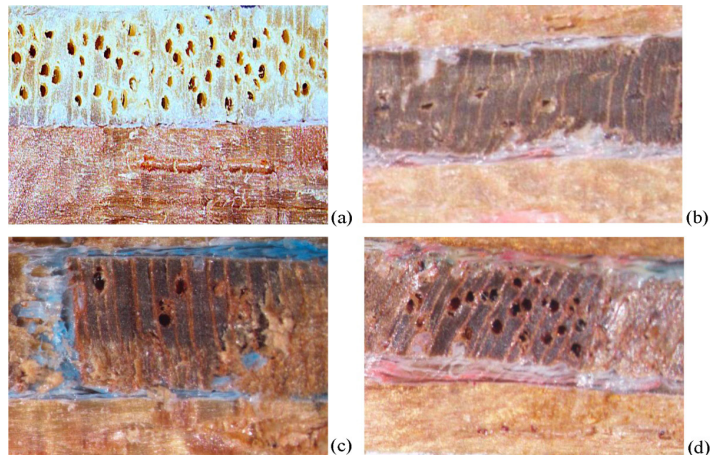


Figure 8: Optical microscopy image of plywood prepared at varying polymer content. (a) 80 g/m², (b) 105 g/m², (c) 210 g/m² and (d) 310 g/m².

CONCLUSIONS

The study demonstrated the suitability of recycling waste milk pouches (LDPE based) as the bonding agent in preparing plywood panels. The formaldehyde free PBP will serve two purposes i.e.us age of waste plastic including milk pouches as well as reduction in formaldehyde emission from the wood panels which is a major concern. For proper melting and embedding of polymer in plywood, it is essential to keep the hot press temperature higher than the polymer and also the quantity of polymer has to be at the optimal level. Low level of polymer content resulted in delamination of veneers on knife test indicating poor bonding. In this study, plywood prepared with 210 g/m² polymer content was found to be the optimum level as there was no significant difference in physical and mechanical properties and the panel possessed good bonding strength without glue-line failure. The improvement in properties is mainly attributed to the mechanical bonding of the veneers due to penetration of hot-molten polymer into wood ultra-structure of the adjacent veneers.

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