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Modification of Urea Formaldehyde Resin with Pyrolytic Oil on Particleboard

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Abstract

Urea formaldehyde resins are widely used in the manufacturing of wood composite and their usage is always combined with release of formaldehyde characterized to be hazardous to health during and after the manufacturing of the products. This study investigates the effectiveness of wood-based adhesive from oil of pyrolysed Triplochiton scleroxylon sawdust for the production of composite board. The wood-derived Pyrolytic Oil (PyO) was blended with Urea Formaldehyde (UF) resin to formed Pyrolytic Oil-Urea Formaldehyde (PyOUF). The obtained PyOUF called Wood-Based Adhesives at four blends and control (UF) viz; 1:1, 1:2, 1:3, 2:1, 1:3 were further employed to prepare the composite board and test for their bonding strength by physical (water absorption-WA and thickness swelling-Th.S) and mechanical properties (modulus of elasticity-MOE, modulus of rupture-MOR, and impact bending-IB). Data obtained was analysed using analysis of variance at α 0.05. The result of analysis of variance conducted on physical properties show significant difference ($p \leq 0.05$) between the WA values obtained when testing the different blending proportion of PyOUF and likewise between 2 and 24 h of immersion. PyOUF had significant effect ($p \leq 0.05$) on Th. S for 24 h but no significant different ($p > 0.05$) for the 2 h period of soaking. The analysis of variance on mechanical properties of the composite board (MOE, MOR, and IB) show significance differences ($p \leq 0.05$) between the strength values obtained when testing the different ratios of PyO with UF. PyO content influenced the properties of the boards and it is evident that PyO can be used in the manufacture of composite board.

Key Words: pyrolysis, pyrolytic oil, urea formaldehyde, wood adhesive, particleboard

Introduction

One of the major challenges associated with wood-based particleboard is the use of formaldehyde resin which causes negative emission into the atmosphere. The contained formaldehyde has been classified as a known *carcinogen* (IARC 2004). The basic raw materials used in the development of Urea-formaldehyde (UF) resin, is derived from petroleum-based resources. However, the sustainable supply of materials has become a serious issue because of the ever-increasing demand for petrochemicals along with continuously declining reserves of petroleum. Hence, over the past few decades, environmental issues and sustainability have become the main drivers of research to produce eco-friendly materials developed from renewable resources.

In this respect, considerable research has been conducted to replace petrol-materials with different kinds of bio-materials extracted from renewable bio-resources (Zhang et al. 2006; Zhang et al. 2013). Amongst various possible alternative bio-materials is pyrolyzed lignocellulosic biomass

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(wood sawdust) called pyrolytic oils. It is a dark-brown organic liquid and also it includes a lot of the organic compounds like phenols, alcohols, ketones, esters, aldehydes, oxygenated hydrocarbons (Goyal et al. 2008; Fuwape et al. 2011; Adegoke and Ayodele 2015). This product can be readily stored, transported, and also used as a chemical feedstock for the production of various industrialized chemicals. The synonyms for pyrolytic oil include bio-oil, pyrolysis oils, pyrolysis liquids, liquefied wood, bio-crude oil (BCO), wood liquids, wood oil, liquid smoke, wood distillates, pyroligneous acid, and liquid wood.

Beside, pyrolyzed lignocellulosic biomass holds an immense potential to replace petro-phenol in various applications. One of these areas is an adhesive application and the development of these new adhesives from renewable natural sources will ensure long term success in the wood composite industry (Antonović et al. 2006). This would cause higher ecological purity of wood-based panels and further expansion of the use of integral wood components. More importantly, the production of pyrolytic oil from wood residues will improve the eco-friendliness of adhesive, reduces waste, increases waste management through optimum utilization of wood residues, provides alternatives to thermosetting adhesives and enhances sustainability.

Therefore, the subject of this research work based on the influence of modification of Urea-formaldehyde (UF) resin with Pyrolytic oil (PyO) to form PyOUF resin on particleboard. Furthermore, this research aimed at evaluating the physical and mechanical properties of the particleboard.

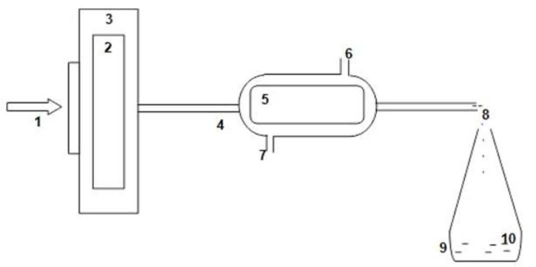

Fig. 1. Schematic diagram of the pyrolyzer experimental set-up (Adegoke et al. 2014).

Materials and Methods

Pyrolysis experiment

Triplochiton scleroxylon (Obeche) sawdust was used for the pyrolysis experiment (Fig. 1). The sawdust was put inside the pyrolytic chamber and then subjected to a predetermined temperature of 500°C. The gases that evolved were distilled in the condenser to form pyrolytic oil which was retained in a conical flask. The oil retained was condensed at 4°C using a reflux system. The above method used for the pyrolysis of Obeche sawdust followed previous work Adegoke et al. (2014).

Preparation PyOUF adhesive

The modification of urea-formaldehyde (UF) resin with pyrolytic oil (PyO) to form PyOUF (Wood-Based Adhesive) was done by blending the different ratios of PyO to UF resin: 1:1, 1:2, 1:3, 2:1, and 3:1 under reflux system on hot plate magnetic stirrer. This process was made at ambient temperature. The above method used follows previous work from Adegoke et al. (2019). Table 1 presents the physical properties of the new adhesive formed.

Production of composite board (particleboard)

The sawdust of Araba (*Ceiba pentandra*) was collected at the point of the saw from a local sawmill within Ibadan, Nigeria. The sawdust was air-dried to eliminate excess free water and prevent degradation by fungi. The samples were later oven-dried at a temperature of 103±2°C for hours until a constant weight was achieved. The sawdust was homogenized using a sieve size of 2 μm. PyOUF was mixed with *Ceiba pentandra* sawdust and spread into a 250 mm× 220

Table 1. Physical properties of PyOUF (Adegoke et al. 2019)

Ratio of PyO:UF	pH	Viscosity (mPa.s)	Solid content (%)	Gel time (s)
1:1	3.38	352.00	51.45	117
1:2	3.39	365.00	52.58	136
1:3	3.43	384.33	53.22	203
2:1	3.53	318.00	47.69	85
3:1	3.77	330.33	48.37	58
UF	3.50	322.00	49.75	25

*Stiasny test, 65%.

mm board mold. The composite board produced follows the pressing parameters in Table 2.

The mixing ratio 1:1, 1:2, 1:3, 2:1, and 3:1 represents the blend of *Ceiba pentandra* sawdust to the PyOUF while particle quantity is the amount (g) of sawdust blended with the PyOUF.

Evaluation of board properties

Before the evaluation of the properties, the boards were conditioned for 5 days at 24°C and 65% relative humidity. The performances of the boards were evaluated according to EN standards. The properties evaluated were on water absorption and thickness swelling (TS) after 2 and 24 h immersion (EN 317 1993). The flexural tests were conducted using a universal testing machine at room temperature according to EN 310 1993. The flexural bending modulus (MOR), bending strength at peak (MOE) and energy to break (IB) were determined by a three-point bending test with the universal testing machine operating with a load cell capacity of 5 kN. A total of five specimens were made and tested.

Data analysis

Data obtained were analyzed statistically. Analysis of variance (ANOVA) was used to test significant difference among different ratio of PyOUF. When the ANOVA indicated a significant difference among different ratios of PyOUF, a comparison of means was conducted, employing the Duncan Multiple Range Test (DMRT) to identify which groups were significantly different at α 0.05.

Results and Discussion

Water absorption

The analysis of variance conducted which shows significant ($p \leq 0.05$) between the WA values obtained when testing the different blending proportion of PyOUF likewise between 2 and 24 h of immersion (Table 3). It is evident from Table 4 that the lowest value of WA (8.23%) obtained from particleboard produced with 3:1 which differed significantly from the other particleboard contained a large proportion of urea in their adhesive blends, and the highest (21.09%) from a panel formed with 2:1, while the values of WA of the other adhesive blend located between the 9.79, 11.80, 14.23, and 19.34% for panel formed with 1:3, 1:1, 1:2 and UF. For the 24 h of immersion, 1:2 had the highest percentage WA while panel formed with 1:3 (36.58%) had the lowest at 14.77%. It appeared that there is an increase in WA as the ratio of PyO decreases in the blends of PyOUF. This decrease is probably due to the formation of condensation products with high bonding potential (Nemli et al. 2003).

The water absorption values for the particleboards produced with UF were lower than those produced with 1:1, 1:2, 1:3, 2:1, and 3:1 (24 h). This fact can be explained by the microstructure formed between the particles and the PyOUF and the ratio of the blending of PyOUF (Fiorelli et al. 2012).

Table 2. Pressing parameters used to manufacture particleboard in the laboratory

Parameter	Value
Replicates	3 boards
Wood particle	Homogenized Sawdust 2 μm
Mixing ratio	1:1, 1:2, 1:3, 2:1, and 3:1
Particle quantity	107, 80, 64, 80, and 64 g
Dimensions	250×220 mm
Total press cycle	4 days
Pressure	3 N/mm^2
Nominal thickness	8 mm
Nominal density	0.7 g/cm^3
Cool temperature	24°C
Cool relative humidity	65%

Table 3. F-Value of various ANOVA table for physical and mechanical properties

Properties	F-value	Sig.
Physical properties (%)		
2 h WA	4.20*	0.05
24 h WA	5.48*	0.03
2 h TS	0.83 ns	0.57
24 h TS	73.38*	0.00
Mechanical properties		
Modulus of Rupture-MOR (N/mm^2)	271.06*	0.00
Modulus of Elasticity-MOE (N/mm^2)	3770.63*	0.00
Impact bending-IB (Nm)	111.17*	0.00

*Significant ($p \leq 0.05$); ns, not significant ($p > 0.05$).

Table 4. Influence of PyOUF on physical and mechanical properties of particleboard

Ratio of PyO:UF	Physical properties				Mechanical properties		
	2 h WA (%)	24 h WA (%)	2 h TS (%)	24 h TS (%)	MOR (N/mm^2)	MOE (N/mm^2)	IB (Nm)
1:1	14.23 (0.65)^{abc}	29.58 (3.26)^{bc}	19.25 (2.12)^{a}	29.40 (4.59)^{bc}	0.64 (0.07)^{a}	77.14 (2.75)^{c}	0.014 (0.004)^{a}
1:2	19.34 (4.30)^{bc}	36.58 (12.17)^{c}	17.45 (1.80)^{a}	25.43 (0.20)^{ab}	0.28 (0.03)^{a}	40.84 (1.31)^{a}	0.004 (0.001)^{a}
1:3	11.80 (6.88)^{ab}	14.77 (2.68)^{a}	20.16 (2.14)^{a}	54.95 (0.41)^{e}	0.28 (0.00)^{a}	22.14 (0.62)^{b}	0.005 (0.001)^{a}
2:1	21.09 (3.20)^{c}	35.21 (3.88)^{c}	15.47 (0.589)^{a}	20.83 (0.52)^{a}	1.07 (0.35)^{b}	133.20 (3.49)^{d}	0.035 (0.011)^{b}
3:1	8.23 (0.22)^{a}	21.22 (1.02)^{ab}	20.33 (5.53)^{a}	38.35 (1.47)^{d}	2.30 (0.12)^{c}	189.05 (4.75)^{e}	0.062 (0.004)^{c}
UF	9.79 (0.62)^{a}	18.42 (0.39)^{ab}	18.33 (2.36)^{a}	33.33 (0.00)^{c}	4.88 (0.03)^{d}	573.55 (9.50)^{f}	0.101 (0.000)^{d}

Means (n=3)±standard deviation in parenthesis.
MOR, modulus of rupture; MOE, modulus of elasticity; IB, impact bending; WA, water absorption; TS, thickness swelling.
Numbers followed by different letters (a-f) in each column are significantly different at the level of $p \leq 0.05$ according to the duncan multiple range test (DMRT).

Thickness swelling

The thickness swelling tests provided information on the bond conditions of the panels after they were immersed in water. The results of variance analysis for thickness swelling (Table 3) show that PyOUF content had a significant effect ($p \leq 0.05$) on this property for 24 h but no significant difference for the period of soaking of 2 h. It appeared that the lowest value (17.45%) of thickness swelling (Table 4) was noticed in panels produced from (1:2), and this value did not differ statistically from other panels (1:1, 1:3, 2:1, 3:1 and UF) as in Table 1. In the case of the 24 h period of immersion, the lowest value (20.83%) was noticed in particleboard produced from: 2:1 as this differs from all other particleboard produced.

However, the thickness as a property was affected negatively as a result of reinforcement of the PyO mix with urea. This may be due to the interference in the curing of the adhesive as explained by Nemli et al. (2003), Polyphenolic extractives can react with formaldehyde even at normal temperature to yield condensation products with high bonding potential, for this reason, the water diffusion and absorption could be decreased, and also probably due to a decrease in the resistance of UF for water in case of immersion for 24 hours. It can further notice and explain that the prolonged soaking in water may result in leaching of the water-soluble substances and hence reduce decay strength.

Generally, based on EN standards, particleboard should have a maximum WA value of 8%. The WA of the particleboard ranged from 8.23 to 21.09% (Table 3). All the panels produced had higher WA values than those allowed by EN 317 standard (max. 8% for 2 h immersion). These high values may be related to the fact that no wax or other hydrophobic substance was used during particleboard manufacture.

Bending modulus

The results of the analysis of variance for bending modulus (modulus of elasticity-MOE) showed, that there are a statistical significance ($p \leq 0.05$) differences between the strength values obtained when testing the different ratios of PyO with UF, also the analysis showed a significant effect of the levels of PyOUF resins content on this property (Table 3).

It is evident from Table 4 that the highest values of MOE regardless of the levels of resin was the particleboard made from UF with the value of 573.55 N/mm^2, followed by 3:1, with bending modulus value of 189.050 N/mm^2, while 1:3 had the lowest bending modulus 22.137N/mm^2.

Bending strength at peak

The analysis of variance in Table 3 shows a significant effect ($p \leq 0.05$) of PyOUF on bending strength at peak-modulus of rupture (MOR) values obtained when testing the different blends. It is evident from (Table 4) that the highest values of MOR regardless of the levels of resin content obtained (4.877 N/mm^2) from UF particleboard and the lowest (0.282 N/mm^2) from particleboard produced from 1:2. It appeared also that there is a decrease in MOR values as the amount of PyO increases in the ratios.

It can be further noticed, that there were no significant differences in bending strength MOR) between 1:1, 1:2, and 1:3 (Table 3). Similar results concerning MOR and MOE values were obtained by Kasir and Al-Zaidbagy (2004) in their study on the utilization of *Eucalyptus camaldulensis* bark extract fortified with UF and Gonçalves et al. (2003) in their study on particleboard made with UF modified with bark extract from mimosa (*Mimosa caesalpiniaefolia*). Both found a decrease in MOR with an increase in bark extract in the resin mix.

Bhagat and Verma (2013) reported that the flexural properties of composites depend critically on the microstructure of the composite and the interfacial bonding between the reinforcement and the matrix. This explains the content in Table 4, that both flexural strength and modulus of the six composites boards increased with an increase in reinforcement content (PyOUF) at all ratios. This was due to the strong interfacial adhesion/bonding between the particles and the matrix which enhances load transfer (Swain 2013). While the later decrease in flexural strength and modulus might be due to agglomerate formation at higher concentrations of the reinforcement of the two composites which is also observed in the tensile behavior.

Energy to break

Energy break -Impact bending (IB) is an important property that indicates overall material toughness. However, the matrix–fiber interfacial bonding, and the properties of both matrix and fiber of the particleboard produced. The results of variance analysis for impact bending (energy to break) showed, that the ratio of PyO: UF was significant ($p \leq 0.05$) in their effects on impact bending (Table 3). In Table 4 presented, it appeared that the highest values of impact bending (0.101 Nm) obtained from a panel made of UF followed by panels formed with 3:1 and 2:1 which had a value of (0.062 Nm and 0.035 Nm) respectively and 1:1, 1:2, and 1:3 did not differ from each other as shown in (Table 4), while the lowest value (0.004 Nm) obtained from panels formed with 1:2. The result has denoted that when the composites undergo a sudden force, the impact energy is dissipated by the combination of fiber pullouts, fiber fracture, and matrix.

Conclusions

In this study, the various blending of PyOUF was used to make particleboard. Based on the preliminary results of this study, the modification made on UF by PyO content influenced the properties of the boards hence PyOUF can be used in the manufacture of composite board. It is suggested that PyOUF levels should be at 1:1, 1:2, 2:1, and 3:1 in the panels if these are to maintain their desirable properties hence, it had higher property values than those allowed by EN standard.

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