

Alternative Functionality of Wood-waste through Applied Binding Mechanism of Urea-Formaldehyde Resin

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ABSTRACT

Sawdust, the major by-products of sawmills and an abundant wood-waste of ≤ 2 mm particle size was used to produce particleboards through the binding mechanism of urea-formaldehyde, to minimize its potentials of posing environmental challenges, and study its suitability with urea-formaldehyde resin for the production of less expensive particleboards of acceptable standard. A known mass of the sawdust (3g – 8g) was first blended with 20cm³ of molten paraffin wax, and then with 30cm³ of urea-formaldehyde resin. The blend was molded into board under electrically heated hydraulic press at 150°C and 10 tons for 4 min. The physico-mechanical properties of the particleboard, including water absorption, thickness swell, density, tensile strength, modulus of elasticity, specific strength and hardness, were investigated using the conventional methods, and were found satisfactory with reference to the Bureau of Indian Standards and Australian standards for thin particleboards. The properties of the particleboards were dependent on the ratio of the sawdust to the urea-formaldehyde resin. The boards are easily produced and cost effective, and should be an inexpensive alternative for wood panel producers and users because, the end-use application of particleboard is subject to the properties, ease of preparation and cost.

Key words: Particleboard, urea-formaldehyde, binding, sawdust, properties.

INTRODUCTION

Wood is the hard, tough substance that forms the trunk of trees, and a 3-dimensional polymer composite made up mainly of 50% cellulose and 20 – 30% lignin, used by early humans to make tools, shelter, fire, and other necessities, and has been widely used as construction material sequel to its physical strength, and low processing cost. Nevertheless, it has some inherent troublesome properties like change in dimension with time due to environmental modulation and structural degradation which result in loss of strength (Rowell, 1983; Deka and Saikia, 1999); a problem that led to the invention of composite lumber or wood panel product such as particleboards, fiberboards, wafer boards and oriented strand boards, which possess a wide range of beneficial properties than standard lumber.

Since the invention of particleboard in 1940s by Max Himmelheber, it has emerged as versatile substituent for wood in many applications, and its advantages over natural solid wood panels lie on the facts that: it is fairly inexpensive to manufacture which translates

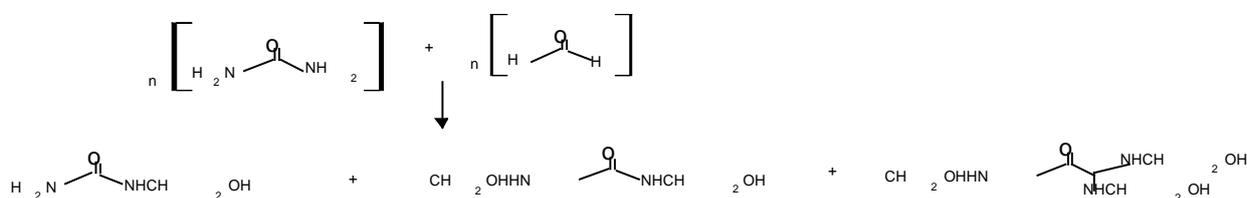
into lower cost for the consumers; it is available in large flat sheets; and easily decorated in different styles with laminated overlays. The applicability of a particleboard largely depends on its properties which include water resistivity/water adsorption, thickness swell, density, tensile strength, hardness, flexural strength, impact strength, nail resistance, screw-withdrawal, surface soundness, and formaldehyde emission. These properties in-turn depend on the processing variables such as pressing time, pressure and temperature, as well as the manufacturing parameters such as the type of binder (resin), filler (eg. sawdust), and other additives used. In particleboard industry, particleboard is basically manufactured by mixing wood particles or other agricultural wastes with a synthetic resin or other suitable binder, and forming the mix into a sheet under heat and pressure. The wood particles (wood-wastes) are used as the primary lignocellulose materials, and sawdust was recommended due to its relatively low specific weight and abundance as a cheap by-product, and unless reprocessed into boards, burned in a sawdust burner or used to make heat for other

operations, it can present a hazard in manufacturing industries especially in terms of its inflammability and potential to collect in piles and add harmful leachates into local water systems (Peter et al. 2002), creating an environmental hazard.

The three most commonly used resins (binder) in wood composite industry are formaldehyde-based resins, including urea-formaldehyde, phenol-formaldehyde and melamine-urea-formaldehyde (Mamza et al. 2014), but with the safety concern on formaldehyde (Keith and Telliand, 1979), several efforts are being made to reduce or replace formaldehyde content in the resin formulations (Nihat et al. 2002; Mozaffer et al. 2004; Yoosup et al. 2008). Thus, there have been increased interests in researches geared towards modification of the common binders and development of new ones to conform to

standards and guidelines of the environment protection agencies and as well maximize its economic value.

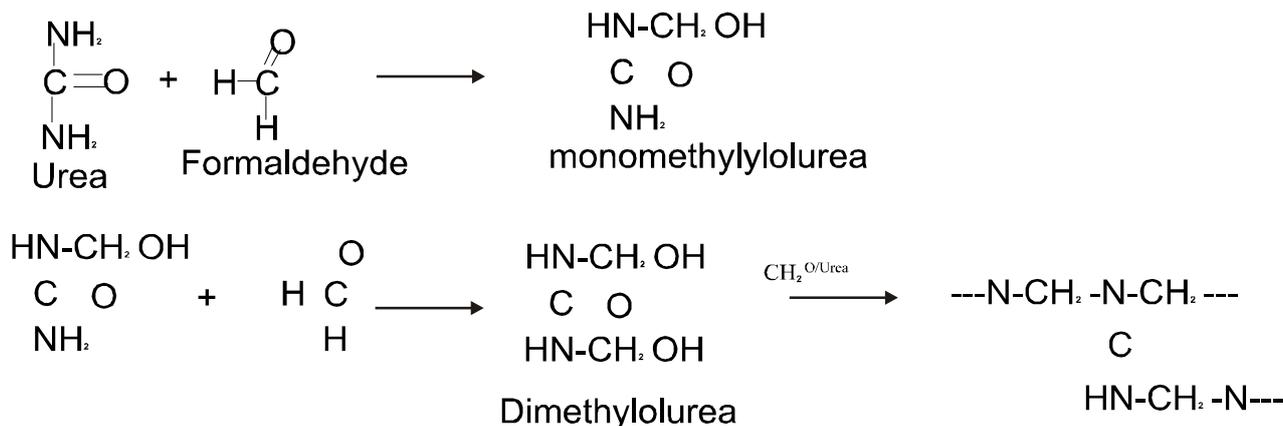
Urea-formaldehyde is composed of molecules that cross-link into clear, hard plastics. It is formed from the reaction between urea (H_2NCONH_2) and formaldehyde (CH_2O). Two main steps are recognized in the reaction of formaldehyde with amino compounds such as urea to form useful amino resins and the success of producing and using the resins largely depends on the precise control of these two chemical reactions (Gordon et al. 1966). The first step, known as methylation or hydroxymethylation, involves simple addition of formaldehyde to introduce the hydroxymethyl group. This reaction is in reality a series of reactions that lead to the formation of mono-, di-, and trimethylolurea, as illustrated below.



The addition of formaldehyde to urea can take place over the entire pH range. However, the reaction rate is dependent on the pH, and the rate for the addition of formaldehyde to successively form one, two, and three methyl groups has been estimated to be in the ratio of 9:3:1 respectively (Nimz, 1983). The exact ratio of course, is dependent on the reaction

conditions employed in the addition reaction.

The second step, referred to as methylene bridge formation, polymerization, resinification, or simply cure, is a condensation reaction that involves the linking together of monomer units with the liberation of water to form a dimer, a polymer chain, or a vast network as shown below.



The hydroxymethylation in alkaline conditions at pH 8.2–8.3 was reported a fast reaction and the chosen conditions lead to a steady equilibrium state characteristic of a concrete reaction mixture (Peep et al. 2006). Although it is often said that the purpose of hydroxymethylation is to obtain 1, 3-bishydroxymethylurea, quite a complicated mixture consisting of mono, di, and tri-substituted urea compounds (as shown above) are formed. In the presence of excess formaldehyde and alkaline condition, dimethylene ether formation favors condensation mechanism, while in aqueous solution of formaldehyde containing some amount of methanol as the stabilizing component, a certain amount of methoxymethylene groups are formed. Under acidic condition, increase in the molecular weight of urea-formaldehyde resin is thought to be a combination of reactions leading to the formation of : (1) methylene bridges between amido nitrogens by the reaction of methylol and amino groups on reacting molecules, (2) methylene ether linkages by the reaction of two methylol groups, (3) methylene linkages from methylene ether linkages by the splitting out of formaldehyde, and methylene linkages by the reaction of methylol groups spitting out water and formaldehyde in the process.

The objective of this research was to significantly and economically improve the functionality of wood-waste through the manufacture of wood panel products using a cost effective and easily prepared binder and sawdust, thereby promoting alternative source of lumber, and reducing the over dependency on natural solid wood that result in continuous deforestation, which is a practice that militates against natural environmental sustainability.

MATERIALS AND METHODS

Materials

Urea was purchased from H & W Ltd Chadwell Heath Essex England and used as received. Formaldehyde and paraffin wax were used as received. Sodium hydroxide pellet was purchased from NAAFCO scientific supplies Ltd London, and was used to prepare aqueous solution of sodium hydroxide. The sawdust was collected from a sawmill; from which stone,

over sized pieces of wood, and other unwanted objects were removed from the sawdust by handpicking, after which it was sieved (using a sieve of mesh-size 2mm) to obtain sawdust of uniform sizes, and sprayed on a clean mat to dry at ambient condition.

Preparation of the Resin

The urea-formaldehyde was prepared by mixing 255cm³ of 40% aqueous formaldehyde in a 500cm³ beaker with 8cm³ of 1molar solution of NaOH. The mixture was heated and stirred for 15min. 150g of urea crystals was then charged into the mixture, stirred for 15min and the pH maintained at 9-10. The mixture was transferred into a quick-fit round-bottomed flask, refluxed for 20min at 60°C, and then cooled at room temperature.

Formulation of the Particleboard

Exactly 8g of the sawdust was first blended with 20cm³ of molten paraffin wax (as water repellent agent) in a mixer for 5min. 30cm³ of the resin was then added and thoroughly mixed for another 5min. The resin-sawdust blend was sprayed on aluminum foil, wrapped and pressed. These processes were repeated using 7g, 6g, 5g, 4g, and 3g of the sawdust with 20cm³ of paraffin wax and 30cm³ of the resin. Each of the blends was separately molded into board (panel) form by compression molding in an electrically heated hydraulic press at 150°C, 10 tons, and 4min using a metal mold with the dimensions (160mm x 150mm x 3mm). The particleboards formed were cooled at room temperature.

Characterization of the Particleboard

For accurate evaluation of the physical and mechanical properties of the particleboards, the particleboard samples were tested empirically for their wood working properties in accordance with the ASTM specifications for particleboards.

Tensile Strength: This was carried out using Hounsfield tensometer; a tensile testing machine, designed to elongate the sample at a constant rate and continuously and simultaneously measure the instantaneous applied force and the resulting elongations. The

samples were configured in dombel (ie dogbone) shape, and each was fixed-clamped at one end and pulled at constant rate from the other clamped end. The dogbone shape of the sample encouraged its failure at the centre, where the stress was highest, and not at the gripsites where stress concentration may otherwise result to premature failure. The tensile response was calculated in terms of stress-strain.

$$\text{Stress} = \frac{\text{Force}}{\text{Cross sectional area.}}$$

$$\text{Strain} = \frac{\text{Change in length}}{\text{Original length}}$$

Density: The density of the particleboard was obtained by dividing the weight of the particleboard with its calculated volume. The weight was obtained using analytical-weighing balance, while the volume was obtained by multiplying their respective measured length, width, and thickness. The density of the particleboard samples were calculated using: $p = m/v$.

Where p = density (g/cm^3), m = mass of sample (g), and v = volume (cm^3).

Water Absorption: The particleboards were tested for their water absorption properties at room temperature and 65°C . This was carried out by weighing the particleboard samples (26mm length, 16mm width, and 3mm thick) before and after soaking in water for 24h. Two sets of the samples were prepared and soaked in water contained in a beaker and were allowed to stand, one at 65°C and the other at room temperature. They were allowed to stand in their respective conditions for 24h, after which they were removed from water and reweighed. The ratio of the difference in weights to the original weight of the sample expressed as a percentage is the percentage water absorption of the particleboard.

Thickness Swell: Thickness swell test was carried out by measuring the thickness of the samples before and after being soaked in water

for 24h. (One set at room temperature and the other set at 65°C). The ratio of the difference in thickness to the original thickness of the sample expressed as a percentage is the percentage thickness swell of the sample.

RESULTS AND DISCUSSION

Beneficiation and screening of the sawdust as earlier stated gave sawdust of particle size $\leq 2\text{mm}$. This was necessary because the properties and potential uses of various types of particleboard differ greatly with regard to the size and geometry of the particles. Better strength, appearance, and resin economy could be achieved by using more uniform sawdust particles.

Urea-formaldehyde pre-polymer was prepared at 60°C , and pH value of 9-10 to avoid the possible curing of the resin at elevated temperature and acidic medium before it is applied for the particleboard formulation. High temperature ($150\text{-}220^\circ\text{C}$) and acidity have been found as some of the factors that facilitate the cross-linking (curing) processes of the resin. Hence, the pH of the resin was maintained at 9-10, such that the cross-linking could only be facilitated by the elevated temperature during the formulation of the particleboards in a compression moulding machine.

In analysis of the sawdust-resin interactions, continuum mechanics was used because the binding mechanism is basically mechanical, in which there is actual penetration of adhesive into surface pores and crevices. The degree of mechanical behavior depends on the strong binding at the sawdust-resin interface. The volume ratio of the two phases may also influence the properties of the particleboard.

Many of the same parameters with which metals are specified -ie. Tensile strength, modulus of elasticity, and yield, are often used explicitly for polymeric materials (Billmeyer, 1984). Hence, the stress-strain tests were employed for the characterization of the particleboards, and the results of the tests are summarized in table 1.

Table 1: Mechanical properties of the urea-formaldehyde particleboard samples.

S/N	%Resin Composition	Tensile Strength (MPa)	Brinell Hardness	Modulus of Elasticity (MPa)	Specific Strength (N.m/gK)
1	85.7	1.817	0.218	52246	245.54
2	56.6	0.752	0.062	50272	85.45
3	55.5	0.213	0.519		27.66
4	54.5	2.554	0.740	64595	359.27
5	53.6	2.503	0.726	81033	272.07
6	52.6	3.247	0.940		382.00
7	51.7	4.398	1.280	18425	488.67

Tensile test is one of the most common mechanical stress-strain tests used to ascertain several mechanical properties of materials. Seven (7) tensile samples were prepared for the test because the ultimate stress resulting from the large and irreversible deformation is a sample properties rather than material properties, and is strongly influenced by sample defects and processing history. However, their average could be an actual representation of the properties of the particleboard. The tensile samples were configured in dogbone shape with rectangular cross-section. The dogbone configuration was chosen so that during the testing, deformation could be confined to the narrow center region (which has a uniform cross-section along its length), and also reduce the likelihood of fracture at the gripsites of the samples. The tensile testing machine is designed to elongate the sample at a constant rate and to continuously and simultaneously measure the instantaneous applied force with loaded cell, and resulting elongation. The dependency of the load-deformation characteristics on the sample size was minimized by normalizing the geometric factors, load and elongation, to the respective parameters of engineering stress and engineering strain (Davis, 2004). The results of the tensile strength tests, as shown in table 1, indicated that the urea-formaldehyde particleboard samples exhibited good strength with reference to Bureau of Indian Standards/specification. All the samples except the samples of 55.5% and 56.6% resin compositions showed higher values than the Bureau of Indian Standards (BIS, 2005) value - 0.8N/mm². This

implies that the urea-formaldehyde resin imparted good mechanical properties in the particleboard, which could be attributed to the three dimensional bonding effect of the binder, and strong binding force and compaction strength at the resin-sawdust interface. The tensile strength decreased with increase in the resin composition from 51.7% to 55.5%. This may be due to the viscous nature of the resin which could not allow easy penetration into the pores and crevices of the sawdust, hence, allowing the filler-influence to dictate the mechanical behavior of the particleboard samples until above 55.5% resin composition when the resin assumed to be enough to exert its adhesion and mechanical interlocking influence, resulting to the experienced increase in tensile strength.

Tensile strength and hardness are indicators of a material's resistance to plastic deformation. The Brinell hardness of the particleboards followed the same trend as the tensile strength. It decreased with increase in resin compositions from 51.7% to 56.6% and then increased from 56.6% to 85.7% resin composition. The urea-formaldehyde particleboards of 56.6% to 85.7% resin composition exhibited relatively good hardness. This can be attributed to more cohesive and adhesive force exerted by the resin on the sawdust particles, resulting to the greater hardness exhibited. Hence, they should possess relatively better toughness, durability and ability to resist abrasion or surface deformation than those of 51.7% to 55.5% resin compositions.

Specific strength (material's strength

“force per unit area at failure” divided by density) of the particleboards was also determined. Expectedly, it followed the same trend as tensile strength; a decrease in specific strength of the particleboard with increase in the resin composition from 51.7% to 55.5%, and then increased from 56.6% to 85.7%. The increase in specific strength implies increase in weight-to-strength ratio. Therefore, the urea-formaldehyde particleboard samples of 56.6% to 85.7% resin compositions which exhibited good specific strength should be more applicable where weight saving are worth the high cost than the urea-formaldehyde particleboards of 51.7% to 55.5% resin compositions.

On an atomic scale, macroscopic elastic strain is manifested as small changes in the interatomic spacing and the stretching of interatomic bond. As a consequence, the magnitude of the modulus of elasticity is a measure of the resistance to separation of adjacent atoms, that is, the interatomic bonding forces. This modulus is proportional to the slope of the interatomic forces-separation curve at equilibrium spacing (Ralls et al. 1976). On particulate scale, the interstitial spaces allow the stretching of the inter-particle bonds formed by the resin. The results of the test for modulus of elasticity of the urea-formaldehyde particleboards shown in table 1 showed high modulus of elasticity with respect to ANSIA 208.1 - 2009 standards for particleboards. In stress-strain plot for polymers, lower modulus implies that higher energy is required to deform the sample to the point of failure (Joel, 2003). Therefore, the urea-formaldehyde particleboard samples which exhibited high modulus of elasticity should be stiffer than particleboards with low modulus of elasticity. This is as a result of stretching tendency of the inter-particle bonds

formed by the urea-formaldehyde resin, and perhaps the slippage of plastic flow along the crystal plane in the urea-formaldehyde resin.

Density of material connotes its mechanical properties. This may give reasons for the decrease in the tensile strength of the urea-formaldehyde particleboards exhibited by the samples of 51.7% to 55.5% resin compositions. The results, as in table 2, show that the density of the samples decreased from 51.7% to 55.5% resin compositions; same trend as the tensile strength of the samples.

Water absorption and thickness swell tests were carried out at different temperatures to ascertain the water resistivity of the particleboards as well as their dimensional stability in a humid environment and at elevated temperature. The response of a particleboard to humidity is a function of the degree of its water absorption. It is a sample rather than material property, and as such, it depends on the composition and processing history of the sample. The urea-formaldehyde particleboards showed relatively good resistance to moisture at room temperature, as indicated in table 2. However, the moisture resistivity reduced at elevated temperature (65°C). This was not unexpected because, principally condensation polymers are susceptible to degradation due to effect of water, and multiple exposures such as moisture and heat can result in accelerated deterioration (Joel, 2003). Results of the thickness swell on the other hand showed that the degree of thickness swell, which is a measure of the dimensional stability of the particleboards in humid environment, is a function of the composition and the prevailing temperature. It decreased with increase in the percentage composition of the urea-formaldehyde resin in

Table 2: Physical properties of the urea-formaldehyde particleboard samples.

S/N	%Resin Composition	%Water Adsorption (Rm Temp.)	%water Adsorption (65°C)	%Thickness Swell(Rm Temp.)	%Thickness Swell (65°C)	Density (g/cm ³)X 10 ⁻³
1	85.7	47.27	110.71	0	63.4	7.4
2	56.6	25.72	84.00	12.9	54.8	8.8
3	55.5	49.12	97.20	8.1	56.8	7.8
4	54.5	51.39	94.56	9.4	50.0	7.1
5	53.6	29.32	81.16	7.7	60.0	9.2
6	52.6	49.20	71.30	15.2	45.5	8.5
7	51.7	22.51	99.89	18.2	81.8	9.0

CONCLUSION

Sawdust; the major wood-waste from sawmills with the potentials of posing environmental challenges, can be profitably utilized with urea-formaldehyde resin for the production of less expensive particleboards, whose properties is a function of the percentage composition of the components. This implies that the properties of the particleboard depend on the resin-filler ratio. Consequently, the properties can be altered by varying the percentage composition of the components.

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