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Research Article

EVALUATION OF PHYSICAL, MECHANICAL PROPERTIES AND FORMALDEHYDE EMISSION OF THE PLYWOOD MANUFACTURED WITH UF/PMDI HYBRID RESIN AS AN ADHESIVE

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A B S T R A C T

The aim of this study was to evaluate the physical, mechanical properties and formaldehyde emission of the plywood made by using UF /pMDI Hybrid resin. In order to improve the water resistance properties, urea-formaldehyde resin are commonly modified with melamine or phenol. In this study water resistance properties of urea formaldehyde resin was enhanced by using polymeric isocyanate (pMDI) having terminal isocyanate –CNO group appx. 12,5 % with urea –formaldehyde resin . 12mm plywood was manufactured by using uf/pmdi resin developed in the laboratory scale and was taken for its performance study in terms of physical and mechanical properties like water resistance, glue shear strength, MOR,MOE etc. and statistical analysis was performed in order to examine possible feasibility of the UF/pMDI resin for manufacture of boiling water resistance plywood. From the analyzing data results shows that incorporation of 10% of pMDI in UF/pMDI resin significantly increases the water resistance, glue shear strength, MOR, MOE value of the plywood and formaldehyde content of the plyboard reduced up to 30% as compared to control sample tested as per IS:13745. The results of this study demonstrate that the UF/PMDI hybrid resin can be an alternate resin for improving the water resistance properties of the plywood.

INTRODUCTION

The PMDI resins are already proved for manufacturing wood panel products like Particle board, Medium Density Fiberboard etc. But its use as an adhesive for plywood manufacturing is limited due to several reasons. Analyses showed that a 40 % addition of isocyanates to UF resin makes it possible to manufacture plywood with an even better water resistance than that specifi ed in the respective standard binding for MF resin (Pizzi *et al*., 1993).

In contrast, Lei *et al*. (2006) stated that an addition of pMDI to MUPF resin generally did not improve mechanical properties, but it considerably improved the board water resistance. These researchers showed that, when pMDI is added to MUPF resin at as little as 5 % tensile strength of particleboards, after 2 h boiling the water resistance increases from 0.09 to 0.21 N/mm^2.

The presented modification method of amine resins, apart from an increase in water resistance, also contributes to a reduction of costs associated with pMDI use.It was reported that mixing isocyanate with PF resin or UF resin adhesives enables it to react faster with water than with other resin and makes each adhesive's methylol group and --NCO group react to create urethane bonds with relatively strong bond strengths (Pizzi 1983, Pizzi and Walton 1992, Pizzi *et al*. 1993, Pizzi *et al*. 1996).

Chelak and Newman (1993) reported that existing formaldehyde-based resins condense while emitting water to serve their purpose as adhesives, whereas isocyanate reacts with to form a polyurea structure as the backbone of its adhesive bond formation. In this context, it was expected that mixing UF resin with isocyanate prepolymer containing urethane bonds and nonreacted--NCO groups would achieve urea bonds and urethane bonds.

In turn, the use of UF/pMDI hybrid adhesive leads to a reduction of the pMDI share by 15 %, & reduces the costs and improves its water resistance, but only to a limited extent. Plywood produced with this adhesive exhibited good water resistance only after 27 min of boiling instead of 2 h, as specifi ed in the standard for melamine based resin (Mansouri *et al*., 2006). The NCO group in PMDI may react with the hydroxyl group present in wood and the curing starts before the assembly of veneers for hot pressing. (papadopoulors etal. 2002)The PMDI resin may also stick with the caul plates placed above the boards while pressing which creates further difficulties in taking out the plywood (Hanns-immo sach *et al*.1997).

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The reaction of isocyanate with water at room temperature can be made slow, if it is emulsified in a starch solution. The mobility of water molecules was expected to be reduced when they were bound to numerous hydroxyl groups present in starch [Long yu *et al*. 2011].

The aim of this study was to to manufactured plywood by using UF /pMDI as an adhesive as binder and to evaluate the physical, mechanical properties and formaldehyde emission of the plywood.

MATERIALS AND METHOD

Materials

Technical-grade urea granules (99%), formaldehyde (37% HCHO) and aqueous solutions of both formic acid (HCOOH) and sodium hydroxide (NaOH) were used for the synthesis of UF resin.The polymeric isocyanate (pMDI) was Yamandur R-11an isocyanate-terminated Prepolymer containing 12.5 % free isocyanate groups having viscosity 15-25 Cps @30^0C and appearance of dark brown colour emulsified liquid. Wood Veneer used for manufacture of plywood belongs to *Dipterocarpus sp.*(Gurjan) as core and face veneer .

Methods

Preparations of the Urea- formaldehyde resin

230-250 parts by weight of formalin (Formaldehyde content 37%) was charged into resin kettle and made alkaline with 50% sodium hydroxide solution to pH 7.5 -8.0. 100 parts by weight of urea was gradually added to the kettle and stirring started. Stirring continued till the end of the reaction. Temperature was raised by passing steam and then set at 92º±2ºC and kept at this temperature under agitation for 1½ - 2 hours. pH were checked time to time and maintained at 7.5 – 8.0. In the second stage, the pH of the solution was lowered to 5.0 – 5.5 by adding 50% solution of acetic acid and reaction was continued under agitation at the same temperature. The progress of the reaction was followed by measurement of viscosity and water tolerance. For ready result, instead of viscosity, flow time of the reaction mixture was measured in B_4 cup. Water tolerance was a measure of the number of times of weight of water which can be mixed with resin before incipient precipitate is formed. The resin was ready when had a flow time of 14-15 seconds in B_4 flow cup and water tolerance of 3-4 times. The reaction was arrested by raising pH to 7.5 – 8.0 by adding 50% alkali and then resin was cooled to ambient temperature.

Preparation of UF/pMDI resins Hybrid

UF/pMDI resins were prepared in the laboratory by adding polymeric isocyanate (pMDI) with different concentrations starting from 1%, 2.5%, 5%, 7.5% and 10% into the liquid UF resin. Five different above UF/pMDI resins were prepared by adding polymeric isocyanate (pMDI) during cooling of the resin when the temperature was at 60 ± 2^0C . Finally, the resin mixture was agitated at high rpm using a laboratory stirrer until a homogenous solution was obtained and cooled to room temperature. Properties of these UF/pMDI resins were presented in Table 1.

Formulation of adhesive mix

Using synthesized UF/PMDI hybrid resin plywood adhesive mix was formulated by mixing synthetic resin, Ammonium chloride(NH_4Cl) as hardener, Liq. Ammonia (NH_3) as buffer and wheat flour as extender to enhance the rheological properties of the adhesive. The flow time of the adhesive mix was standardized under stirring with a speed regulated stirrer. (Table No-3). The mixing was continued for 30 minutes until a homogeneous mixture was obtained. Viscosity of the glue after mixing was taken in B_6 Cup (IS:3944,1982) .

Plywood manufacturing

Plywood with a size of 300mm×300mm×4mm was manufactured by using *Dipterocarpus sp.*(Gurjan) as core and face veneer and UF/B-pMDI resins of different mixing concentration of polymeric isocyanate. The amount of glue spread used was appx.250 g/m^2 for manufacturing 3-ply plywood. The plywood was hot-pressed at 110 -115°C and 11-12 kg/cm^2 specific pressure for 14 minutes for 12mm plywood. The core temperature of the plywood was measured with a thermocouple placed in the core veneer during the hot-pressing.

Evaluation of plywood properties

Plywood properties such as glue shear strength in both dry and wet conditions, Water resistance properties were tested as per IS:1734 (1983) and formaldehyde emission were evaluated as per IS:13745(1993). The bond strength of plywood was checked by knife test as per IS 848-2006 in both MR and BWR Grade. The tensile strength of the plywood was tested as per IS:1734(1983) in both along and across the grain.

RESULTS AND DISCUSSION

Properties of UF/B-pMDI resins

The gel time of hybrid UF/pMDI resins and control samples was determined to compare their activity and the results obtained are displayed in Tab.-1

Table No 1 Properties of UF/PMDI Hybrid resin

Parameters	Results
Solid content(%)	51.8
Water tolerance	1:04
PH	8.0
Viscosity (cps)	76
Gel time(s) @100^0C	68

Table No 2 Properties of adhesive mix

Parameters	Results
Flow time of adhesive mix in B6 flow cup in seconds	18
Solid content of adhesive mix(%)	60.2
Adhesive pH	7.1
Pot life (hrs)	06

Table No 3 Composition of the adhesive mix used to prepare 9 ply plies (12 mm thickness)

Adhesive mix		Board parameters for 7 ply plywood	
Component	Parts by weight	Characters	Board Parameters(mm)
Resin UF Liquid	200	Number of plies	9
NH_4Cl	1.0	Face longitudinal	0.5
Liquor. NH_3	1.6	Cross band (cross grain glued)	1.8
Wheat flour	6.0	Long core (Longitudinal grain	1.8
Insecticide(GLP)	0.5	Cross band (cross grain glued)	1.8
		Long core (Longitudinal grain	1.8
		Cross band (cross grain glued)	1.8
		Back (Longitudinal grain)	0.5

Table No 4 Plywood manufacturing condition

Sl. No.	Adhesive Type	Grade of Plywood	Hot Press Temperature ^{0}C	Hot pressing pressure Kg/cm^2	Hot pressing time (mnt.)
1	UF Resin adhesive	MR	110±5	10	12 minutes for 9ply 12mm

CONCLUSION

The study investigated the feasibility of manufacturing of water resistance plywood by using UF resin modified with polymeric isocyanate by manufacturing UF/PMDI hybrid resin.

Table No 5 Bonding quality of the plywood tested as per IS; 848-2006

Test	Test Method	Criteria for conformity	Results
BWR GRADE (Boiling water Resistant) Three cycles: Each cycle consisting of 8 hours boiling in water and thereafter drying at 65±2^{0}C for 16 hours.	Clause 4 and 7.3.2 Of IS :848 - 2006(Indicated in table -1)	No separation of plies at the edges and /or surface at the end of three cycles. On forcible separation of plies with knife, wood failure shall be predominant and shall be more than 75% for excellent bond and not less than 50% for pass standard. For less than 50% wood failure, the specimen shal be considered as failed.	Separation of plies at the edges and /or surface at the end of three hrs. Boiling water resistance sustained up to 03 hrs.
MR GRADE (Moisture Resistant) Three cycles: Each cycle consisting of 3 hours 60±2^{0}C in water and thereafter drying at 65±2^{0}C for 8 hours.	Clause 4 and 7.3.2 Of IS :848 - 2006(Indicated in table -1)	No separation of plies at the edges and /or surface at the end of three cycles. On forcible separation of plies with knife, wood failure shall be predominant and shall be more than 75% for excellent bond and not less than 50% for pass standard. For less than 50% wood failure, the specimen shall be considered as failed.	No separation of plies at the edges and /or surface at the end of three cycles. 60% Wood Failure Pass Standard

Table 6 Physico-Mechanical properties of the plywood on the basis of pilot scale using UF/ pMDI resin@10% conc.

Sl. No.	Powder resin grade (%)	Average Glue Shear Strength						Static Bending				Tensile Strength, N/mm2	
		Dry State		Wet State (After cyclic test)		Resistance to Mico-organism		MoR, N/mm2		MoE, N/mm2			
		Load, N	Wood Failure, %	Load, N	Wood Failure, %	Load, N	Wood Failure, %	Along	Across	Along	Across	Along	Across
1	UF	950	65	780	50	730	50	39.46	46.67	4895	2948	31.25	30.22
3	UF/ pMDI	1125	70	880	60	810	55	44.27	67.10	4863	5937	39.45.	42.95

Table 7 Formaldehyde emission (perforator value) of the plywood testes as per IS 13745:1993

Sl. No.	Type of resin used	Tests	Clause No	Test Method	Requirement as per IS 3087:2005	Results
	2	3	4	5	6	7
1	C0	Perforator value (mg formaldehyde/100gm dry board.)	Clause 9.7	IS 13745:1993	8mg/100gm For E1 Class	44.44
2	C1					38.6.
3	C2					32.7
4	C3					32.6
5	C4					29.8.5
6	C6					26.5

Adhesion performance of plywood i.e dry and wet glue shear strength of plywood bonded with UF/pMDI resin after the addition of 1 and 10% pMDI resin were determined to compare the adhesion of hybrid resins, and the results are presented in Fig. 7. As expected, the pMDI resin had an obvious influence on the glue shear strength of plywood with both dry and wet. Glue shear strength values increasing as the concentration increased up to 10%. Both dry and wet glue shear strength values increased by 20% and 25%, respectively, compared with that of the control. These high dry glue shear strength values are attributed to the formation of various bonds between the isocyanate group and the $-CH_2OH$ groups of UF resins or those between the isocyanate group and the hydroxyl group of wood. Boiling water resistance properties of the plywood manufactured with UF/ pMDI resin showed better then control samples .This might indicate an improvement in the water resistance of the plywood due to the incorporation of the pMDI resin into the UF resin. Though plywood does not show water resistance properties upto BWR grade plywood when tested as per IS 848(2006), but able to withstand up to 3 hr at 100^{0}C in cyclic test.

The results showed that the plywood with improved glue shear strength, Static bending strength, tensile strength, water resistance and considerable limited formaldehyde content may be obtained in the case of Urea formaldehyde modified with emulsifiable polymeric isocyanate (PMDI) at a concentration of 10%,However the plywood does not confirms to the boiling water resistance required as per the BWR Grade when tested as per IS :848 -2006. The results obtained from this study suggest that the modification of UF resins with pMDI resin is a way of improving the water resistance and though formaldehyde content does not confirms to E1 emission standard for all concentrations, However values reduced 20 to 30% as compared to control sample.

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