Effect of adhesive performance of liquid urea formaldehyde (UF)resin when used by mixing with solid UF resin for manufacturing of wood based panels

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Abstract- The purpose of this paper is to investigate the effect of solid powder urea formaldehyde resin on the quality of the plywood when manufactured by mixing with liquid urea formaldehyde resin. The finding obtained would benefit the manufacture of urea formaldehyde adhesive bonded composite panels. The study is focused on new solution for production of high quality wood based panels by using technique of mixing of powder resin with liquid urea-formaldehyde resin to make better adhesive system. In order to improve the adhesive and bonding properties solid powder urea formaldehyde resin was mixed with liquid Urea formaldehyde resin, 12mm plywood was manufactured and bond quality was studied as per IS:848:2006. Conventional hardener – so called latent catalyst namely ammonium chloride was used to react with existing free formaldehyde of the resin forming a strong acid that promotes resin cure. From the study, data shows that glue shear strength for both dry state, weight state and in mycological state, tensile strength and static bending strength values shows satisfactory results. There is a significant change in results of MoR and MoE of plywood samples manufactured by using powder resin mixed with liquid resin. It was also found that after using 10% concentration of solid UF resin, the pre press tackiness was improved drastically which helps to improve the quality of plywood. The study developed a new solution or technique that can give a solution to manufacture plywood by using pre press technique and manufacture of wood based composites having more mechanical and bonding strength by using urea formaldehyde resin.

Index Terms- Bonding, glue shear strength, MoR and MoE of plywood, pressing, resins, rheological properties, wood composite materials.

I. INTRODUCTION

Urea–formaldehyde (UF) resins are one of the most widely used wood adhesives with wide applications in the manufacture of wood-based composite panels such as plywood, particleboard, and medium-density fiberboard. Some of the advantages of UF resins over other adhesives are: easy handling, low cost, and good performance in panel products. UF resins also have high reactivity and hence by the shorter hot press time achievable as well as full hardening, clear glue line, cold tack, absence of organic solvents, synergetic effect with other adhesives, and non-flammability. However, there are two most important disadvantages: low moisture resistance and formaldehyde emission from wood-based composite panels in service which is known to cause sick building syndrome. These drawbacks are driving research to understand basic properties of UF resins that can lead to developments aimed at reducing formaldehyde emission from wood-based composite panels bonded with UF resins.

Urea-formaldehyde (UF) resins are the most used resins for interior-grade plywood application. However, the construction of the plywood panel itself affords better protection of the glue line from water attack than in other types of wood panels. The major defect of UF resins is their high sensitivity to water attack, forti- fication with small amounts of melamine being generally used to yield better results, even for interior-grade panels. Hybrid resins in which polymeric isocyanate (pMDI) is used to reinforce the characteristics of full exterior-grade resins are already known for several applications (Adcock et al. 1999, Pizzi and Walton 1992, Pizzi et al. 1993, Simon et al. 2002, Shadurka et al. 2003, Des- pres et al. 2005). Combining then hybrid resins with the more suitable plywood panel architecture led to the use of small pro- portions of pMDI to upgrade the water resistance of UF plywood glue lines in the same manner as is current practice with melamine addition to UF resins

Wood adhesives are polymeric materials that are capable of interacting physically or chemically or both with the surface of wood in such a manner that stresses are transferred between bonded materials. Adhesives are to be applied in the form of liquid to spread over and wet the surface of the wood. Urea Formaldehyde Resin is most important type of adhesive in the wood industry since last 60 years especially for the production of wood based panels. More convenient spray dry UF powder is in the market for the last two decades. Small and medium private based wood panel industry prefers to use powder resin since they are more convenient for small capacity production. During last two decades, certain effort have been undertaken for its improvement like reduction of gel time, free formaldehyde emission, adhesive bond fortifying, prolongation of storage life and more convenient transport and application. Liquid UF resin concentration up to 50 to 55% solid content were prepared within wood industry complex is having storage life of 40 days maximum were more convenient but at the same time spread. UF powder resin can be used by re-dissolved and re-dispersed in water at different concentration just before application.

So efforts have been made in this study to make a glue system for wood based panel industry by combination of powder

resin with liquid UF resin. Petroleum is a finite natural resource subject to the vagaries of market forces. This improves the durability - stability of UF resins as a potential substitute for more petrochemical dependent, less versatile, and more supply - sensitive PF resins which will be a welcome development for both manufacturer and users alike. The present study is one of such attempts.

II. Materials and Methods

2.1 Stage Materials

Phenol, formalin, caustic, urea etc. used for resin synthesis was collected from local market which belongs to commercial grade. Wood Veneer used for manufacture of plywood belongs to Dipterocarpussp.(Gurjan). Other chemicals used during testing were of analytical grade purchased from local market. MR Powder resin (SB MR – 202G) taken from M/s ARCL, Kolkata, India which are powder Melamine Urea Formaldehyde resins for Plywood Industry specially developed for MR Grade.

Properties	**MR -SB MR – 202G**
Appearance	White free flowing powder
Solubility	Easily soluble in water
Bulk Density	0.69 gm/cc
PH (50 % Solution)	8-9
Flow time (1 part resin + 1 part Water) 25°C, in Seconds)	16-18
Gel time	118 seconds

2.2 Methods

2.2.1 Synthesis of UF 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 was checked from time to time and was 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 steady result instead of viscosity, flow time of the reaction mixture was measured in B4 cup (as per IS: 3944:1982). 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 flow time of 16-18 seconds in B4 flow cup and water tolerance of 3-4 times was observed. The reaction was arrested by raising pH to (7.5 – 8.0) by adding 50% alkali and then the resin was cooled.

2.2.2 Formulation of adhesive mix

Using synthesized UF resin, plywood adhesive mix was formulated by mixing synthetic resin, hardener, buffer and UF/MR/BR powder resin at various concentrations from at concentration level from C0 to C6 to standardize the flow time of glue under stirring with a speed regulated stirrer (Ref Tab 3). The mixing was then continued for 30 minutes until a homogeneous mixture was obtained. Viscosity of the glue after mixing was taken in B6 Cup (IS:3944-1982) and the adhesive mix was taken for glue durability test through water resistance test as per IS :303-1989.

2.2.3 Viscosity assessment

A change in glue viscosity exerts large effects on glue spreader rates. Glue mix with high viscosity is difficult to spread and leads to higher glue spread which is undesirable from quality and economic point of view. For a resin, viscosity is a measure to assess the progress and extent of polymerization of the resin either during manufacture or use and for adhesive it should be satisfactory coverage up to 28-30 gms /sq. ft. In plywood industry the actual viscosity of the adhesive are studied by use of standard flow cup. In this study the flow time of the adhesive mix was studied by B6 flow cup (as per IS:3944-1982) at different concentration level. The results for standardization of flow time are given in Tab 4.

2.2.4 Spreading process

Spread rate will largely depend on the nature of the work being done and the method of cure employed. Rates are normally defined by gms of glue required per 1m2 of board surface to be glued. Spread rates will range from 28 -30 gms/ft2 depending on the nature of the work, time constraints of the glue-up, and method of cure. Care should be taken to check the thickness of veneer and relative porosity of the species to be used and the spread to be adjusted accordingly. In many instances, a thin spread coupled with mandatory open assembly time will be necessary to prevent excessive bleed-through. Hot Press pressure also has great bearing on the spread rate. Lower pressures should have less glue in the glue line, as thick glue lines will take longer time to cure.

2.2.5 Pot Life assessment

Once mixed with water, the resin will advance in its cure stage until it is unusable. The amount of time between first mixed until it reaches the unusable point is termed pot life. As with most directions for gluing, this is a dynamic value. It will change primarily due to temperature. Hotter temperatures yield a shorter pot life and vice versa with colder temperatures. At 250C pot life will be 5-6 hours and at30-350C pot life will be 3-4 hours.The results for standardization of flow time is given in Tab 4.

2.2.6 Layup and Stand time assessment

Since layup time depends on the number of plies in a panel, type of core, glue formulation, number of panel etc. and stand time depend on veneer moisture content, veneer temperature, pressure, temperature and spread rate. So proper layup and stand time has to be maintained during manufacture of plywood. The time after spreading the glue, before full pressure is applied is referred to as assembly time. Open assembly is after the glue is applied to the veneer but before it is put together with the mating surface. Closed assembly is after the surfaces have been placed together but before full hot press pressure is applied. Open and closed assembly times are interrelated. The maximum closed assembly time is only reached when virtually no open

assembly time is given and is reduced about 2 minutes for every minute of open assembly time. As temperature increases, allowable assembly times decrease. Maximum assembly time at 25-28^{0}C is 45 minutes and at 30-35^{0}C is 22 minutes. The results for standardization of flow time are given in Tab 5.

2.2.7 Stock Preparation

The best results are obtained with the best stock preparation. The closer these conditions are adhered to, the better the results. Equilibrium moisture content (EMC) should be 8-10% max of 12% for cold pressing which may go maximum up to 10% for hot pressing. It is critical to know the destination of the end piece, so that moisture content change will not be extreme. Surfaces should be free of dust, dirt, grease or other contaminants.

2.2.8 Pressing and Curing

As with pot life, both hot press pressure and cure time are dynamic depending on the conditions at hand. Pressure required to develop sufficient bond strength is dependent upon the construction and the wood species used with the guiding rule that surfaces must be in intimate contact with the glue.

2.2.9 Pre Pressing Technique

The technique of pre-pressing involves a temporary bond formation between the glue coated veneers before hot pressing. The glue coated veneer assembly is compressed near to its final thickness during the short pre-pressing cycle. In a pre-pressed panel, the uncured glue is used to hold the veneers together. The technique of pre-pressing is becoming more and more popular in the manufacture of plywood particularly where hot presses of more than ten daylight openings are used. In this study 1ft x 1ft size 10 nos plywood was manufactured by using Eucalyptus sp. as core veneer and gurjan spp. as face veneer. 10% of BR-201G was mixed with Liquid MUF and UF resin for achieving prepress tackiness of the glue. Details of pre- pressing condition, glue properties and mechanical properties are given in Tab 9 to Tab 11.

III. Results and Discussion

3.1 General Resin Properties

The specific gravity, solids contents as well as the viscosity values of the synthesized resins showed little differences and these values are within the normal ranges of industrial values (Ref Tab 3).

3.2 Adhesive Mix

Adhesive mix for plywood manufacture using three powder resin with liquid resin shows homogeneous mixture after mixing of 30 minutes in glue spreader. The powder resin doesn't shows any chemical reactions with liquid UF and MUF resin even after mixing with hardener and buffer which implicates satisfactory results for adhesive mix. The concentration of adhesive mix from extension level C0 to C6 shows satisfactory concentration for glue spreader and adhesive mix viscosity after mixing properly in a glue spreader minimum 30 minutes (Ref Tab 3).

3.3 Assessment of Viscosity and physicochemical properties

From Viscosity study (Ref Tab 4), data shows that viscosity of adhesive mix measured in terms of flow time in B6 (as per IS 3944-1982) shows satisfactory result at extension level C3 for smooth operation of Glue Spreader. From the data, it has also been observed that the viscosity of adhesive mix increases with increasing extension level. Solid content of adhesive mix increase as significantly as the concentration of the powder resin increases in the adhesive mix with satisfactory PH and pot life.

3.4 Layup and Stand type assessment

Since all gluing operation have to be completed within a giving period of time i.e. from the spreading of adhesive on veneer to the application of pressure in the assembly, so layup and stand time of the adhesive mix should resist within the period. Layup time should not be too short or high to avoid solvent loss and time during panel assembly. From the data (Ref Tab 5) reveals that powder resin mixed with adhesive mix shows significant result towards bond quality during boiling water test when total layup and stand time lays for 60 minutes. But it shows unsatisfactory in terms of glue shear strength and percent of wood failure when both layup and stand time exceeds 60 minutes.

3.5 Adhesive bond quality assessment

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Bonding quality was tested according to IS 848:2006 for both UF, MR and BR grade powder resin. The results are summarized in Tab 5 to Tab 8. MR and BR grade resin at 10% (w/w) of liquid resin were used to test for BWR Grade plywood and results are summarized in Tab 13. UF, MR and BR grade powder resin shows excellent results in cyclic test. Glue shear strength value shows more in comparison to MR and UF grade (Ref Tab 7). During the study of glue shear strength for dry state, wet state and mycological state, tensile strength and static bending strength, results shows satisfactory value an there is a significant change in results of MoR and MoE has been obtained for plywood samples manufactured by using powder resin mixed with liquid resin (Ref Tab 8).

3.6 Pre pressing and Hot pressing Parameter

TABLE 4
Lay up and stand time standardization

Powder resin grade	Level of conc	Flow time in B6 flow cup (sec.)	Lay out time (min)	Stand time (min)	Quality	
					Cyclic test MR Grade	% of wood failure
MR (202G)	C5	24	20	15	Pass standard	85
			30	15	Pass standard	85
			45	15	Pass standard	75
			60	15	Failed	40
			75	15	Failed	40

In plywood industries the former process is known as cold pressing and the later process is known as hot pressing and both the process leads to curing of resin. From the study for manufacture of plywood to assess the rheological properties of powder resin, the parameters for hot pressing and cold pressing (Ref Tab 9 to Tab 12), it shows satisfactory results specially in case of bond quality and surface finishing of plywood, result shows plywood delaminates after 3 hours in case of MR Powder resin with liquid UF resin and delaminates after 9 hours in case of BR 201G with liquid MUF resin tested as per IS 848- 2006.

TABLE 3
Properties of adhesive mix with Powder resin MR

Properties	Level of concentration (%)						
	C0	C1	C2	C3	C4	C5	C6
Flow time of adhesive mix in B6 flow cup in seconds	20/B4	10	13	18	23	26	31
Solid content of adhesive mix (%)	48.0	49.2	52.4	53.5	53.7	54.9	56.2
Adhesive pH	6-6.5	6-6.5	6-6.5	6-6.5	6-6.5	6-6.5	6-6.5
Pot life (hrs)	> 6	> 6	> 6	> 6	> 6	> 6	> 6

IV. TABLES

TABLE 6
Composition of the adhesive mix used to prepare 7 ply plies (12.0 mm thickness)

Adhesive mix		Board parameters for 9 ply plywood	
Component	Parts by weight	Characters	Board Parameters (mm)
Resin UF Liquid	200	Number of plies	9
NH4Cl	1.0	Face longitudinal	0.35
Liquid NH4	1.6	Cross band (cross grain glued)	1.8
Powder resin	3.0	Long core (Longitudinal grain)	1.8
Insecticide (GLP)	0.5	Back (Longitudinal grain)	0.35

TABLE 1
Properties of synthesized Urea-formaldehyde resin

Gel Time at 100ºC (seconds)	Flow Time (B4 cup) (seconds)	Solid content (%)	Water tolerance	pH	pH of Cured film	Free formaldehyde (%)
74	20	48.8	1:4	8.0-8.5	5.6	0.90

TABLE 2
Adhesive mix with different concentration of Powder resin (MRGRADE)

Component	Parts by weight (Pbw) Level of concentration (%)						
	C0	C1	C2	C3	C4	C5	C6
UF Resin Liquid 48% solid	200	200	200	200	200	200	200
Powder resin MR	0	4.0	6.0	8.0	10.0	12.0	14.0
NH4Cl	1.0	1.0	1.0	1.0	1.0	1.0	1.0
Liquid NH4	1.6	1.6	1.6	1.6	1.6	1.6	1.6

TABLE 5
Properties Effect of coverage on Glue Shear Strength with respect to coverage spread = 350±10 gms/m^2 DGL

Powder resin grade	Conc Level	Dry State		After cyclic test (MR Grade)	
		Load (N)	Wood Failure (%)	Load (N)	Wood Failure (%)
MR (202G)	C0	1020	65	754	55
	C1	1154	65	787	55
	C2	1248	65	812	55
	C3	1298	70	832	60
	C4	1365	70	888	60
	C5	1420	70	906	60
	C6	1486	70	916	60

TABLE 8
Static Bending and Tensile Strength of the plywood on the basis of pilot scale using powder resin.
of synthesized Urea-formaldehyde resin

Sl. No.	Powder resin grade (%)	Static Bending				Tensile Strength (N/mm^2)	
		MoR, N/mm^2		MoE, N/mm^2			
		Along	Across	Along	Across	Along	Across
1	UF+Conv Extender	39.46	46.67	4895	2948	31.25	30.22
2	MR	44.27	67.10	4863	5937	39.45	42.95

TABLE 9
Board Pressing conditions of adhesive mix with Powder resin (MR GRADE)

Parameters	Cold Pressing	Hot Pressing (UF Resin)
Pressure	8 Kg/cm^2	10.0 Kg/cm^2
Temperature	NA	110°C
Time	15-20 minutes	12 minutes/12 mm ply

TABLE 7
Glue Strength of the plywood on the basis of pilot scale using powder resin of synthesized Urea-formaldehyde resin

Sl. No.	Powder resin grade (%)	Average Glue Shear Strength					
		Dry State		Wet State (MR Grade)		Mycological State	
		Load (N)	Wood Failure (%)	Load (N)	Wood Failure (%)	Load (N)	Wood Failure (%)
1	UF+Conv Extender	1375	70	780	60	730	55
2	MR	1525	70	880	60	790	55

TABLE 11
Pre-pressing Conditions

Adhesive Type	Moisture Content of Veneers (%)	Closed Assembly Time (Minutes)	Prepressing Pressure (Kg/cm^2)	Pre-pressing Time (Minutes)
UF Resin	8-10	20-30	8	15-20
UMF- 50 resin	6-8	20-30	8	15-20

TABLE 12
Hot Pressing Conditions Conditions

Adhesive Type	Grade of Plywood	Hot Press Temperature (^0C)	Hot pressing pressure (Kg/cm^2)	Hot pressing time (Minutes)
UF Resin	MR	110±5	10	12 minutes for 9 ply, 12mm thk

TABLE 10
Adhesive Formulations for Pre-pressing Pressing conditions of adhesive mix with Powder resin (MR GRADE)

Type of Resin	% of Powder resin on the wt. of liquid resin	% of filler/ Extender	Type of buffer and its % on the wt. of liquid resin	Catalyst and its on the wt. of liquid resin	Flow time (B6 Cup)	Grade of plywood
UF Resin	BR-201G 10%	10%	Liquor ammonia (sp. Gr. 0.91):0.4	NH_4Cl 0.075 - to 0.1	35-40	MR

V. Conclusion

The different grade of powder resin having solid content 100% doesn't show any chemical reaction with Liquid resin even after mixing with hardener and buffer rather than forming creamy and is lump-free during mixing. Satisfactory rheological properties of the adhesive and physio co-mechanical properties of the plywood made with mixing resin technology was achieved. Satisfactory pre-press results have been achieved at higher concentration of powder resin mix. The chances of pre-cure of dry out was reduced because veneers are held in close contact with each other. The adhesive wets the veneer surface since the layers of veneers have an opportunity to come into intimate contact with each other for greater period of time. Hence the quality of the plywood improved by overcoming warping, pre-curing etc. Plywood delaminates after 3 hours in case of MR Powder resin with liquid UF resin and delaminates after 9 hours in case of BR 201G with liquid MUF resin tested as per IS 848 - 2006 for BWR grade plywood by adding 10% (w/w), however plywood confirms to MR Grade in all the cases.

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