"A Natural Additive Approaches to Enhance the Performance of Formaldehyde Based Adhesive for Plywood Manufacturing"

S. C. Sahoo, A. Sill., C. N Pandey

Abstract- There is a need for additive that enhance the different properties like rheological , tackiness etc. Of the adhesive mix during plywood manufacturing bu using non formaldehyde based adhesive. The present study relates the efficacy study of the natural additive, which is basically fully based on starch and protein available from renewable sources containing a natural tackifier and viscosity enhancer. The adhesive is based predominately on a natural product for example protein, starch based natural material, lignin etc. Thus complementing the use of a lignocelluloses as the primary material. The additive was analysed for physical and chemical properties as per is : 1508 Like P^H ash content. Moisture, protein content etc, in order to evaluate the suitability of bond quality and other properties during manufacture of Plywood. 12mm thickness plywood were manufactured with non Formaldehyde adhesive using natural additive at different concentration and evaluated different physical, che3micals properties and board manufacturing condition. Data revels that additive of three to four percent concentration gave satisfactory results in terms or board quality, rheological properties and mechanical properties etc. Five to six percent additive concentration gave excellent result. Addition of higher percent gave unsatisfactory results for manufacturing of ply boards.

Keywords: - Additive, starch and protein, natural product, rheological properties and mechanical properties, natural tackifier and viscosity enhancer.

I. INTRODUCTION

Adhesive formulations are frequently based on room and high temperature curing for synthetic resins such as phenol-formaldehyde, phenol-resorcinol-formaldehyde, urea-formaldehyde, urea-melamine-formaldehyde, and polyvinyl acetate glues. These resins are extended with additives which can have some adhesive properties in addition to the capable of controlling viscosity or other rheological properties of the formulation. Fillers used in North American structural-plywood adhesives often contain up to 80 percent carbohydrates and therefore they are often referred to as "Extenders."Fillers and extenders are the materials added to synthetic resin adhesive to improve their spreading characteristic to control excessive adhesive penetration into the substrate and to reduce the amount of primary binder required per unit area. The material may be organic or inorganic. In the wood bonding industry, the conventional adhesive extender used is wheat flour. Wheat flour prices have tripled in recent years and supply is getting difficult as it is mainly used as food material. Fillers used are usually the powder of corn cobs, tree barks or walnut shells. Their supply is inconsistent and their prices have also increased rapidly. Wheat flour is used in the art of adhesives as an extender in urea-formaldehyde plywood glues it can represent, for example, 25 percent by weight of the total glue. Since wheat flour is a staple foodstuff, it is a relatively expensive ingredient for glue mixes, and its availability varies with the world commodities market. A less expensive substitute for wheat flour which will perform similar to wheat flour in such glue mix is therefore desirable for plywood manufacturing and other adhesives applications. The search for a substitute extender and filler material becomes one of the most important tasks to the plywood and laminating industries.

Wood adhesive extenders are amylose compounds with some protein content that have adhesive action and contribute to the rheological properties of the glue mix. The physico-chemical properties of these extenders are very important to establish. For instance, high ash, high crude fat and high fiber contents pose unpredictable viscosity problems and increase wash water requirement. Protein has been found to influence the water taking capacity of flour (Robertson, J 977).

To reduce costs, synthetic glues are mixed with an extender that can be either imported maize starch (US$650/tons) or food grade wheat flour (US$500/tons). Typically 50 kg of synthetic glue will make 55-60 *1/8"* plywood sheets, with an extender th is increases to 80-85 sheets of *1/8"* plywood sheets. For each 50 kg batch (of synthetic glue), either 10 kg of maize starch or 25 kg of wheat flour is required (Graffuam *et al. 1997).*

Proteinaceous and amylaceous materials are most often called extenders because they can enter into the matrix of the binders. Fillers in North American structural-plywood adhesives often contain up to 80 percent carbohydrates; therefore, they are often referred to as "extenders." Fillers are extensively utilized in adhesives and sealants, coatings, plastics, medicines, and cleaning compounds (Sellers 1994).

Manuscript Received on December 2014.

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One of the main changes that has occurred in wood construction in the last 50 years has been the adoption of glue technology, to bond structures together and to make wood available in a more stabilized, laminated sheet form as plywood. Plywood mills in India use maida/wheat flour as extender in the adhesive mix for plywood production. Maida andWheat as a commodity is not grown in India and is therefore imported. Its market price keeps soaring-up because it has competing uses. It is used in India as a food source particularly for bread and pastries making in addition to its use as extender in plywood mills.

Authors like Narayan Murti et al (1943, 1962), Jain, et al (1963, 1975), Narayan Prasad, et al (1969, 1976), Roy, et al (1971), Mohan Das, et al (1976) has reported the used of various materials such as casein, Ground nut cake powder, Lignin rich materials, paddy husk gel, Tamarind seed powder, Deoiled maize gluten in various form into adhesive formulations based on PF resin for bonding BWR plywood. Raghunath Rao, et al, (1993) have review the effect of commonly used protinious and sturch based fillers and extenders on phenolic resin bonded plywood. Naha, et al (1995) have studied the utilization of various oil seed cakes flours such as Iarati, Mahua, as extenders for UF and PF resin for bonding different grades of plywood.

II. MATERIALS AND METHODS

A. Materials

Materials used for manufacture of additive like soya powder,ground nut shell powder,cocunutshell powder,wheatflour ,guar gum,xanthungum and cellulosic materials (commercial grade) was procured from local market .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 Dipterocarpus sp (Gurjan). Other chemicals used during testing were of analytical grade purchased from local market.

B. Methods

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 7.2 -7.5. 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 is checked time to time and maintained at 7.5 – 7.8.

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 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 viscosity is 80-100 cp or have a flow time of 22-30 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. .

Synthesis of Modified PF resin :- Novolac resin: 100 parts by weight of phenol and 60 parts by weight of formalin (37% formaldehyde) were charged into resin kettle. Stirring started and continued till the end of reaction. 2 parts by weight of oxalic acid in 33 parts by weight of water was added in case of acid catalyzed novolac resin. The reaction was carried out at 90±2ºC for two hours. At the end of first stage reaction white insoluble product is formed which separates from aqueous layer on keeping. The resin was then cooled to 60ºC.

Resol resin: 16 parts by weight of sodium hydroxide dissolved in 66 parts of water was added in case of acid catalyzed novolac resin. 120 parts by weight of formalin (37% formaldehyde) was added next. Exothermicity was carefully controlled at this stage. Reaction is further continued at 85ºC. When viscosity of the resin is 80-100 cp or flow time 20-30 secs in B_4 flow cup of IS: 3944-1982, the resin was cooled and discharged from the kettle.

C. Formulation of Natural additive for plywood adhesive -

Additive for plywood adhesive was formulated by taking wheat flour 50 parts,soya powder 10 parts, ground nut shell powder 15parts ,cocunutshell powder,wheatflour 05 parts ,guar gum -2 parts ,xanthungum 03 parts,Sodium cmc -2 parts and next make upto 100 parts with filler.

D. Formulation of adhesive mix:-

Using both synthesized UF and PF resin each plywood adhesive mix was formulated by mixing synthetic resin, hardener, buffer and Additive at various concentrations from extension level from C_0 to C_6 to standardize the flow time of glue under stirring with a speed regulated stirrer. (Table No-2) .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) for both amino and phenolic resin by taking four different types of extender used for both prepress and hot pressing purposes (Table No-3). The adhesive mix was taken for glue durability by boiling water and mycological test. The coverage study was carried for both UF and PF resin based adhesive mix after addition of Additive. The glue shear strength in dry, wet and mycological was carried out by Tensometer for different extension level (C_0 to C_6).- (Table No-7 ,8)

E. Physical and chemical properties of Additive :-

Additive was evaluated for certain physical characteristics as per IS:1508 i.e. Color, moisture content. PH, ash content etc.. The size of extender was determined by sieve analyzer as per IS: 460-1962. Sieve designation 100 microns and Sieve designation 80 microns on percent basis was taken for this study. Each Additive was also analyzed for fat and protein contents etc. (Table No-4)

F. Viscosity assessment:-

A change in glue viscosity exerts large effects on glue spreader rates.

Published By:
Blue Eyes Intelligence Engineering
& Sciences Publication

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. 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(IS:3944-1982) flow cup at different concentration level Additive using in both phenolic and amino resin . The results for standardization of flow time was given in Table No-3.

G. Layup and Stand time assessment:-

Since layup time depends on the no of plies in a panel, type of core, glue formulation, no of panel etc. and stand time depend on veneer moisture content, veneer temperature., pressure and ambient temperature. and spread .So proper layup and stand time is required to maintain during manufacture of plywood. In this study layup and stand time for individual Additive extender/filler has been studied and optimized and the time and bond quality has been studied . Table No-5.

H. Plywood Manufacture (Laboratory scale) :-

Seven ply plywood was manufactured by taking Gurjan (Dipterocarpus) species as core, face and back veneer of size 2' x2' and 12mm thickness .Each adhesive mix was applied on the veneer and conditioned for moisture content up to 14% with a roller coater at a spread rate 300 – 350 gms/m^2 on both sides basis .The spread veneers were then pressed in a hot press at pressure 10.5 Kg/cm^2, 14 Kg/cm^2 and temperature 110^0C ,140^0C for for UF and PF resin respectively for 12 minutes to get 12mm thick plywood .The total assembling time ,which includes from first adhesive application to hot pressing was taken one hour. (Table No-8)

I. Experimental design :-

In the adhesive bond quality evaluation (dry, wet and Mycological tests), a 4 x 5 x 3 factorial experiment in completely randomized design (CRD) was employed: 4 types of extender x 5 adhesive extension levels x 3 conditions. This makes a total of 60 nos combinations study. The extenders used were Additive of four different grade. The extension levels were 1 to 6% based on liquid resin and the plywood pressing times were 6 and 12 minutes. as per the thickness of the plywood. Adhesive mix without extender served as a control.

J. Adhesive bond assessment:-

The manufactured plywood were tested in boiling water resistance and boiling waterproof plywood .Glue shear strength, résistance to microorganism, tensile strength ,percentage of wood failure and static bending strength etc. tested as per IS:1734/1983 for assessment of bond quality . (Table No-6&7) .

III. RESULTS AND DISCUSSION

The ash content of extender is an important value to asses knife wear test low ash content shows low knife wear test and vise versa. Data of this study (Table no-4) reveals the value of ash content varies from 10.6 to 12% in case of Additive. The screen mesh size grain or particle size value of four extenders shows significant result rising from 89.4 to 90.7% as per IS : 460-1962. Sieve designation of 80 micron result shows that the four extenders is suitable for plywood manufacture for adhesive mix instead of causing glue spreader problem. Moisture content, Protein and fat content result shows satisfactory results. Adhesive mix for plywood manufacture using four types' extenders shows homogeneous mixture after mixing of 30 minutes in glue spreader. . The concentration of adhesive mix (Table No.-2) from extension level C_0 to C_6 shows that the extension level C_3 i.e. 3 percentage concentration of liquid resin in both UF and PF. Rheological proprieties such as Viscosity can be directly co-related to the evolving Physical and Mechanical Proprieties during the resin period. From Viscosity study (Table No-3) data shows that viscosity of adhesive mix measured in terms of flow time in B_6 IS : 3944-1982 shows satisfactory result at extension level C_3 for both UF and PF resin for smooth operation of Glue Spreader. The flow time measured in B_4 Cup for UF301 at 3 percentage concentration in 13 seconds and for PF-502 it is 12 seconds , From the dat

a of the study it has been observe that the viscosity of adhesive mix increases with increasing extension level. Solid content of adhesive mix increase as significantly as the concentration of the extender increases in the adhesive mix with satisfactory P^H and pot life. 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 (Table No-5) reveals that Additive mixed with adhesive mix shows significant result towards bond quality during boiling water test when total layup and stand time lies 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.Bond quality assessment study (Table No-6,7) data shows that during study glue shear strength for both Dry , Weight and Mycological, Tensile strength and Static bending strength values shows satisfactory result there is a significant change in results of MOR and MOE of plywood samples made by Additive during pilot scale study. Glue shear strength in a Dry, Weight and Mycological lies between 800 to 2100 N range for PF resin which is also shows satisfactory results.

Data from different coverage rate has shown that at the minimum coverage 330 gms/m^2 (D.G.L) adhesive bonding strength in dry.wet and mycological ware also satisfactory for PF resin wood failure percentage lies in the range 60 to 85 % .

IV. CONCLUSION

Relative advantages and disadvantages of Additive used as extender/filler in PF/UF resin in plywood industries, was study under this project may be summarized as it is recommended to incorporate less concentration of Additive (C_3 extension level) in comparison to used conventional extender/filler for smooth operation of glue spreader and to get the better bond quality of both phenolic and amino resin bonded plywood. Additive was found to be better in comparison to starch based extender with phenolic resin for manufacture of BWR, BWP plywood and other wood based panel products.Glue shear strength value in dry, wet state of plywood bonded with PF and UF resin after incorporation with Additive increases however the glue shear strength value after mycological test does not give satisfactory results.It is possible to meet the challenges of the global requirement for continuous improvement of the panal performance by using integrated binder syste (resin + additive).

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Results :-

Table no. 1(a) - Properties of synthesized Urea-formaldehyde resin

Gel Time at 100°C (seconds)	Flow Time (B4 cup) (seconds)	Solid content (%)	Water tolerance	pH	PH of Cured film	Free formaldehyde (%)
65	22	48.5	1:4	8.2	2.62	0.56

Table no. 1(b) - Properties of synthesized Phenol formaldehyde resin

Gel Time (minutes)	Flow Time (B4 cup) (seconds)	Solid content (%)	Water tolerance	pH	Free phenol (%)
23	24	49	1:4	10.14	0.20

Table no. 2(a) - Adhesive mix with different concentration of Additive.

Component	Parts by weight (Pbw)						
	Level of concentration (%)						
	C_0	C_1	C_2	C_3	C_4	C_5	C_6
UF Resin Liquid 48% solid	200	200	200	200	200	200	200
Additive	0	2.0	4.0	6.0	8.0	10.0	12.0
NH_4Cl	1.0	1.0	1.0	1.0	1.0	1.0	1.0
Liquid NH_4	1.6	1.6	1.6	1.6	1.6	1.6	1.6

Table 2(b) - Adhesive mix with different concentration of Additive

Component	Parts by weight (Pbw)						
	Level of concentration (%)						
	C_0	C_1	C_2	C_3	C_4	C_5	C_6
UF Resin Liquid 48% solid	200	200	200	200	200	200	200
Extender	0	2.0	4.0	6.0	8.0	10.0	12.0
NH_4Cl	1.0	1.0	1.0	1.0	1.0	1.0	1.0
Liquid NH_4	1.6	1.6	1.6	1.6	1.6	1.6	1.6

Table 3(a) - Properties of adhesive mix with Additive

Properties	Level of concentration (%)						
	C_0	C_1	C_2	C_3	C_4	C_5	C_6
Flow time of adhesive mix in B6 flow cup in seconds	19/B_4	9	10	13	15	16.5	18
Solid content of adhesive mix(%)	48.5	49.6	50.1	51.3	52.5	53.2	53.5
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

Table 4-Physical and Chemical characters of Additive

PHYSICAL AND CHEMICAL PROPERTIES	
Color	Off white
Fitness to Granules (%)	90.5
Moisture content (%)	2.84
Ash Content (%)	6.8
Fat Content (%)	2.4
Protein Content (%)	11.5

Table 5-Lay up and stand time standardization

Sl. no.	Additive	Level of concentration	Flow time in B_6 flow cup (sec.)	Lay out time(min)	Stand time(min)	Quality	
						Cyclic test	% wood failure
1	Additive	C_3	13	20	15	Pass standard	85
				30	15	Pass standard	85
				45	15	Pass standard	75
				60	15	Failed	40
				75	15	Failed	40

Table 6-Effect of coverage on Glue Shear Strength with respect to coverage spread = 350± 10 gms/m^2 DGL

Type of Additive	Concentration Level	Coverage / m^2 DGL/kg Liquid PF resin 50 % solid	Average Glue Shear Strength					
			Dry State		Boiling		Resistance to Mico-organism	
			Load, N	Wood Failure, %	Load, N	Wood Failure, %	Load, N	Wood Failure, %
Additive	C_0	5.60	1368	65	729	55	658	50
	C_1	5.61	1391	65	741	55	672	50
	C_2	5.72	1425	65	768	55	691	50
	C_3	5.79	1449	70	785	60	709	55
	C_4	5.83	1465	70	802	60	724	55
	C_5	5.90	1494	70	831	60	739	60
	C_6	5.94	1506	70	848	60	752	60

Table 7-Effect of coverage on Glue Shear Strength with respect to coverage spread = 300 ± 10 gms/m^2 DGL

Type of Additive	Concentration Level	Coverage / m^2 DGL/kg Liquid PF resin 50 % soln.	Average Glue Shear Strength					
			Dry State		Boiling		Resistance to Mico-organism	
			Load, N	Wood Failure, %	Load, N	Wood Failure, %	Load, N	Wood Failure, %
Additive	C_0	6.52	1341	65	708	55	616	50
	C_1	6.58	1381	65	726	55	647	50
	C_2	6.65	1409	65	738	55	667	50
	C_3	6.71	1432	70	769	60	691	55
	C_4	6.78	1450	70	781	60	706	55
	C_5	6.84	1474	70	804	60	720	60
	C_6	6.91	1499	70	823	60	747	60

Table 8-Composition of the adhesive mix used to prepare 7 ply plies (1.2 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	7
NH_4Cl	1.0	Face longitudinal	1.0
Liquid NH_4	1.6	Cross band (cross grain glued)	2.2
Extender (Additive)	3.0	Long core (Longitudinal grain	2.2
Insecticide(GLP)	0.5	Cross band (cross grain glued)	2.2
		Long core (Longitudinal grain	2.2
		Cross band (cross grain glued)	2.2
		Back (Longitudinal grain)	1.0

Table 9

Sl. No.	Type of Extender used (%)	Average Glue Shear Strength						Static Bending				Tensile Strength, N/mm2	
		Dry State		Wet State		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	Additive	1375	70	780	60	730	55	39.46	32.67	4895	2948	31.25	30.22

Table 10 -Board Pressing conditions

Parameters	Cold Pressing	Hot Pressing	
		UF	PF
Pressure	12 kg/cm^2	10.5 kg/cm^2	14 kg/cm^2
Temperature	NA	110°C	140°C
Time	30 minutes	12 minutes	12 minutes