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Low Formaldehyde Emitting Urea Formaldehyde Resin for Plywood

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A B S T R A C T

Urea formaldehyde resins were prepared in the laboratory using keratin through copolymerization reaction. Conventional Urea formaldehyde resins were also manufactured without the addition of keratin during the reaction. Through the determination of such reaction technology as molar ratio, adding a sequence of keratin and adding amount of keratin, we synthesized the low toxic urea resin. The resins were admixed with suitable hardener and the panels were made. Bond quality and formaldehyde content were evaluated. The results show that adding amount of 5 % keratin and adding keratin with the third feeding of urea is the best choice. From the studies, it was found that keratin addition reduces the formaldehyde content in the panel without affecting the strength properties.

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Introduction:

Urea formaldehyde resin with low cost, the source of the raw material, manufacturing process is simple, a high bonding strength after curing the sub lighter color, and advantages in the wood based panel widely used in industry. But in recent years, urea formaldehyde resin has high content of free formaldehyde in the production and use of the process of bonding product release formaldehyde issue has drawn increasing attention. Thus, a study to address these problems has become the main direction of urea formaldehyde.

Urea formaldehyde resins are mainly used in manufacturing panels for indoor uses. During the recent years, these have been serious concern over the formaldehyde emission rates and are reported to be higher from furniture and other panels. It has also been reported that when formaldehyde emission rates are higher one can sense eye burning. Investigation reveals that formaldehyde at higher concentration causes nasal cancer in animal and human beings.

Keratin is a structural protein of ectoderm cells, widely found in the organizational structure of organisms, such as animal hairs, egg shells, animal hoof cell medium. Normally the keratin molecule inside the disulphide bond that peptide internal and peptide occur between crosslinking, form the three dimensional network structure, so that the natural keratin difficult to dissolve. Alkaline reagent easier to open the disulfide bonds in keratin, keratin degradation of amino acids into different sizes. In which amino and urea formaldehyde glue in the reaction, thereby reducing the urea formaldehyde resin content of free formaldehyde.

Emission of formaldehyde from wood products is more where bonding agent is amino formaldehyde resin, many countries find wide application in interior uses due to its color appearance, easy workability and economic price. Although the emission of formaldehyde is mainly due to higher formaldehyde content in the resin manufacturing process, it cannot eliminate completely due to the fact that the condensation reaction such as urea formaldehyde content/emission in the panel can be minimized by the addition of higher quantity of second area which would react with the unreacted formaldehyde that may present during the reaction. Also the addition of suitable chemicals or absorbing agents to the resin during manufacture reduces the emission from panels.

Keeping in view the harmful effect on the end uses and environmental conditions keratin modified urea formaldehyde has been synthesized as a binder for the preparation of plywood. While the objective was to compare the characteristic properties of board prepared with keratin modified urea formaldehyde resin to the boards prepared with neat urea formaldehyde resin.

Materials and Methods:

Materials:

1) **Species of wood veneers for plywood:** Rotary cut veneers of thickness 1.6 mm from poplar were used in this study

2) **Raw material:** Urea, formalin, caustic soda, glacial acetic acid, and keratin powder were procured from M/s Vijays Biocytes Private Ltd, Chennai.

Experiment:

Preparation of Keratin Modified Urea formaldehyde resin (KMUF):

The weight ratio of formaldehyde with urea (F/U) selected 2.3:1 Urea is add in steps, first time 65% of the total amount of urea, the second for 25% of the total and the third of the total 10%. Added keratin was selected in the first, second and third inputs of urea, the keratin adding amount to a percentage of the total mass of urea was 3%, 5% and 7% respectively.

920 gm of formalin is taken in a beaker and the pH is adjusted to 7.5-8 by using 33 % caustic solution. The formalin is first charged into the reactor. 400 gm of urea is weighed. The first batch of urea (65%) was added to the reactor containing formalin. The pH of the reactant mixture was maintained around the 7.5-8 and heating is done to raise the temperature to 90±20C for 30 min and then added the second batch of urea. The reactants are allowed to react at pH 7.5-8 at 90±20C under reflex to ensure complete formulation of methylol urea. Further reaction is completed by adding traces of dilute acetic acid (pH 4.5-5) leading to UF polymer formation. Once desired viscosity is obtained the pH is raised to 7.5-8 until the precipitate is formed when a drop of resin is poured in water contained in a beaker.

After the precipitate formation the reaction is arrested by increasing the pH to 7.5 - 8. 5 percent of keratin powder and remaining quantity of third urea is added to the reaction mixture and cooled immediately to room temperature.

Urea Formaldehyde Resin:

1150 gms of formalin is taken in a beaker and the pH is adjusted to 7.5 - 8 by using 33 % caustic solution. The formalin is first charged into the reactor. 500 gms of urea is weighed. 90% first urea was added to the reactor containing formalin. The pH of the final reactant mixture was maintained around 7.5-8. The reactant mixture is heated and condensed at 90±20C for about 90 min. Then the pH is reduced to 4.5-5 by using glacial acetic acid and condensed at 90±20C until the precipitate is formed when a drop of resin is poured into water contained in a beaker, pH is then raised to 7.5-8. The remaining second urea of 10 % was added and the resin is cooled to room temperature (Table 1).

Table 1: Properties of resin

S. No.	Properties	Conventional Urea formaldehyde resin	Keratin modified urea formaldehyde resin
1.	Flow time of Resin in B4 flow cup	20 ec	22 ec
2.	Gelation time at 1000C	73 ec	65 ec
3.	pH of the resin	8.1	9.3
4.	Solid content (%)	49	50
5.	Water tolerance	1:3-4	1:3

Method of adhesive mixing for UF and KMUF resin:

Resin was mixed with requisite quantities of water along with extender and other additive like buffers, fortifier and hardener and mixed with the help of a mechanical stirrer in order to get homogenous adhesive (Table 2).

Table 2: Adhesive formulation for Plywood (5 Ply)

S. No	Particulars	Resin
1	Resin	300 gm
2	Maida	30 gm
3	Melamine	7.5 gm
4	Hardener	1.5 gm in requisite quantity of water
5	Liq Ammonia	3 ml

Preparation of Plywood:

In the laboratory experiments, 1.6 mm thick veneers of size 0.33 X 0.33 m were used for making 5 ply plywood. In the laboratory scale studies veneers were brushed coated. The adhesive mixture prepared with urea formaldehyde and keratin modified urea formaldehyde resin for the making interior grade plywood and condition adopted for the manufacture of plywood are given below:

Species of veneer used- poplar
Size of veneer- 33 cm X 33 cm
Thickness of veneer-1.6 mm
Moisture content of veneer- 8-10%
Hot press temp- 110±50C
Specific pressure- 12 kg/cm2
Hot press time- 6 min for 5 ply for 7 mm thick

Plywood panels were conditioned and tested for confirming to MR grade as per IS 848:2006 (Table 3) and 303: 1989 (Table 4) and the formaldehyde content emission was determined by perforator method as per IS 13745:1993 "Method for determination of formaldehyde content is by extraction method" (Table 4). The method involved boiling of the specimen in toluene, collecting the driven off formaldehyde in water and analysing by iodometric method. The formaldehyde content/ emitted is obtained by the formula given:

$$= \frac{3.0\ (V_0 - V_1)\ (100 + H)}{M}$$

= 'x' mg/100 gm of oven dry board

V_0 is the consumption in ml of 0.010 mol/I thiosulphate solution for the blank test

V_1 is the consumption in ml of 0.010 mol/I thiosulphate solution for the test

H is the moisture content of the in %

M is the mass in grams of test pieces before the extraction.

Table 3: Properties of keratin modified Urea formaldehyde resin based plywood panel

S. No	Knife test	Criteria for conformity	Result
1	MR GRADE (Moisture Resistant) Three cycles : Each cycle consisting of 3 hours at 60 ± 20C in water and thereafter drying at 65 ± 20C for 8 hours	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% of 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. 60-65% wood failure Pass standard

Table 4: Physical and Mechanical properties of MR grade plywood panel

S. No	Tests	Test Method	Requirement as per IS:303-1989	UF Resin	KMUF Resin
1	Moisture content, %	IS : 1734-1983 (RA 2003) Part-1	5 – 15	6.43	6.43
2	Resistance to water, Adhesion to plies (3 cycles of 3 hr. @ 60 ± 2° C in water & 8 hr. drying @ 65 ± 2° C)	IS : 1734-1983 (RA 2003) Part-5 and Part-6	Min. Pass Standard	Pass standard	Pass standard
3	Resistance to micro-organisms, (Adhesion to plies)	IS :1734-1983 (RA 2003) Part-5 and 7	Min. Pass Standard	Pass standard	Pass standard
4	Static Bending Strength Modulus of elasticity, N/mm2 i) Parallel to the face grain a) Average b) Minimum individual ii) Perpendicular to the face grain a) Average b) Minimum individual	IS : 1734-1983 (RA 2003) Part-11	 4000 3600 2000 1800	 6424 5789 2204 2139	 7732 7536 2255 2052
	Static Bending Strength Modulus of rupture, N/mm2 i. Parallel to the face grain a) Average b) Minimum individual ii. Perpendicular to the face grain a) Average b) Minimum individual	IS : 1734-1983 (RA 2003) Part-11	 30 27 15 13	 35.09 30.41 25.69 23.05	 56.07 49.73 30.88 29.21
5	Formaldehyde emission	IS: 13745	E1:Fc ≤8 mg/100 g E2:8 <Fc≤ 30mg/100g	4.8	1.9

Result and Discussion:

Keratin modified urea formaldehyde resins were prepared and compared with conventional urea formaldehyde. Keratin modified urea formaldehyde resin was prepared incorporating keratin during the third stage of urea addition. Plywood were made and tested for its strength properties and formaldehyde content.

From Table 1 it can be seen that the pH of the resin sample was found 8.1 and 9.3 indicating that the resins are alkaline. It can be also seen that viscosity of the keratin modified resin is higher than the conventional resin. This may be due to the inclusion of keratin in modified samples. This is in agreement with the fact that the higher the viscosity the better the reactivity of the resin. Generally the purpose of determining the gel time of a resin for a use as adhesive is ascertain the resins reactivity during hot pressing and storage. The gel time of the resin is strongly affected by its solid content and pH of the resin. The gel time of the UF resin decreased with increasing solid content. It is also depended on final pH of resin and pH also depended on percentage of solid content. More water in system diluted the curing reaction and acted as an energy barrier to resin curing. Therefore the cure rate decreased and this resulted in a long time. Thus, it is important to control the moisture content of raw materials in the manufacture of wood composite products. From Table 5 it is observed that with 5% keratin addition during the reaction the reaction has brought down the formaldehyde content to about 1.9 mg/100 gram of oven dry board. From the Table 5, it is recorded that the mechanical properties of panels meets all the requirement of IS 303 for MR grade plywood.

Conclusion:

Keratin modified urea formaldehyde resin and urea formaldehyde resins were used as binder in the preparation of plywood and boards were compared based on their mechanical properties. Keratin addition during the reaction reduces the formaldehyde content in the panels without affecting the strength properties. Internal addition of keratin after second stage of reaction gives good strength properties.

Formaldehyde content was brought down to the minimum of 1.9 mg/100gms of oven dry plywood by using the resin developed under this project compared to that of conventional urea formaldehyde resin.

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