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SPENT SULFITE LIQUOR-UREA FORMALDEHYDE RESIN: A POTENTIAL BINDER FOR PARTICLEBOARD

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Spent sulfite liquor (SSL), obtained from the paper industry based on the grassy pulp material was concentrated to 50% total solids and condensed with sulfuric acid at pH 3. The modified SSL was copolymerized with different amounts of urea formaldehyde (UF) and used as a binder for exterior grade particle board. Various types of poplar boards were prepared i.e. (i) by using different composition of SSL: UF. (ii) at different press time and (iii) at different press temperatures. The properties of these boards meet the requirement imposed for international standards.

Key words: Grassy pulp materials, SSL-UF resin, Particleboard.

Introduction

The presence of lignin in the waste product of pulp mills has made it an attractive raw material for making adhesives over since the begining of sulfite pulping process of wood. By this process only half of the wood is used as pulp while the other half forms SSL which is generally discharged into the rivers creating water pollution problems. Only 20% of SSL is used for various purposes [1], but the technical utilization of lignin on a large scale is still an unsolved problem. As lignin molecule has structural similarities with phenolic resin, therefore, many attempts have been made to cross link lignin molecule via phenolic reactions. Such as epoxide [2], isocyanates [3-5], ethyleneimines [6] and phenolformaldehyde [1,7,8].

The rapid increase in particleboard production during the last few decades demand an adhesive that is cheap, available in large quantities and independent of crude oil.

Beside meeting these requirements, lignin does not release formaldehyde which has become serious problem with urea formaldehyde (UF) adhesives. Most of the world paper mills are using wood as a pulp material. SSL obtained from these mills has already been utilized as a binder for particleboard. Some of these investigations used SSL as such [9] while the others used it in combination with phenol formaldehyde [9] and urea formaldehyde [10]. No such studies have been done in this country where SSL is obtained from paper mills based on grassy pulp mateiral (bagasse, Kaigrass and wheat straws).

In our previous study [11], we prepared particleboard from SSL alone. The results showed strength properties well comparable to the boards prepared from urea formaldehyde. The physical properties of the board, although passed the requirements imposed from American Standard [12], but still its water absorption was not comparable to the UF boards prepared during the study.

The aim of the present study was two fold (i) to prepare particleboards from SSL, co-polymerised with different ratios of UF and, (ii) to find out optimum press conditions for the production of an econimical board.

Experimental

SSL was obtained from Adamjee Paper and Board Mills, Nowshera, NWFP, Pakistan where sodium based sulfite pulping process is being used. The pulping material is mainly composed of bagasse *(Saccharum officinarum)* Kaigrass *(Saccharum spotarum)* and wheat straws *(Triticum vulgare)*.

Condensation of SSL. SSL was concentrated to 50% total solid at 50° under reduced pressure. Condensation was carried out with concentrated sulfuric acid and pH of the liquor was adjusted to 3.

Chipboard preparation. Medium density boards ($0.7 gm/cm^3$) having a size of 50 x 55 x 1.27 cm were prepared from poplar chips. Condensed SSL and UF (60% total solids) were sprayed separately at an air pressure of 2 kg/cm^3 followed by 2 mins. blending time in each case. A fixed ratio 1:9 (w/w on ovendry basis) of the glue (SSL and UF) to chip was used. Following three types of the board were prepared.

(i). Chipboard from SSL, in combination with different ratio of UF (SSL:UF, 90:10, 80:20, 70:30 etc.). The boards were pressed at 30 kg/cm^2, 210° for 16 mins.

(ii). Boards from SSL and UF at a fixed ratio of 70:30. Boards were pressed at 30 kg/cm^2 and 210° for different press times (8-16 mins.).

(iii). Boards were prepared from SSL and UF at a fixed ratio of 70:30. The boards were pressed at 35kg/cm^2 for 12 mins. Temperature of the press was changed in the range of 180-220°. A 0.5% paraffin emulsion was also used to act as water repellant. The boards were conditioned for one week at room temperature and tested according to American Society of Testing Materials [13]. The results reported are the average of values obtained for five specimen from each board.

Results and Discussion

Effect of glue compositin on the properties of chipboard. Figure 1a represents the strength properties of the boards at various glue compositions of SSL and UF. Modulus of rupture (MOR) remained almost constant upto SSL concentration of 40%. Further decrease in SSL concentration resulted in a slight increase of MOR, while modulus of elasticity (MOE) increased with the decrease of SSL upto 70% concentration and remained constant upto 40% of SSL and then decreases with the decreasing amount of SSL. It is clear from Fig. 1b that thickness swelling (T-S) at 2 and 24 hrs, and water absorption (W-A) (Fig. 1c) at 2 hrs. soaking decreases with the gradual addition of UF upto 30%. Further addition of UF showed a constant response upto 90% addition of UF, while water absorption for 24 hrs. soaking gave a constant response beyond 50% addition of HF in the resin. Linear expansion (L-E) at 90% relative humidity (Rh) for 24 hrs. did not give any conclusive results. These results indicate that MOR of the board is not much affected by UF addition, and MOE of the board is even better with 40% SSL in the resin, while the boards became water resistant by 30% addition of UF in the resin. The results were compared with neat UF board prepared during the study. It is interesting to note that properties of the boards prepared from SSL-UF were superior than neat UF in

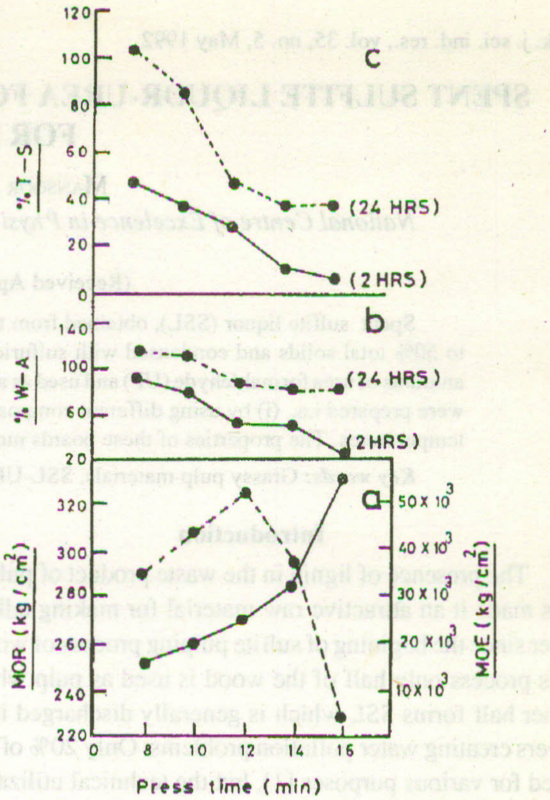

Fig. 2(abc). Effect of press time on (a) MOR and MOE (b) Water absorption (2 and 24 hrs. soaking) and (c) Thickness swelling (2 & 24 hrs soaking) of poplar boards.

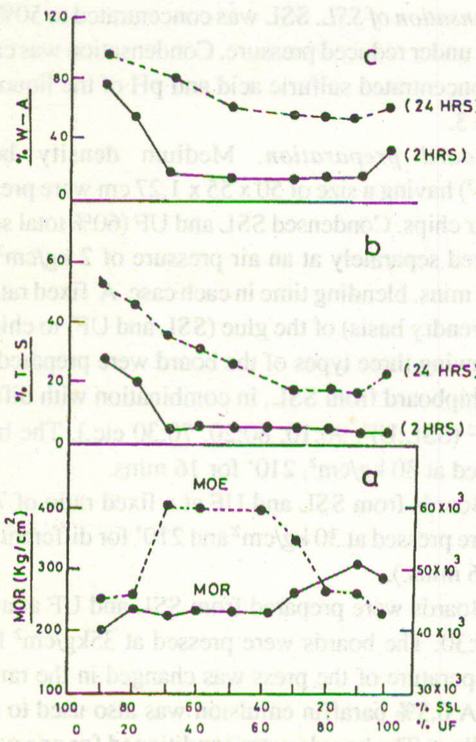

Fig. 1(abc). Effect of glue composition on (a) MOR and MOE (b) Thickness swelling (2 & 24 hrs, soaking) and (c) Water absorption (2 & 24 hrs, soaking) of poplar boards.

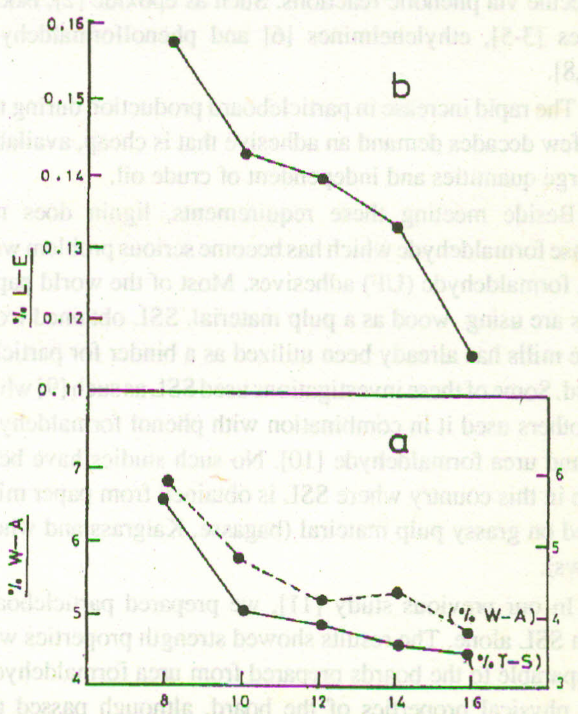

Fig. 3. Effect of press time on (a) Water absorption and thickness swelling (b) Linear expansion (90% Rh, 24 hrs) of poplar boards.

each respect. This may be due to the complex structure of SSL-UF which is copolymerized during condensation reactions giving a highly branched polymer which may be more water resistant and have high binding affinity to the board particle. T-S of the board at the glue composition 70:30 of SSL to UF was compared with German Standard DIN 68761 [14], while the other properties were compared with the American Standard CS236 [12] (Table 1). All the results were found to be equally good.

Effect of press time of the properties of chipboard. Figure (2a) represents the strength properties of the boards prepared from 70:30 of SSL to UF at various press time. MOR of the boards increases by increasing press time upto 12 mins. Further increase of press time decreases the MOR, while MOE increases with the increase of press time. It is obvious from Fig. 2b,c that both T-S and W-A decrease with the increase of press time and resulted in a more water resistant board.

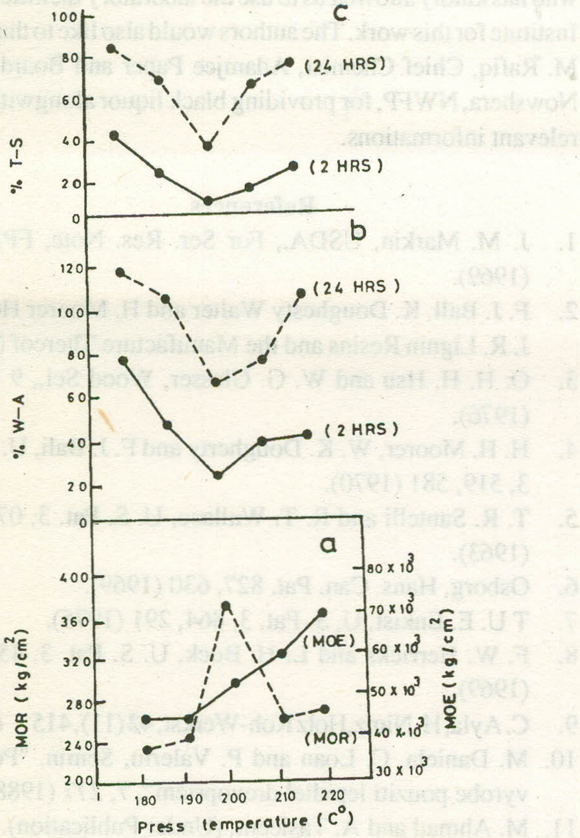

Fig. 4(abc). Effect of press temperature on (a) MOR and MOE (b) Water absorbed (2 & 24 hrs soaking) and (c) Thickness swelling (2 & 24 hrs soaking) of poplar board (Glue comp. 70:30 of SSL to UF, presss time 12 mins. pressure 35 kg/cm²).

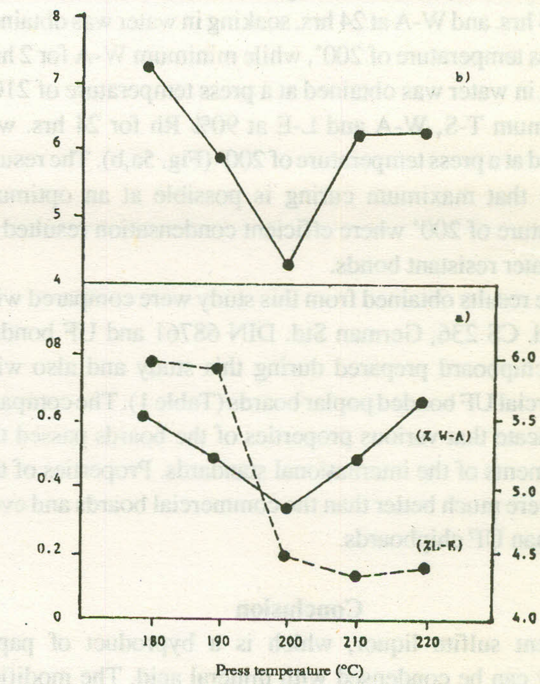

Fig. 5(a,b). Effect of press temperature on (a) Lar expansion and water absorption and (b) Thickness swelling (90% RH, 24 Hrs) of poplar boards. (Glue composition 70:30 of SSL to UF, press time 12 mins., pressure 35 kg/cm²).

Table 1. Comparison of Board Properties with Standard and Commercial Boards.

Properties	Am.Std. CS 236 (1966)	Ger. Std. DIN 68761 (1966)	a	b	c	d	e	f
MOR kg/ cm²	112	193.0	294.00	305.00	229.00	228.00	323.00	367.00
MOE kg/cm²	17577	19827.0	–	42074.00	26412.00	55504.00	26063.00	53941.00
%WA 2 hrs., soaking	160 (1 hr.)	–	–	39.86	94.58	21.24	50.70	18.49
%WA 24 hrs., soaking	–	–	–	69.69	118.76	84.00	87.14	59.11
%TS 2 hrs., soaking	–	6.0	–	2.64	20.79	4.90	27.00	5.14
%TS 24 hrs., soaking	–	–	–	23.87	25.01	35.00	43.31	31.07
%TS 90%RH	–	–	–	4.184	3.509	53.00	5.21	4.85
%ST 90% RH	–	–	4.01	2.962	1.317	–	3.89	4.33
%LE 90% RH	3.35	–	0.458	0.147	0.169	–	0.139	0.187

(a). UF bonded poplar board prepared in Pakistan Forest Institute, Peshawar; (b). UF bonded poplar board prepared during this study; (c). UF bonded commercial poplar boards; (d). SSL: UF (70:30%) poplar board pressed at 30 kg/cm², 210° for 16 mins; (e). SSL:UF (70:30%) poplar board pressed at 30kg/cm², 210° for 12 mins; (f). SSL : UF (70:30%) poplar board pressed at 35 kg/cm², 200° for 12 mins.

Figure 3 showes that T-S, W-A and L-E at 90% Rh for 24 hrs.. decreases with the increase of press time. All properties of the board were found to meet the requirement of CS 236 [12].

Effect of press temperature on the properties of chipboard. Figure (4a) shows the strength properties of chipboard prepared from a glue ratio 70:30 of SSL UF at various press temperature. Maximum MOR was found at a press temperature of 200°. Low MOR at values at 180 and 190° may be due to incomplete curing of the resin, while low MOR at 210 and 220° may be due to charring and fiber destruction at these high temperatures. It is shown in Fig. 4b,c that a minimum T-S at 2 and 24 hrs. and W-A at 24 hrs. soaking in water was obtained at a press temperature of 200°, while minimum W-A for 2 hrs. soaking in water was obtained at a press temperature of 210°. A minimum T-S, W-A and L-E at 90% Rh for 24 hrs. was observed at a press temperature of 200° (Fig. 5a,b). The results indicate that maximum curing is possible at an optimum temperature of 200° where efficient condensation resulted in more water resistant bonds.

The results obtained from this study were compared with Am. Std. CS 236, German Std. DIN 68761 and UF bonded poplar chipboard prepared during this study and also with commercial UF bonded poplar boards (Table 1). The comparison indicate that various properties of the boards passed the requirements of the international standards. Properties of the board were much better than the commercial boards and even better than UF chipboards.

Conclusion

Spent sulfite liquor, which is a byproduct of paper industry can be condensed with mineral acid. The modified SSL can be copolymerised with different amounts of UF and can be used as a binder for particle board industry. It is concluded that water resistance of the boards remained constant at and above 30% UF in the glue while MOR of the board is not much affected with the amount of UF. The press cnditions for boards of maximum strength and resistance were found to be pressure 35 kg/cm^2, temperature 200° and press time 12 mins.

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References

1. J. M. Markin, USDA., For Ser. Res. Note, FP2-0206 (1969).
2. F. J. Ball, K. Doughesty Walter and H. Moorer Howard, J. R. Lignin Resins and the Manufacture Thereof (1962).
3. O. H. H. Hsu and W. G. Glasser, Wood Sci., **9** (2), 97 (1976).
4. H. H. Moorer, W. K. Dougherty and F. J. Bali, U. S. Pat. 3, 519, 581 (1970).
5. T. R. Santelli and R. T. Wallace, U. S. Pat. 3, 072, 634 (1963).
6. Osborg, Hans. Can. Pat. 827, 630 (1969).
7. T U. E. Enkist, U, S. Pat. 3, 864, 291 (1975).
8. F. W. Herricks and L. H. Bock, U. S. Pat. 3, 454, 508 (1969).
9. C. Ayla, H. Nimz, Holz Roh-Werkst, **42** (11), 415 (1984).
10. M. Daniela, C. Loan and P. Valeriu, Semin. "Pokroky vyrobe pouziti lepidiel drevopriem", 7, 277 (1988).
11. M. Ahmad and A. Tasleem, (Under Publication).
12. American Standard CS. 236 (1966).
13. American Society of Testing and Materials (ASTM), (1965).
14. German Standard DIN 68761 (1966).