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Reaction pH of Urea-formaldehyde Resins as Related to Strength Properties of Southern Pine Particleboard*1

Chung-Yun HSE*2

サウザンパイン・パーティクルボードの材質におよぼすユリア・ホルムアルデヒド樹脂の反応 pH の影響*1

Chung-Yun HSE*2

反応混合物の pH を最初の1時間アルカリ側 (4水準) に，その後弱酸 (3水準) に調整する組合わせで，生成樹脂によるボード材質を検討した。

中性 (pH 7) または弱アルカリ性 (pH 8) でつくられた樹脂は，強アルカリでつくられた樹脂より高い接着強度を示した。反応初期 pH が8または9のとき，後段の酸性側の pH 変化は樹脂接着力にほとんど影響しなかった。しかし初期 pH 7 の場合，酸性反応時の pH の増加につれて，また初期 pH 10 の場合は酸性反応時の pH の減少につれて接着力は向上した。初期 pH 7 と酸性反応時の pH 5.8 の組合わせの樹脂は，最高の接着力を示した。

Twelve urea-formaldehyde resins were prepared with factorial combinations of 4 alkaline and 3 acidic reaction phases; *i.e.*, the reaction mixture was adjusted to pH 7, 8, 9, or 10 for the first hour and then made weakly acid to pH 5.8, 4.8, or 3.8.

On average, the resins formulated at neutral (pH 7) or weakly alkaline (pH 8) conditions made boards of higher internal bond and modulus of rupture than resins formulated at stronger alkaline conditions. Change in acidic phase had little effect on resin adhesion strength when the initial pH was 8 or 9, but caused increases at initial pH 7 and decreases at initial pH 10. Acid phase pH 5.8 in combination with initial pH 7 yielded the resin with the best adhesion strength but required very long formulation time.

1. INTRODUCTION

In the formulation of urea-formaldehyde adhesives, the initial reaction products are methylol compounds, formed under neutral or weakly alkaline conditions. The methylol ureas are not resinous. At an appropriate time, therefore, the reaction mixture is made weakly acid to promote condensation leading to resin formation. Both the rate of reaction and the type of polymer formed can be controlled by varying the pH. The study reported here explored the relationship between reaction pH and properties of southern pine particleboard.

*1 Received Dec. 24, 1973.

*2 Southern Forest Experiment Station, Forest Service, U.S. Department of Agriculture, Pineville, Louisiana

2. PROCEDURE

Resins were formulated with factorial combinations of 4 alkaline and 3 acidic phases. Specifically, the reaction was adjusted to pH 7, 8, 9, or 10 for the first hour and then to pH 5.8, 4.8, or 3.8. The resulting 12 resins were replicated.

Adhesion properties were determined by measuring internal bond strength, modulus of rupture (MOR), and modulus of elasticity (MOE) of boards. Details of resin and board preparation were similar to those described in the preceding paper.[1)]

Data were tested by variance analysis at the 95 % level of probability.

3. RESULTS AND DISCUSSION

Individual values of board properties varied widely, ranging from 2.99 to 10.39 kg/cm^2 for internal bond; from 95.6 to 140.6 kg/cm^2 for

MOR, and from 16050 to 20700 kg/cm² for MOE. Averages for the 12 resins are shown in Table 1.

Internal bond and MOR differed with changes in the alkaline phase. The change of acidic phase was significant only in the MOE measurement.

As Table 2 shows, resin formulated at neutral (pH 7) or weakly alkaline (pH 8) conditions produced boards with higher internal bond and MOR than resins formulated at stronger alkaline conditions (*i. e.*, pH 9 and 10). This result may be largely attributed to the conversion of formaldehyde to methanol and formic acid by the Cannizzaro reaction. The reaction is greatly increased at high reaction pH,[3] and may effectively reduce the CH_2O/urea ratio. As shown in the previous paper, a decrease in the ratio resulted in reduced board quality.

MOE was significantly related only to pH of the acidic phase; MOE increased as pH decreased (Table 2).

The interaction of the alkaline and acidic phases was significant (Fig. 1). Change in pH of acidic phase had only minor effect on MOR and internal bond when the initial pH was 8 or 9. If initial pH was 7 or 10, MOR and internal bond increased or decreased as pH of the acidic phase increased. This pH effect was especially important in the weakly acid range of 5.8, *i. e.*, pH 5.8 in combination with initial pH 7 yielded the resin with the best strength.

To check on batch-to-batch consistency, resin viscosity was measured with a Cannon-Fenske opaque viscometer at 30-minute intervals after acidic adjustment. It was noted that viscosity as a function of reaction time was reproducible but differed for resins formulated by various combinations of pH in alkaline and acidic phases. It accelerated as pH of acidic phase decreased (Fig. 2). Total reaction time varied substantially, therefore, and became significant in optimizing the pH condition. At pH 5.8, where adhesion strength was greatest, the rate of condensation

Table 1. Strength properties of particleboards assembled with resins formulated under various pH conditions.

Alkaline phase	Acidic phase	IB	MOR	MOE
		kg/cm²		100 kg/cm²
pH 10	5.8	3.85	106.5	223.8
	4.8	5.24	116.6	221.7
	3.8	5.41	124.4	216.3
pH 9	5.8	4.75	110.9	218.3
	4.8	4.40	111.1	206.6
	3.8	5.17	112.2	200.6
pH 8	5.8	6.42	121.8	231.5
	4.8	6.45	126.4	228.9
	3.8	4.58	124.2	203.7
pH 7	5.8	8.35	127.6	221.2
	4.8	8.31	116.4	192.6
	3.8	5.30	112.3	206.7

Table 2. Effects of alkaline and acidic phases on board quality.

		Internal bond	MOR	MOE
		kg/cm²		100 kg/cm²
Alkaline phase	7	7.32	118.8	
	8	6.52	124.2	
	9	4.77	111.4	
	10	4.83	115.8	
Acidic phase	3.8	—		224.3
	4.8			208.8
	5.8			203.9

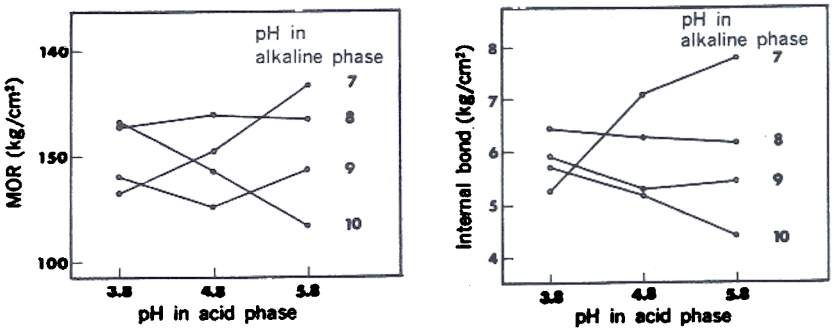

Fig. 1. Interaction of pH in alkaline and acidic phases with internal bond and MOR of particleboard.

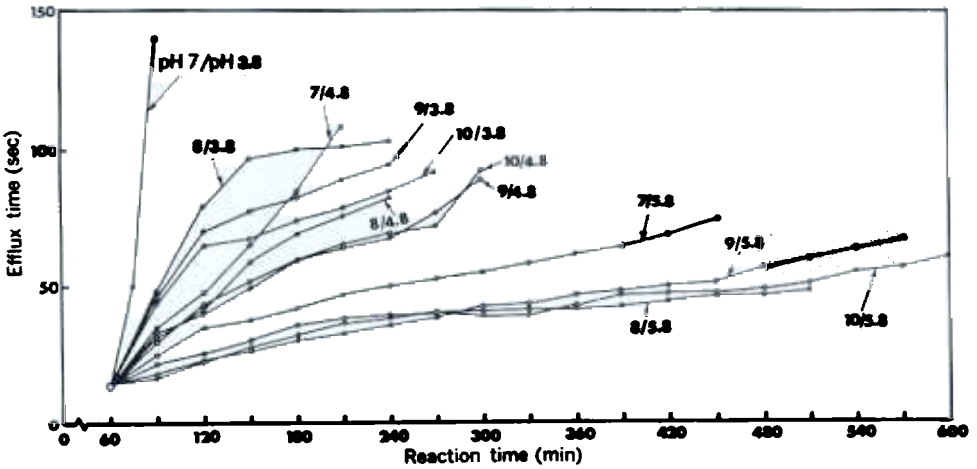

Fig. 2. Relationship between resin viscosity and reaction time at various pH levels in alkaline and acidic phases.

(measured as viscosity increase) was so slow as to be economically infeasible in commercial practice.

The poor performance of the resin formulated at pH 7/3.8 may be due to its extremely fast condensation. The rate to condensation affects the chemical homogeneity; *i.e.*, the faster the rate, the less homogeneous is the resin with respect to both molecular weight and molecular constitution. In an inhomogeneous resin not all the molecules of different fractions will react, and those that do will not react similarly. And, in the curing process, the lower molecular fraction, having to start growing from a smaller size, takes longer to catch up. It is likely, therefore, that gel formation and curing properties may be adversely affected by rapid condensation.

REFERENCES

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