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Effect of Synthesis Variables on Tack in Urea-Formaldehyde Resin

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The degree of tack—the ability of resin to adhere to another surface under light pressure and over short time—in urea-formaldehyde resin varies throughout the particleboard manufacturing process and affects mill maintenance and board quality. A three-part study was conducted to evaluate the effects of certain synthesis variables— molar formaldehyde : urea ratio, ingredient concentration, reaction pH, and reaction catalyst—on tack of urea-formaldehyde resin in particleboard manufacturing. Southern pine particleboard mats were formed and prepressed in the laboratory, and tack was evaluated with the push-off method. Reaction pH significantly affected tack and viscosity, which were highly related. On the average, reducing formaldehyde : urea ratio and ingredient concentration increased tack; these two variables may be significant to tack development and resin morphology. Reaction catalyst did not affect tack or other resin morphology. Reaction catalyst did not affect tack or other resin characteristics, but resin surface tension contributed significantly to tack development. Further study should better define optimum manufacturing time and make tack development in particleboard manufacturing more predictable.

KEY WORDS Particleboard; reaction catalyst; reaction pH; tack; urea-formaldehyde resin; effect of resin-synthesis variables.

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INTRODUCTION

In the particleboard industry, resin tack—the ability of resin to adhere to another surface under light pressure and over short time[1]—plays an important role in production and ultimately in panel characteristics. Maximum resin tack needs to develop as the particles are felted. Premature tack development or excessive tack in the blenders can cause serious maintenance problems and downtime. However, latent tack development or lack of tack can make the prepressed mat difficult to convey and result in high panel-rejection rates.

Because the factors influencing tack and its development in urea-formaldehyde resins as applied to southern pine particleboard have not been understood, we investigated the relationship between tack and several key resin-synthesis variables in a three-part experiment. Southern pine particleboard mats were formed and prepressed in the laboratory with laboratory-prepared resin, and tack was evaluated with the push-off method. More specifically, we measured the effects on tack of molar formaldehyde:urea ratio and ingredient concentration (Part I), reaction pH (Part II), and reaction catalyst (Part III).

LITERATURE BACKGROUND

Generally, tack is considered a low-order adhesion. Whether the adhesion force of tack is a function of separation force[2] as identified by the Stefan equation, morphology of the tack surface,[2] or viscoelastic response to dynamic deformation[1,2] is a point of argument. Undoubtedly, the controversy exists because tack is a function of several resin morphological properties.[3,4,5] Resin properties such as elasticity and viscosity[3,6] as well as molecular characteristics involving degree of polymerization and cross linking[7,8] have been identified as key features to "stickiness".

Methods of measuring tack have been reported by numerous investigators.[1,2,3,6,9,10,11] Some[1,2,9] found that the probe test provided a reasonable approach to measurement. Although this test is simply conducted, probe topography and composition can influence tack measurement.[1] Push-off and mat separation tests, both of

which require laboratory-formed prepressed mats, are commonly used by industries related to the particleboard industry. The disadvantages of both methods have been enumerated.[10] However, these tests apparently provide the closest approximation to end-use conditions in the particleboard industry and therefore have gained acceptance.

The particleboard industry also is concerned with absorption of the solvent by the wood substrate. Studies using veneer contact faces on metal anvils in a glueability tester[10] showed that tack development and duration partly depend on how water is dissipated through absorption, adsorption, or evaporation, that the time required to develop or lose tack is about equal, and that resin tackiness does not change as rapidly in a formed mat as in loose resin-coated particles because of reduced evapotranspiration.

How molar ratios of urea-formaldehyde resin and reaction pH levels and catalysts affect strength properties of southern pine particleboard have been investigated.[12,13,14] However, tack was not evaluated at that time. Data from those tests inidicated that the variables which affect resin bond properties might also be related to tackiness. More recently, the chemical structure of urea-formaldehyde resins[15,16,17] and a proposed colloidal dispersion model[5,17] lend further support to the hypothesis relating tack to resin-synthesis variables.

EXPERIMENTAL METHODS

Resin synthesis

Urea in pelletized form was introduced to the resin kettle, to which formaldehyde as a 37% solution and distilled water were added. Temperature was increased at a maximum rate to 95°C with a thermostatically-controlled steam coil while the resin was cooked for 1 hour in an alkaline phase and then 4 hours in an acidic phase or until a viscosity of 100 seconds was achieved, as measured with a standard capillary viscometer. After cooking, the resin was quickly cooled to 25°C, and pH was adjusted to 7.5 to suspend the reaction.

Early laboratory tests demonstrated that resin solids content would be critical to tack. The 40% solids level, based on ovendry

weight, was selected as a standard which represented a good comprise between industrial reality and experimental convenience. Solids content was adjusted by gently heating the evacuated reaction vessel and distilling the off-gassed solvent.

The resin was generally allowed to sit overnight at room temperature before viscosity and surface tension were determined, and during this time many of the resins became cloudy, a suggestion of colloidal behavior. Viscosity was measured by a Brookfield† viscometer with spindle no. 1 at 50 rpm. Surface tension was tested by a Fisher surface tensiometer equipped with a 6-cm platinum ring.

Mat fabrication

Particles from a Johns Manville Corporation particleboard mill in Monroe, Louisiana, were fractionated by a vibrating shaker with three sieves (2, 4, 8/in.) and a pan. Particle size distribution by fraction (ovendry basis) was 0.01, 0.50, 7.2, and 92.3%, respectively.

A sufficient quantity of resin to yield two mats with 6% resin content by weight (ovendry basis) was acidified to pH 4.4 with an HCl reagent and sprayed at 40 psi into a drum mixer containing particles. One mat was formed with the blended particles immediately after mixing (0 min. assembly time) in a mat-forming box divided in half by a plastic film (removed after prepressing) to produce two samples from each mat. A second mat was formed in the same fashion with the remaining blended particles 20 min. later (20 min. assembly time). All mats were prepressed at 100 psi to ½-in. stops.

Tack testing apparatus

A glueability tester was briefly tried but discarded because it required extensive refinement. Instead, tack was tested with a push-off apparatus (Figure 1).

The apparatus was constructed on a metal lathe. A long threaded rod was set in the chuck of the lathe. A threaded fitting was turned

† The mention of trade names or commercial products does not constitute endorsement by the authors or Oregon State University.

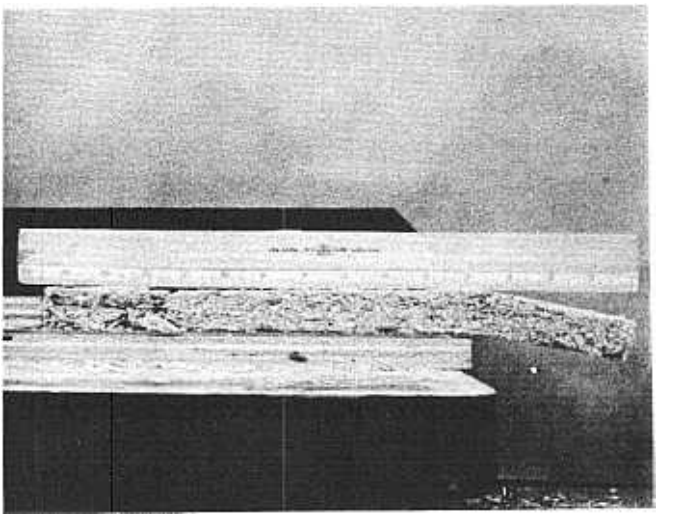

FIGURE 1 Apparatus for testing tack of urea-formaldehyde resin with the push-off method.

onto the rod, to which a push bar was affixed. Because the lathe could be adjusted to turn at various speeds, the push-off rate also could be varied.

A Plexiglas® base and sheet metal guide with high-gloss paint finish were mounted on a $\frac{3}{4}$-in. plywood platform; the guide, attached to one side of the Plexiglas®, maintained sample alignment. Plexiglas® provides a relatively low friction coefficient and a sharp edge for mats to break over. The platform assembly was aligned with the push bar. During the push-off test, a prepressed mat was slid across and then off the edge of the Plexiglas®; tack was expressed as the length of mat overhanging the edge when the mat broke off.

Preliminary refinement of the push-off apparatus focused on push-off rates and variability. Results showed 6 in./min. was a good test rate and that a 15% coefficient of variation should be expected.

Part I: Molar ratio and ingredient concentration

Resin preparation and characteristics have been related to glue-bond quality in southern pine particleboard.[12] The ratio of

formaldehyde to urea (F:U ratio) was cited as an important factor in molecular development.[2,17] However, resins with F:U ratios above 2:1 generally suffer inferior moisture resistance unless further modified. Since earlier studies by Hse,[12,13,14] molar ratios for industrial resins have been reduced in response to problems with latent release of free formaldehyde. The objective of Part I was to determine whether molar F:U ratio at various ingredient concentrations affects tack. The relatively high molar ratios of this study reproduced those by Hse[12,13,14] and were aligned with those of another recent investigation.[17]

To test this objective, a 3×2 factorial experiment was designed: three molar F:U ratios (2.3:1.0, 1.9:1.0, and 1.5:1.0) were combined factorially with two ingredient concentrations (50 and 35%) for a total of six resin cooks (treatments). Each cook was replicated twice, and the resin-production sequence was randomized within each replication. Alkaline phase pH, 8.0, was controlled with a reagent of 20% NH_4OH and 50% NaOH mixed 1:1 by volume. Acidic phase pH, 5.0, was controlled with a 20% HCl reagent.

Part II: Reaction pH

Urea-formaldehyde resins are synthesized through a condensation reaction between excess formaldehyde and urea in the presence of acid or base.[2] Reaction pH was shown to be an important variable in resin synthesis and ultimately affected strength properties of southern pine particleboard.[13] The objective of Part II was to determine whether alkaline and acidic phase reaction conditions and their interactions affect tack.

To test this objective, a 4×3 factorial experiment was designed: four pH levels in the alkaline phase (7.0, 8.0, 9.0, and 10.0) were combined factorially with three pH levels in the acidic phase (5.8, 4.8, and 3.8) for a total of 12 resin cooks. Each cook was replicated twice, and the resin-production sequence was randomized within each replicate. Resin was formulated with a molar F:U ratio of 2.3:1.0 at an ingredient concentration of 50%. The same reagents as described in Part I were used to control pH in each reaction phase. Laboratory and testing procedures were as previously enumerated.

Part III: Reaction catalyst

Catalyst selection to optimize urea-formaldehyde resin performance in southern pine particleboard has been previously studied.[14] Reaction catalysts in resin production were shown to affect panel properties significantly. In addition, synthesis time to the reaction end point was related to the catalyst employed. The objective of Part III was to determine whether reaction catalyst influences tack.

To test this objective, a 4×3 factorial experiment was designed: four acidic phase catalysts (HCl, CH_3COOH, NH4Cl, and H_3PO_4) were combined factorially with three alkaline phase catalysts (NaOH/HMTA, NaOH/TEA, and NaOH) for a total of 12 resin cooks. HMTA (hexamethylenetetramine) and TEA (tetraethanolamine), when not combined with NaOH, have resulted in condensation rates too fast to control.[14] Therefore, HMTA and TEA were mixed with NaOH, which is known to promote a comparatively slow reaction. Each cook was replicated twice, and the resin-production sequence was randomized within each replication. Resin was formulated with a molar F:U ratio of 1.7:1.0 at an ingredient concentration of 50%. The same reagents as described in Part I were used to control pH in each reaction phase. Laboratory and testing procedures were as previously enumerated.

RESULTS AND DISCUSSION

Part I: Molar ratio and ingredient concentration

The resins prepared at 1.5:1.0 F:U ratio at both ingredient concentrations were not sufficiently stable for laboratory testing (they gelled during short-term cold storage). Thus, tack testing in Part I was restricted to the other two F:U ratios. Loss of two treatments from the experimental design reduced the number of available observations below a level reasonable for formal analysis of variance.

Syntheses at 35% ingredient concentration yielded resins with solids contents of about 25% by weight (ovendry basis) before solvent extraction; syntheses at 50% ingredient concentration yielded resins with roughly 35% solids contents before solvent

extraction. This meant that substantially more solvent had to be removed from the low- than the high-solids resins to achieve the standard 40% level.

The F:U molar ratio influenced viscostiy, surface tension, and tack characteristics of the cooked resins (Table I). When only the main effect of molar ratio was considered, average tack was 6.0 (1.9:1 ratio) and 2.6 in. (2.3:1 ratio) for 0-min. assemblies and 6.8 (1.9:1 ratio) and 3.1 in. (2.3:1 ratio) for 20-min. assemblies. Viscosity averaged 59.6 cps (1.9:1 ratio) and 41.0 cps (2.3:1 ratio). Surface tension averaged 63.8 dynes/cm (1.9:1 ratio) and 56.2 dynes/cm (2.3:1 ratio).

The effects of ingredient concentration became apparent after results for viscosity, surface tension, and tack were averaged over the molar ratios. Average tack was 5.2 in. (35% concentration) and 3.4 in. (50% concentration) at 0-min. assemblies, 6.0 in. (35%) and 3.8in. (50%) at 20-min. assemblies. Viscosity averaged 55.5 cps (35%) and 45.2 cps (50%), and surface tension was unaffected by ingredient concentration.

Using advanced analytical methods, Rammon *et al.*[15] demonstrated that higher molar F:U ratios increased molecular branching and slowed condensation. Setting the results of this investigation within the context of those of Rammon *et al.*[15] suggests that viscosity and tack were reduced by the increased molecular branching induced by the higher molar ratios. In addition, when the

TABLE I

Effect of molar ratio of formaldehyde to urea (F:U) on tack of urea-formaldehyde resin and its characteristics at two ingredient concentrations and two assembly times, as measured by the push-off method

Independent variables		Resin characteristics			Tack (in.) at two assembly times ($n = 4$)	
F:U ratio	Ingredient concentration (%)	Solids (%)	Viscosity[a] (cps)	Surface tension (dynes/cm)	0 min.	20 min.
1.9:1.0	35	40.6	65.0	63.6	7.4	8.0
	50	40.5	54.2	64.0	4.6	5.5
2.3:1.0	35	39.8	45.9	57.7	3.0	4.1
	50	41.6	36.1	54.8	2.2	2.1

[a] Measured by Brookfield viscometer, spindle no. 1, 50 rpm.

results for resins of higher and lower ingredient concentrations were compared, the negative influence of excess levels of formaldehyde on reaction kinetics and resulting resin characteristics, as noted by Rammon *et al.*,[15] were shown herein.

Part II: Reaction pH

The relationship between resin viscosity and reaction times at various pH levels in both the alkaline and acidic phases was not different from that previously reported.[13] The reaction was very rapid for the 7.0–3.8 resin cook, which was terminated because of viscosity criteria. Few of the other resin cooks were terminated because of viscosity.

Viscosity was sensitive to pH level (Table II). In general, viscosity decreased with increasing alkalinity in the alkaline phase and increased sharply with increasing acidity in the acidic phase. Surface tension was unaffected.

Tack at 0-min. assembly time differed significantly ($\alpha = 0.05$, paired t-test) from tack at 20-min. assembly time. Therefore, the tack data were separated by assembly time for all analyses.

TABLE II
Effect of alkaline and acidic phase reaction pH on tack of urea-formaldehyde resin and its characteristics at two assembly times, as measured by the push-off method

Reaction pH		Resin characteristics			Tack (in.) at two assembly times	
Alkaline phase	Acidic phase	Solids (%)	Viscosity[a] (cps)	Surface tension (dynes/cm)	0 min.	20 min
7.0	5.8	42.2	38.2	62.4	1.72	2.10
	4.8	41.4	62.9	68.2	5.00	5.72
	3.8	41.2	86.7	60.7	6.25	6.50
8.0	5.8	40.7	30.4	64.6	1.25	1.66
	4.8	42.0	47.0	57.6	2.84	3.47
	3.8	41.6	63.5	59.6	5.09	6.06
9.0	5.8	41.0	29.6	61.6	1.38	1.56
	4.8	41.8	41.5	56.0	2.25	2.72
	3.8	42.0	57.6	65.8	5.44	4.84
10.0	5.8	41.0	27.0	61.5	1.38	1.59
			50.3	60.4	2.22	2.44
			41.8	57.2	3.94	4.28

[a] Measured by Brookfield viscometer, spindle no. 1, 50 rpm.

Regardless of assembly time, both alkaline and acidic phase pH significantly affected tack ($\alpha = 0.05$, analysis of variance). However, the alkaline-acidic phase interaction was not significant.

At 0-min. assemblies, tack decreased with increasing alkalinity and increased with increasing acidity (Figure 2A, B). At 0 min. in the alkaline phase, tack at the pH 7.0 level differed significantly ($\alpha = 0.05$, Duncan's Multiple Range Test) from that at pH 8.0, 9.0, and 10.0 (Table III). However, mean tack values at pH 8.0, 9.0,

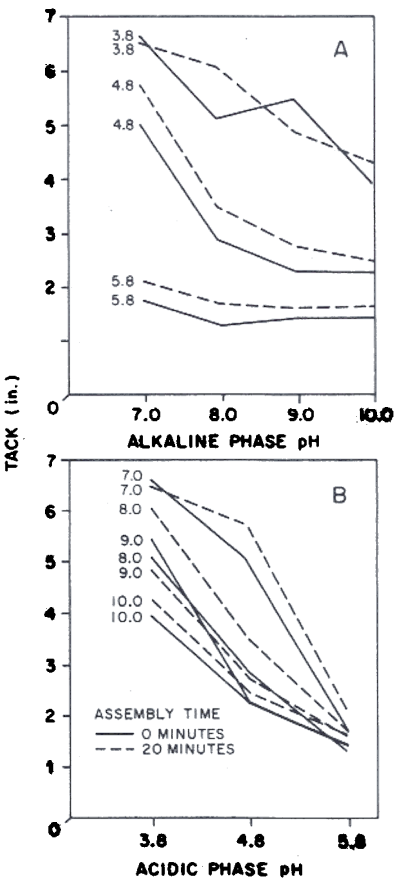

FIGURE 2 Effect of alkaline (A) and acidic (B) phase pH on tack of urea-formaldehyde resin at two assembly times, as measured by the push-off method.

TABLE III
Effect of alkaline ($n = 6$) and acidic ($n = 8$) phase reaction pH, by phase, on tack of urea-formaldehyde resin at two assembly times, as measured by the push-off method

Phase, by pH level	Mean tack (in.) at two assembly times[a]	
	0 min.	20 min.
Alkaline		
pH 7.0	4.45a	4.78a
8.0	3.06b	3.73ab
9.0	3.02b	3.04b
10.0	2.51b	2.77b
Acidic		
pH 3.8	5.27a	5.42a
4.8	3.08b	3.59b
5.8	1.43c	1.72c

[a] Within each assembly time and phase, mean values followed by different letters are significantly different ($\alpha = 0.05$, Duncan's Multiple Range Test).

and 10.0 did not differ from each other. At 0 min. in the acidic phase, however, mean tack values at pH 3.8, 4.8, and 5.8 differed significantly from each other. Trends for the 20-min. assemblies were similar to those for 0 min. (Table III). A multiple linear regression model for tack at both assembly times, including variables for alkaline pH, acidic pH, their interaction, surface tension, and viscosity, indicated that acidic and alkaline pH as well as viscosity, indicated that acidic and alkaline pH as well as viscosity significantly affected tack ($r^2 = 0.89$ at 0 min., $r^2 = 0.85$ at 20 min.).

A simple linear regression demonstrated that tack and viscosity, which exhibited similar trends, were closely correlated ($r^2 = 0.83$). Viscosity may indicate degree of polymerization as well as coalescence of polydisperse colloidal particles. We do not know whether increased tack and viscosity were affected by the mechanism of polymerization or coalescence. However, results in Table III indicate that pH level in the acidic phase of synthesis was ultimately most crucial to resin performance.

Part III: Reaction catalyst

Tack at 0-min. assembly time differed significantly ($\alpha = 0.05$, paired t-test) from tack at 20-min. assembly time. Therefore, the tack data were separated by assembly time for all analyses. Regardless of assembly time, tack increased significantly ($\alpha = 0.05$) with time for all cooks tested (Table IV). However, neither the alkaline or acidic phase catalysts nor their interaction significantly affected tack ($\alpha = 0.05$, analysis of variance); therefore, Duncan's Multiple Range Test was not conducted. Reaction times were similar for all treatments, and neither viscosity nor surface tension was affected by catalyst (Table IV). However, a multiple linear regression model including variables for alkaline pH, acidic pH, their interaction, surface tension, and viscosity indicated that surface tension significantly affected tack at 0 and 20 min.

Our results suggest that the catalytic effects of HMTA and TEA may have been suppressed by the NaOH. Therefore, the degree of polymerization may not have approached that possible without the moderating influence of NaOH. This area needs further study.

TABLE IV

Effect of alkaline and acidic phase reaction catalyst on tack of urea-formaldehyde resin and its characteristics at two assembly times, as measured by the push-off method

Reaction catalyst[a]		Resin characteristics			Tack (in.) at two assembly times	
Alkaline phase	Acidic phase	Solids (%)	Viscosity[b] (cps)	Surface tension (dynes/cm)	0 min.	20 min.
NaOH	HCl	34.9	18.2	60.0	5.81	5.99
	CH_3COOH	40.0	13.3	57.7	4.03	4.75
	NH_4Cl	40.2	14.8	63.2	6.06	6.31
	H_3PO_4	39.8	18.6	58.4	5.40	5.66
NaOH/TEA	HCl	40.5	14.0	62.0	4.16	4.72
	CH_3COOH	40.1	15.1	58.2	2.84	2.88
	NH_4Cl	41.5	22.5	58.8	4.47	5.25
	H_3PO_4	40.2	17.7	58.6	6.06	6.70
NaOH/HMTA	HCl	40.2	12.3	59.4	3.69	4.75
	CH_3COOH	41.0	17.8	59.9	6.00	6.51
	NH_4Cl	39.8	17.6	61.2	5.62	6.46
	H_3PO_4	41.0	17.2	54.5	4.49	5.56

[a] TEA = tetraethanolamine; HMTA = hexamethylenetetramine.
[b] Measured by Brookfield viscometer, spindle no. 1, 50 rpm.

CONCLUSIONS

1. Molar F:U ratio and ingredient concentration may be significant variables to tack development and crucial to resin morphology. On the average, tack was increased by reducing the F:U ratio from 2.3:1.0 to 1.9:1.0 and by decreasing ingredient concentration from 50 to 35%.
2. The pH level in the alkaline and acidic phases of resin synthesis significantly affects tack and viscosity. Apparently, tack can be developed in either the alkaline or acidic phase, but tack development does not depend on an interaction of pH levels. Controlling pH is more crucial to tack development in the acidic than in the alkaline phase. However, whether the tack development process is a function of polymerization or coalescence of colloidal particles is not yet known.
3. Reaction catalyst can affect reaction rate but, within the limits tested, did not affect resin characteristics or tack. Further testing is needed in this area.
4. Tack is time dependent, probably because of absorption of the resin carrier and progressive changes in resin morphology. With further study, optimum manufacturing timing could be defined.
5. Multiple linear regression models indicated tack could be influenced by viscosity or surface tension. The correlation between tack and viscosity was especially strong. Understanding these relationships should make tack within the manufacturing system more predictable.

Acknowledgments

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References

1. P. J. C. Counsell, *The Nature of Tack: Aspects of Adhesion*, D. J. Alner and K. W. Allen, Eds (Transcripta Books, London, 1973).
2. W. C. Wake, *Adhesion and the Formulation of Adhesives* (Applied Science Publishers Limited, London, Chap. 7, pp. 138–192.

3. R.R. Meyers and C. J. Knauss, in *Handbook of Adhesives*, 2nd Ed., I. Skeist, Ed. (Van Nostraud Reinhold, New York, 1962), Chap. 4, pp. 72–91.
4. M. Petronio, *ibid.*, Chap. 5, pp. 92–113.
5. T. J. Pratt, W. E. Johns, R. M. Rammon, and W. L. Plagemann, *J. Adhesion* **17**, 275 (1985).
6. J. S. Autenrieth, in *Handbook of Adhesives*, 2nd. ed., I. Skeist, Ed. (Van Nostrand Reinhold, New York, 1962), Chap. 14, pp. 222–232.
7. J. Delmonte, *The Technology of Adhesives* (Reinhold Publishing Corp., New York, 1947), Chap. 14, pp. 341–349.
8. C. Dewalt, *Adhes. Age* **13**, 38 (1970).
9. H. Kambe and K. Managata, *J. Appl. Polym. Sci.* **13**, 493 (1969).
10. O. Udvardy and G. Plester, *Proc. Wash. State Univ. Symp. Particleboard* (Maloney, Pullman, WA, 1972), pp. 13–23.
11. W. C. Wake, "Elastomeric Adhesives" in *Treatise on Adhesion and Adhesives*, Vol. 2, R. L. Patrick Ed. (Marcel Dekker, Inc., New York, 1969).
12. C.Y. Hse, *Mokuzai Gakkaishi* **20**, 483 (1974).
13. C. Y. Hse, *ibid.*, pp. 491.
14. C. Y. Hse, *ibid.*, pp. 538.
15. R. M. Rammon, W.E. Johns, J. Magnuson, and A. K. Dunker, *J. Adhesion* **19**, 115 (1986).
16. A. K. Dunker, *et al.*, *ibid.* **19**, 153 (1986).
17. J. Stuligross and J. A. Koutsky, *ibid.* **18**, 281 (1985).