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Title:	THE INFLUENCE	OF VARIOUS	RESIN	PARAM	ETERS ON	THE	PHYSICAL	
	PROPERTIES OF	OAK FLAKEB	OARD					
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The substantial increase in resin costs due to the recent energy crisis, increased public awareness to the problem of free formaldehyde from boards bonded with urea-formaldehyde resins, and a projected increase in the production of exterior board have emphasized the need for new and substitute adhesive systems. One new development is the use of lignosulfonates in combination with conventional phenolformaldehyde resins for the production of particleboard.

An experiment was carried out to test the applicability of such a substituted adhesive system. A commercial phenol-formaldehyde resin and six ammoniated-lignosulfonate-phenol-formaldehyde resins of varying viscosity were used in the fabrication of randomly oriented oak flake-boards. Resin application rates of 3, 4 1/2, and 6 percent were employed in combination with nominal board densities of 40 and 45 lbs/cu.ft.

Test results indicated similar properties (internal bond, modulus of rupture, modulus of elasticity, linear expansion, thickness swell, and weight gain) between the commercial phenolic resin and the substituted resins. Internal bond strength increased with increasing

resin viscosity in the range studied, while modulus of rupture, modulus of elasticity, thickness swell, and weight gain properties generally decreased. Increasing resin application rate and board density resulted in an increase in board strength properties.

In a second study, resin wetting and resin molecular weight properties were related to internal bond and modulus of rupture strength values. Contact angle measurements were used as indicators of resin wetting properties. Measurements were taken at 5, 30, 60, 300, and 600 second intervals. Resin molecular weight distributions were broken into low, medium, and high fractional molecular weight areas.

No significant relationships were found between wetting properties and bond strength (IB, MOR). Wetting alone was not sufficient for the formation of a strong adhesive bond. Significant relationships were found, however, between fractional areas under the molecular weight distribution curve and internal bond strength. IB values were negatively related to low and medium molecular weight resin molecules, while high molecular weight resin molecules were positively related to IB values. Medium molecular weight area exhibited the highest degree of correlation. No correlation was found between resin molecular weight distribution and modulus of rupture values. In addition, no significant relation was found between IB and MOR properties and resin pH.

The Influence of Various Resin Parameters on the Physical Properties of Oak Flakeboard

by

Gregory Lynn Jay

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submitted to

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TABLE OF CONTENTS

STUDY ONE:	The Effect of Resin Type, Resin Viscosity, and Resin Application Rate on the Physical Properties of Oak Flakeboard
I	Introduction1 Statement of Objective2
II	Review of Literature
III	Experimental Design
IV	Results and Discussion
V	Summary and Conclusions
STUDY TWO:	The Effect of Resin Wettability and Resin Molecular Weight Distribution on Bonding in Oak Flakeboard64
I	Introduction
II	Review of Literature
III	Experimental Design
IV	Results and Discussion

V	Summary and Conclusions88
	Bibliography90
Appendix	A91
Appendix	B94
Appendix	C100

LIST OF FIGURES

Figure		Page
1	Cutting pattern for test specimens.	19
2	Internal bond values as affected by resin viscosity, resin rate, and board density.	24
3	Effect of resin rate and board density on internal bond properties.	26
4	Effect of resin viscosity on internal bond properties.	27
- ′5	Modulus of rupture values as affected by resin viscosity, resin rate, and board density.	33.
6	Effect of resin viscosity on modulus of rupture properties.	35
7	Modulus of elasticity values as affected by resin viscosity, resin rate, and board density.	39
8	Effect of resin viscosity on modulus of elasticity properties.	41
9	Effect of resin viscosity on linear expansion properties.	45
10	Effect of resin rate on linear expansion properties.	46
11	Thickness swell values as affected by resin viscosity and resin rate; 40 lbs/cu.ft. nominal board density.	49
12	Thickness swell values as affected by resin viscosity and resin rate; 45 lbs/cu.ft. nominal board density.	50
13	Effect of resin viscosity on thickness swell properties.	51
14	Weight gain values as affected by resin viscosity and resin rate; 40 lbs/cu.ft. nominal board density.	56
15	Weight gain values as affected by resin viscosity and resin rate; 45 lbs/cu.ft. nominal board density.	57
16	Effect of resin viscosity on weight gain properties.	58
17	Measurement of contact angle.	67
18	Molecular weight distribution break points.	80
19	Internal bond strength versus LOW/TOTAL molecular weight	83

20	Internal bond strength versus	MEDIUM/TOTAL molecular		
	weight area.		.	34
21	Internal bond strength versus	HIGH/TOTAL mole	ecular weight	
,	area.		{	85

LIST OF TABLES

Table		Page
1	Summary of experimental design.	13
2	Resin properties.	15
3	Particleboard process conditions.	17
4	Internal bond values for the viscosity × rate × density interaction.	23
5	Effect of resin type on IB properties; 39.24 lbs/cu.ft. adjusted board density.	29
6	Effect of resin type on IB properties; 45.17 lbs/cu.ft. adjusted board density.	30
7	Modulus of rupture values for the viscosity \times rate \times density interaction.	32
8	Effect of resin type on MOR properties; 40.39 lbs/cu.ft. adjusted board density.	36
9	Effect of resin type on MOR properties; 45.41 lbs/cu.ft. adjusted board density.	37
10	Modulus of elasticity values for the viscosity \times rate \times density interaction.	38
11	Effect of resin type on MOE properties; 40.39 lbs/cu.ft. adjusted board density.	42
12	Effect of resin type on MOE properties; 45.41 lbs/cu.ft. adjusted board density.	43
13	Thickness swell values for the viscosity \times rate \times density interaction	48
14	Effect of resin type on TS properties; 40 lbs/cu.ft. nominal board density.	52
1:5	Effect of resin type on TS properties; 45 lbs/cu.ft. nominal board density.	53
16	Weight gain values for the viscosity \times rate \times density interaction	55
17	Effect of resin type on WG properties; 40 lbs/cu.ft.	59

18	Effect of resin type on WG properties; 45 lbs/cu.ft. nominal board density.	60
19	Relationship between molecular weight of polyethylene glycol and the nonsolvent-water content of green wood.	72
20	Bulking of cross sections of Englemann spruce at 25°C.	74
21	Bulking of cross sections of Englemann spruce at 90°C.	7.5
22	Regression equations relating IB properties (psi) to fractional molecular weight areas under the distribution curve.	82

The Influence of Various Resin Parameters on the Physical Properties of Oak Flakeboard

STUDY ONE: The Effect of Resin Type, Resin Viscosity, and Resin Application Rate on the Properties of Oak Flakeboard

I. INTRODUCTION

Particleboard is made by combining wood flakes and/or particles with resin binders and consolidating them into panels of various sizes under heat and pressure.

The raw materials required in the production of particleboard are wood furnish, adhesive, and wax. Wood furnish consists mainly of planer shavings, sawdust, and plywood trim. The adhesive for the manufacture of particleboard consists of two main types: ureaformaldehyde, used extensively for particleboard intended for interior applications, and phenol-formaldehyde, used when the particleboard will be exposed to moist conditions. Urea- and phenol-formaldehyde adhesives are derivatives of the natural gas and petroleum industries respectively, and as a result of the recent energy crisis their costs have substantially increased.

At present, the major portion of particleboards produced are for interior applications and use urea-formaldehyde resins. However, production of exterior board is projected to increase, emphasizing the need for a means of reducing phenolic resin costs. Also, due to increasing public awareness over free formaldehyde, which is more prevalent in boards containing urea-formaldehyde than those containing phenol-formaldehyde, there has been interest in developing new and substitute adhesives. One of the developments reported is the use of

spent sulfite liquor, both alone and in combination with commercial phenolic resins, for exterior particleboard. A substitution of lignosulfonate (a waste liquor from sulfite pulping processes) might be made in place of the more expensive phenol to aid in the reduction of resin costs. This study deals with the use of such substituted phenolformaldehyde resins.

Statement of Objective

The objective of this study was to determine the effect of resin type, resin viscosity, and resin application rate on the physical properties of a randomly oriented oak flakeboard to be used as a corestock in veneer-particleboard composite panels. The physical properties examined were internal bond (IB), modulus of rupture (MOR), modulus of elasticity (MOE), linear expansion (LE), thickness swell (TS), and weight gain (WG).

II. REVIEW OF LITERATURE

The Use of Spent Sulfite Liquor as a Binder for Particleboard

Lignin, a natural bonding agent in wood, is a polymeric substance located between the cells and in the cell walls of wood. Since lignin is the natural bonding agent in wood, it would seem logical to utilize it as a binder for reconstituted wood products. Spent liquors from both sulfite and sulfate pulping processes could be used as raw materials in the preparation of adhesives. Spent sulfite liquor is an abundant and low cost waste product, thus aiding in the reduction of ever-increasing resin costs (Forss and Fuhrmann, 1976).

Several studies concerning the use of spent sulfite liquor as binder, both alone and in combination with phenol-formaldehyde resins, have been carried out. At the present there are a number of patents describing methods of preparing a resin binder from spent sulfite liquor for composite wood products, however, none has been put to commercial use thus far (Shen, 1974).

In the mid-1960s, Pedersen and Jul-Rasmussen at the Dansk Spaanplade Kompagni patented a process for the production of particle-board using lignosulfonates. As described by Johansen (1971), this process involved a two step procedure: first pressing the board, and second treating it in an autoclave to increase the board's water resistance properties. Wood chips of oak, beech, birch, and other hardwoods were blended with a spent sulfite liquor having a pH of about 3.0, and pressed for 20 minutes at 350°F and 300 psi. The pressed boards were then placed in an autoclave in a vertical position at a

temperature of 400°F for 90 minutes to obtain a waterproof bond. The economic advantage of using the inexpensive spent sulfite liquor was largely offset by the cost of the long press time and autoclaving.

Shen (1974) found that spent sulfite liquor, when modified with a strong mineral acid, could be used as a binder in exterior particle-board with a curing rate comparable to that of a conventional phenolic resin. Poplar wafers were sprayed with a 15 to 20 percent solution of sulfuric acid and then mixed with a powdered calcium lignosulfonate. Shen found that the physical properties of boards produced using a resin of this type generally exceeded the requirements of the Canadian Standards Association standards for exterior waferboard [CSA 0188 (68)].

In a second report, Shen (1974) described a refined process using an acid-catalyzed spent sulfite liquor powder. This refined process was found to be superior to the original in that only one blending operation was required using the acid-catalyzed-spray-dried powder; binder and acid content in the finished board were lower; and direct spraying of dilute acid on the flakes was avoided.

Calculated amounts of poplar wafers and spent sulfite liquor powder were loaded into a rotary drum blender (a blending operation similar to that used in commercial production of waferboard in which powdered phenolic resin is used) and blended. Board physical properties were improved over previous work using this simplified blending operation, and acid requirements in the finished board were reduced.

Shen found that the spent sulfite liquor adhesive could replace phenolic- and urea-based resins, and that it was suitable for the manufacture of boards for exterior use, at a resin cost of about one

third that of phenolic resin.

Although the use of acidified spent sulfite liquor as a binder for particleboard shows promise and offers economic advantages, there are certain disadvantages associated with its use. The high acid content associated with the resin can reduce wood strength as a result of wood hydrolysis. The hydrolysis of wood in the presence of a strong acid, particularly under hot-pressing at high temperature and pressure, will proceed at a rapid rate. In addition, the high board acid content may have a corrosive effect on nails and other hardware used to fabricate articles from the panel. Equipment which comes into contact with the binder, binder handling and spraying equipment, the blender, the forming equipment, and the caul plates would require a coating with acid resistant materials. These disadvantages have kept this type of binder from commercial usage at the present time.

At the German Wilhelm-Klauditz-Institut, Roffael and Rauch (1971) studied the use of spent sulfite liquor in the manufacture of particle-board. They found that the addition of an acid-tolerant phenol-formaldehyde resin increased the binding properties of sulfite lignin and led to a practically water-insoluble thermosetting adhesive without using a post thermal aging step. Particleboards produced through the application of this method had higher mechanical properties than boards produced using the Pedersen process discussed earlier. The board properties were equal to or better than those in which spent sulfite liquor was used alone as a binding agent. Strength and swelling properties were found to be affected by the amount of phenol-formaldehyde added, the pH value of the sulfite black liquor, and the

pressing temperature. A pH range between 4 and 5 was found to be optimum.

In a further study Roffael and Rauch (1973) found that ammoniumand sodium-lignin sulfonates could be used as a partial substitute or
extender for conventional alkaline phenolic resins used in the production of particleboards. In the case of one layer boards 0.750-inch
thick, they found that about 25 percent of the resin used could be
replaced by sulfite liquor. Board physical properties were found to be
dependent to a great extent on the pH value of the sulfite liquor and
the alkali content of the phenolic resin used. The pH value of sulfite
liquor seemed to have an optimum value in the range of 7 to 10. Results
of three layer boards (also 0.750-inch thick) indicated that about 35
percent of the phenolic resin used in upper layers could be replaced by
sodium-lignin sulfonates without any significant decrease in the
mechanical properties of the boards. Additional results showed that
the phenolic resin could be substituted by up to 50 percent sulfite
liquor and still have properties above German standards.

Holderby, Olson and Wegener (1967) at the Pulp Manufacturers

Research League in Appleton, Wisconsin, developed an adhesive containing

60 percent lignosulfonates and 40 percent acid tolerant phenol
formaldehyde resin. The adhesive was used in the production of 3-ply

birch/fir/birch and Douglas-fir interior plywood. When tested, this

adhesive gave a glue bond meeting the requirements of the accepted

commercial standards for interior grade plywood.

Research by Forss and Fuhrmann (1976) at the Finish Pulp and Paper Research Institute has resulted in a type of lignin based adhesive intended for use by the plywood, particleboard and fibreboard industry. This adhesive is known as KARATEX. KARATEX is prepared by the mixture of phenol-formaldehyde resin with lignosulfonates. The adhesive was tested in both laboratory and mill scale production conditions using Douglas-fir veneer. Pressing conditions, i.e. press temperature, pressure, and time, were similar to those used with conventional phenolic resins.

Spent sulfite liquor appears to show promise as a commercially acceptable new binder for wood particleboard.

Effect of Resin Viscosity on Board Properties

Sullivan and Harrison (1965) state that the viscosity of any resin system has important implications when the movement of an adhesive is considered. An adhesive must (1) flow laterally to form a continuous film; (2) transfer from the wood surface on which it is spread to the opposite unspread surface; (3) penetrate into both wood surfaces; (4) wet the wood substance; and (5) solidify into a strong mass. The stages of flow and transfer occur at a rate determined in part by the fluid nature of the adhesive. Surface texture and cleanliness, pressure, and temperature must also be considered. Penetration is controlled by the molecular size and surface tension of a resin, of which viscosity is an index. Viscosity may also directly influence the geometry of the glueline, particularly the thickness. For adhesives with identical solids content, the lower viscosity material will lose solids due to squeezeout and excessive penetration. Providing no changes are made in pressure and temperature, as viscosity increases, penetration and

squeeze-out decrease, resulting in a higher solids content on the glueline and a thicker glueline.

Rice (1965) in some work performed on Douglas-fir plywood, found good correlation between resin viscosity of urea-formaldehyde resins and bond durability, with higher viscosity resins having greater durability. Rice studied viscosities over the range of 314 cps to 41,600 cps (as measured at 25°C with a Brookfield LVF viscometer). He attributed the increased bond durability with increased resin viscosity to reduced flow characteristics into the wood cavities and subsequently the retention of adequate quantities of glue between wood interfaces. Thicker gluelines exhibited more durable bonds.

Snyder, Rice and Hart (1967) looked at the effect of varying resin viscosity on the physical properties of internal bond, modulus of rupture, modulus of elasticity, and dimensional stability of a yellow-poplar flakeboard. Resins used were (1) a 150 centipoise viscosity (55 percent solids) urea-formaldehyde commercial resin, (2) resin 1, to which a thickener was added in the amount of 0.25 percent to produce a viscosity of 1400 centipoise at 25°C, and (3) resin 1 aged at 50°C to a viscosity of 1100 centipoise at 25°C. The authors found that resin viscosity had a significant effect on internal bond properties, but not upon static bending or dimensional stability.

Bryant and Garcia (1967) found in studies with Philippine luan plywood, that migration of phenolic resin into the wood before hot pressing was influenced by the viscosity of the resin, which was a function of the resin's average molecular weight. Low molecular weight resins are expected to move further away from the glueline during

assembly times than a more advanced resin through capillary movement and cell wall diffusion, decreasing the volume of resin present at the glueline for bonding.

Effect of Resin Application Rate on Board Properties

There have been many investigations concerning the effect of resin application rate on board physical properties. All investigators are unanimous in their findings that as the adhesive content increases strength properties of the resultant particleboard increase (Kelly, 1977).

Lehmann (1970) looked at the effect varying resin content had on the physical properties of boards made from a uniform mixture of Douglas-fir flakes and particles. Three levels of resin content were studied: 2, 4, and 8 percent based on ovendry weight of wood. He found that values for modulus of rupture and modulus of elasticity increased rapidly with an increase in resin content from 2 percent to 4 percent and reached optimum levels between 4 and 8 percent resin content. For internal bond, the same effects were evident as resin content increased, except that the leveling-off tendency was less apparent. Internal bond strengths continued to increase with increasing resin content in the range studied. A leveling-off in thickness swelling as resin content increased was especially evident. Lehmann found that at low resin contents low density boards were the most stable, but at high resin contents the reverse was true, though differences were not large. He concluded that static bending and thickness swelling approached optimum levels below 8 percent resin content, but internal bond strength

continued to increase with increasing resin content.

Post (1958, 1961) investigated the effect of resin content on the bending strength properties of oak flakeboard. He found that increases in percent resin content can be expected to improve bending strength only to a moderate degree. He also found that stiffness was not materially influenced by resin content. Flake size was the controlling factor.

Talbott and Maloney (1957) investigated the effect of resin content on the strength of particleboards made from two screen fractions of green Douglas-fir planer shavings. A powdered phenolic resin was applied at rates of 2, 4, and 8 percent. They found an increase in modulus of rupture values with increasing resin content. This increase, however, was negligable in the range from 4 to 8 percent. In the case of internal bond, an increase in resin content resulted in a nearly proportional gain in internal bond strength, a sharp contrast to the modulus of rupture results.

Jorgensen and Odell (1961) studied the effect resin spread had on the dimensional stability of oak flakeboard. They found that the magnitude of dimensional changes in a board was altered by the quantity of non-hygroscopic resin included for bonding. Higher resin spreads produced the most stable product.

Larmore (1959) found that the amount of resin used in fabricating boards of aspen and yellow birch flakes influenced the percent change in thickness. As resin rate increased, thickness swelling decreased. He found that 8 percent resin solids resulted in reduced thickness change over a 4 percent resin level.

In general, improved board properties can be found with increasing resin application rates.

III. EXPERIMENTAL DESIGN

A study was designed to compare the effect of resin type, resin viscosity, and resin application rate on the physical properties (internal bond, modulus of rupture, modulus of elasticity, linear expansion, thickness swell, and weight gain) of a randomly oriented oak flakeboard. A commercial phenol-formaldehyde resin (PF) and six ammoniated-lignosulfonate-phenol-formaldehyde resins (ALS-PF) of varying viscosity and pH were applied at rates of 3, 4 1/2, and 6 percent, ovendry wood weight basis. Nominal board densities were 40 and 45 lbs/cu.ft. (based on ovendry board weight).

A completely randomized 2- by 3- by 7-factorial experiment was run. Order of board manufacture was determined using a random numbers table to minimize the effects of external conditions during board fabrication. Three replications (boards) were made for each combination of independent variables. A total of 126 boards were made $(2\times3\times7\times3)$. Table 1 contains a summary of the experimental design.

Preparation of Boards

The approximately 500 pounds of oak flakes used in this study were secured through the Weyerhaeuser Company. The flakes were obtained from a clear cutting of mixed hardwoods in the Oklahoma-Arkansas region. The logs were then commercially flaked using a drum flaker and dried to approximately 4 percent moisture content. The flakes were then shipped to the Forest Research Laboratory, Oregon State University. The furnish consisted of 72 percent oak (Quercus spp.), 17 percent gum (Liquidambar styracifula L.), and 11 percent ash (Fraxinus spp.), as determined by a

Table 1. Summary of Experimental Design.

Factor	Number of	Levels	Levels Studied
Resin viscosity	7		320 cps ¹ (PF) 84 cps (ALS-PF) 100 cps (ALS-PF) 270 cps (ALS-PF) 300 cps (ALS-PF) 850 cps (ALS-PF) 540 cps (ALS-PF)
Resin rate	3		3.0 percent 4.5 percent 6.0 percent
Board density	2		40 lbs/cu.ft. 45 lbs/cu.ft.
Replications/ Combination	.		3 replications/combination

 $^{^{1}\}mathrm{Brookfield}$ viscometer LVT at 25°C.

random sample. The flakes were screened using a #6 mesh screen to remove the fines and dust particles generated during the flaking operations, after which they were placed in plastic bags and sealed in fiber drums to maintain the moisture content until use in board fabrication.

The resins used in this study were a commercial phenol-formaldehyde resin manufactured by Reichhold Chemical, Inc., and six substituted ammoniated-lignosulfonate-phenol-formaldehyde resins supplied by the Weyerhaeuser Company. Resin properties are summarized in Table 2. The wax used in this study was Borden Chemical's EW-403-H.

Wax and resin were applied to the flakes using a laboratory type rotary drum blender. The flakes were brought up to an in-press moisture content of 10 percent (based on ovendry wood weight) through the spraying of water before spraying the wax and the resin. Wax was applied at a rate of 0.5 percent based on ovendry weight of the furnish. Resin application rates were 3, 4 1/2, and 6 percent respectively, also based on ovendry wood weight. The phenol-formaldehyde resin was applied at 47 percent solids content and the ammoniated-lignosulfonates were applied at 41 percent solids content. Wax solids content was 47 percent. The amount of furnish required to fabricate the three board replications was placed in the blender for each resin application rate. After blending, all furnish was removed from the blender and sealed in a plastic bag until use in board fabrication.

The flakeboard mats were hand formed onto an 18- by 18-inch aluminum caul plate. Flake orientation was random. The amount of blended flakes used per board was determined according to the ovendry

Table 2. Resin Properties.

			Viscosity at 25°C Brookfield (LVT)				
Sample Number	Type ^l	Spindle Number	RPM	mPa's (poise)	% Solids	рН	
A	PF	2	60	320	47.0	10.4	
В	ALS-PF	1	60	84	41.0	12.1	
С	ALS-PF	1	60	100	40.8	10.6	
D	ALS-PF	- 2 · 2	60	270	41.3	12.3	
E	ALS-PF	2	60	300	41.0	10.6	
F	ALS-PF	4	60	850	40.9	12.2	
G	ALS-PF	4	60	540	41.1	10.6	

 $^{^{1}\}mathrm{PF}=\mathrm{phenol-formaldehyde}$ resin; ALS-PF = ammoniated-lignosulfonate-phenol-formaldehyde resin.

board density desired (40 and 45 lbs/cu.ft.). Only the amount of flakes necessary for the fabrication of a single board were removed from the plastic bag at one time.

The felted mat was placed in a 24- by 24-inch high pressure steam heated hot press and pressed to stops. Stop thickness was 0.290 inch. Press closing time was 30 ± 5.0 seconds. An initial closing pressure of 720 psi was held for 30 seconds. The pressure was then released over the next 30 seconds to 240 psi. This pressure was held for a period of four minutes after which time the pressure was reduced to zero and the mat was removed from the press. Total press time was six minutes. Press platen temperature was $400^{\circ}F$.

After pressing all boards were hot stacked in a rigid foam insulated plywood box for 24 hours, after which time they were stickered and stacked in a conditioning room operating at 70°F and 65 percent relative humidity. The boards were allowed to reach equilibrium conditions for a period of two weeks before testing. Table 3 contains a summary of the process conditions used in this study.

Testing of Boards

The pressed boards were conditioned at 70°F and 65 percent relative humidity (RH) for a period of two weeks, and sanded on both sides to a nominal 0.250 inch thickness prior to testing. All boards were sanded using a 24-inch pressure fed belt sander at the particleboard laboratory of Weyerhaeuser Company. Four passes of 0.01 inch each were made on alternate faces of the boards to remove the material equally from each face.

Table 3. Particleboard Process Conditions.

Factor	Condition
Board density	40 lbs/cu.ft. 45 lbs/cu.ft.
Board thickness	0.290 inch; sanded to 0.250 inch
Wood furnish	72 percent oak 17 percent gum 11 percent ash
Resin solids	47.0 percent; PF-320 cps-10.4 41.0 percent; ALS-PF-84 cps-12.1 40.8 percent; ALS-PF-100 cps-10.6
	41.3 percent; ALS-PF-270 cps-12.3
	41.0 percent; ALS-PF-300 cps-10.6 40.9 percent; ALS-PF-850 cps-12.2 41.1 percent; ALS-PF-540 cps-10.6
Resin content in board	3 percent 4 1/2 percent 6 percent
Wax solids	47.0 percent
Wax content in board	0.5 percent
Press closing time	30.0 ± 5.0 seconds
Press cycle	6 minutes
Press temperature	400°F
Mat M.C. into press	10.0 ± 0.5 percent

After conditioning and sanding the boards were cut into test specimens according to the cutting diagram in Figure 1. Each internal bond (IB), modulus of rupture (MOR), modulus of elasticity (MOE), and linear expansion (LE) specimen was measured in length and thickness to the nearest 0.001 inch. Specimen dimensions were 2 inches square for IB, 3 inches by 8 inches for MOR and MOE, and 3 inches by 12 inches for LE in accordance with ASTM D 1037-72a. The weight of each test specimen was measured to the nearest 0.1 gram. From the measurements the density (lbs/cu.ft.) of each test specimen was calculated.

Test procedures outlined in ASTM D 1037-72a were followed for the testing of IB, MOR, and MOE test specimens, however, the test used for LE and TS was more severe than that outlined. ASTM D 1037-72a calls for test conditions of 68°F, 50 percent RH to 68°F, 90°percent RH. The test conditions used in this study were 90°F, 30 percent RH to 90°F, 90 percent RH.

IB specimens were tested using a Tinius-Olsen testing machine with a cross-head speed of 0.02 inch per minute. MOR and MOE specimens were tested using an Instron testing machine with a cross-head speed of 0.12 inch per minute. Chart speed was 1.2 inch per minute. LE test specimens were placed in conditions of 90°F, 30 percent RH for a period of two weeks, after which time their length, width, thickness, and weight were measured. The specimens were then placed in conditions of 90°F, 90 percent RH for a two week period, at the end of which time their length, thickness, and weight were again measured. The specimens were

¹ASTM D 1037-72a. American Society for Testing and Materials, Wood-Base Fiber and Particle Panel Materials. Revised 1972.

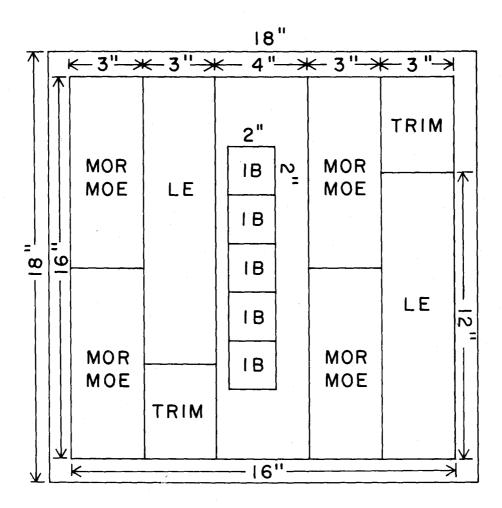


Figure 1. Cutting pattern for test specimens.

placed back in the 90°F, 90 percent RH conditions for an additional two week period to assure that the specimens had reached an equilibrium moisture content. Results indicated, however, that equilibrium conditions were reached in the initial two week conditioning period.

LE and TS were calculated as percent of initial conditioned dimensions (90°F, 30 percent RH). WG was calculated as percent of initial conditioned weight.

Fifteen IB specimens, twelve MOR and MOE specimens, and six ${\tt LE}$ specimens were tested for each experimental combination.

IV. RESULTS AND DISCUSSION

Strength Properties

The strength properties for each resin viscosity, resin application rate, and board density were compared statistically with an analysis of variance. Board density was found to exhibit rather large variations both between and within replications, so all strength property values were first adjusted to a common density. This was possible because the slopes of the regression lines fit to the data for a given resin viscosity were not significantly different between resin application rates. IB, MOR, and MOE properties increased in a linear fashion with increasing board density. The regression equations used in adjusting the data for IB, MOR, and MOE are contained in Appendix A. LE, TS, and WG values were not adjusted because these board properties were not found to exhibit any relationship with board density. Also, density variations between LE specimens were found to be small. The results of the IB, MOR, MOE, LE, TS, and WG tests from each board were averaged to obtain one value per board, and these average values were used in the analysis of variance. Thus the test result for IB was the average of five values per replication, MOR and MOE the average of four values, and LE, TS, and WG the average of two values per replication. The values for each replication were kept separate, as averaging of these values would cause elimination of the error term in the analysis. The data was analyzed as a 2- by 3- by 7- by 3-factorial experiment.

The analysis was broken into the main effects of resin viscosity, resin rate, and density, and also into the interactions of

viscosity × rate, viscosity × density, rate × density, and viscosity × rate × density. The analysis of variance tables can be found in Appendix B. When interaction terms are found to be significant, they are of primary importance over the main effects when extracting information from factorial analyses. If no significant interaction terms were found, the main sources (resin viscosity, resin rate, and density) showing significant differences were investigated. Significant differences were determined using Tukey's method of multiple comparisons. This method has the property that if some or all of the differences between pairs of means are tested, the probability that no erroneous claim of significance will be made is ≥ 0.95.

Internal Bond

Investigation of the analysis of variance table for IB (Appendix Table B-1) reveals that the main effects as well as the interaction terms exhibit significant differences at the .95 and .99 levels. However, since the three way interaction of viscosity × rate × density shows significance, this term is the most meaningful and will contain all the information available. The significance of this interaction indicates that IB values obtained will depend upon not only the resin viscosity being observed, but also upon the resin application rate and board density employed. Table 4 contains the adjusted IB values for the viscosity × rate × density interaction. Figure 2 is a graphical representation of the data contained in Table 4, helping to illustrate the effect of resin viscosity and resin rate on IB values for the two board densities.

Table 4. Internal Bond Values for the Viscosity \times Rate \times Density Interaction.

	Adamatad		IB (psi)		
	Adjusted Board Density	Resin Application Rate			
Resin Type	(lbs/cu.ft.)	3.0%	4.5%	6.0%	
PF-320 cps-10.4 ¹	39.24	55.0	70.0	90.4	
	45.17	92.2	124.1	148.9	
ALS-PF-84 cps-12.1	39.24	61.2	73.1	75.7	
	45.17	112.2	122.1	151.3	
ALS-PF-100 cps-10.6	39.24	43.4	55.5	54.3	
	45.17	69.4	91.5	115.1	
ALS-PF-270 cps-12.3	39.24	58.4	74.4	88.8	
	45.17	112.9	125.6	126.7	
ALS-PF-300 cps-10.6	39.24	44.2	53.6	65.4	
•	45.17	78.2	90.0	116.1	
ALS-PF-850 cps-12.2	39.24	64.6	74.2	88.4	
	45.17	109.9	136.6	140.0	
ALS-PF-540 cps-10.6	39.24	48.6	56.8	64.6	
	45.17	84.0	110.5	105.6	

 $^{^{1}\}mathrm{Resin}$ type, resin viscosity, resin pH.

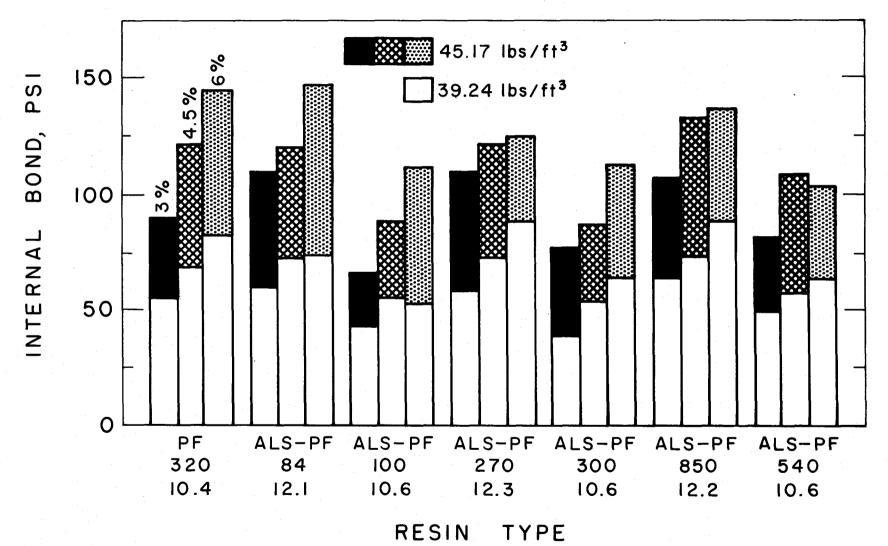


Figure 2. Internal bond values as affected by resin viscosity, resin rate, and board density.

Observation of Table 4 and Figure 2 shows that as resin rate increases over the range of 3 to 6 percent, IB values also increase. This trend can also be seen in Figure 3 illustrating the effect resin rate and board density exhibit on IB values. However, it should be pointed out that the effect of resin viscosity has not been considered here.

As percent resin content increases, resin droplet distribution and resin coverage increase, the result being an increase in board strength properties.

The effect board density exhibits on IB values can also be observed in Figure 3. An increase in nominal board density from 40 to 45 lbs/cu.ft. resulted in a substantial increase in IB values. Board density can be increased by increasing the volume of raw material used in forming a particleboard mat of a given thickness, or by pressing a given volume of raw material to a smaller thickness. In either case the wood particles are brought into closer proximity to one another, increasing the chances of particle to resin droplet contact. A higher degree of particle to particle bonding results in a board of greater strength. Resin content is also increased as board density increases. Application of resin solids is based on board weight, therefore higher density boards contain a greater quantity of resin solids.

Unfortunately the three way interaction of viscosity × rate × density makes observation of the effect resin viscosity had on IB values difficult. Figure 4, relating IB values to resin viscosity alone, indicates an increase in IB values with increasing resin viscosity. It must be pointed out here, however, that this illustration does not take

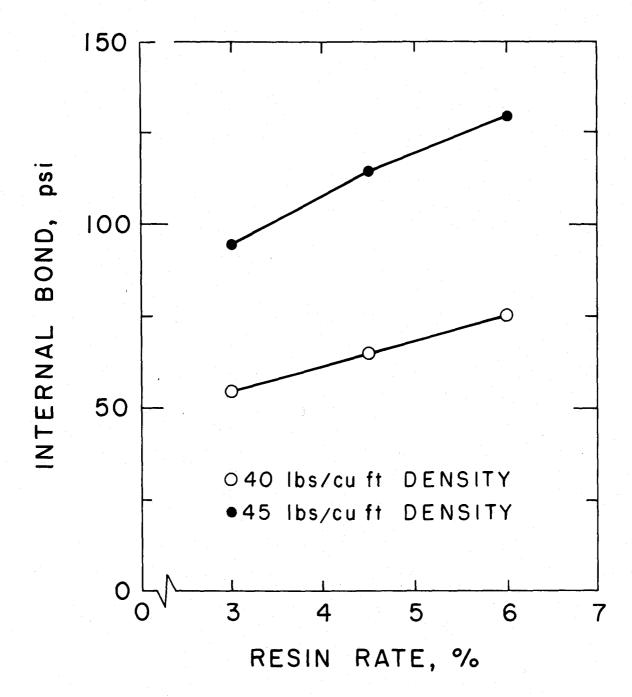


Figure 3. Effect of resin rate and board density on internal bond properties (points represent average values for the resin viscosities studied).

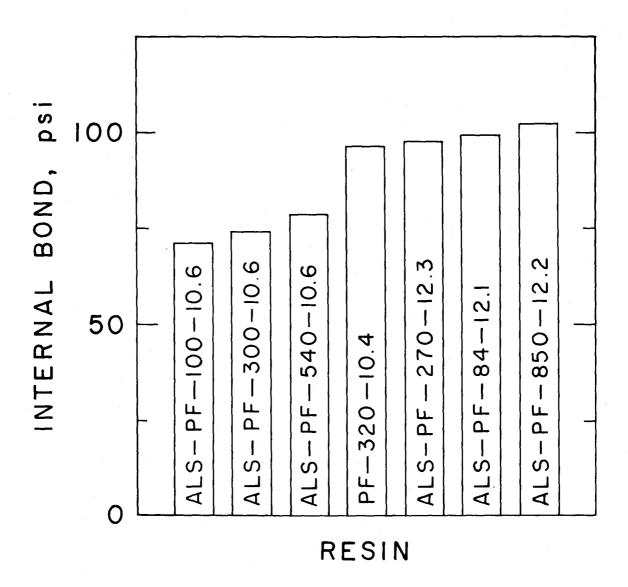


Figure 4. Effect of resin viscosity on internal bond properties. $^{1}\text{Resin type, resin viscosity, resin pH}$

into account the effect of resin rate or board density. It would also appear from Figure 4 that the lower pH ALS-PF resins (refer to Table 2) gave IB values inferior to those obtained using the ALS-PF resins of higher pH. It was hypothesized that a curing problem might have been involved with the set of lower pH ALS-PF resins. A brief study was run to test this hypothesis in which press time was increased from four to ten minutes. IB test results indicated no significant increase in board values, leading to the conclusion that a curing problem was not involved. Rather, higher pH resins improve bonding for oak in some undefined manner.

A change in resin viscosity within a given set of ALS-PF resins did not have a significant effect on IB values. However, in both sets of resins increasing resin viscosities resulted in increasing IB strengths.

Tables 5 and 6 indicate the effect resin type (PF vs. ALS-PF) had on IB values. Tukey's cutoff value for significant differences was 18.95 psi. The results indicate a close agreement between IB values obtained using the commercial phenolic resin and those obtained using the higher pH ALS-PF resins. This would suggest that this type of substituted resin could be successfully utilized in board manufacture. The lower pH ALS-PF resins, however, generally exhibited inferior values as compared to the phenolic resin.

Modulus of Rupture

Observation of the analysis of variance table for MOR (Appendix Table B-2) indicates that the main effects of resin viscosity, resin

Table 5. Effect of Resin Type on IB Values; 39.24 lbs/cu.ft. Adjusted Board Density.

Resin Type	Resin Rate (%)	IB (psi)	Change from Control (psi)
PF-320 cps-10.4	3.0 4.5	55.0 70.0	
	6.0	90.4	*
ALS-PF-84 cps-12.1	3.0	61.2	+6.2
	4.5	73.1	+3.1
	6.0	75.7	-14.7
ALS-PF-270 cps-12.3	3.0	58.4	+3.4
	4.5	74.4	+4.4
	6.0	88.8	-1.6
ALS-PF-850 cps-12.2	3.0	64.6	+9.6
	4.5	74.2	+4.2
	6.0	88.4	-2.0
ALS-PF-100 cps-10.6	3.0	43.4	-11.6
	4.5	55.5	-14.5
	6.0	54.3	-36.1
ALS-PF-300 cps-10.6	3.0	44.2	-10.8
	4.5	53.6	-16.4
	6.0	65.4	-25.0
ALS-PF-540 cps-10.6	3.0	48.6	-6.4
	4.5	56.8	-13.2
	6.0	64.6	-25.8

Cutoff value for significant differences = 18.95 psi.

Table 6. Effect of Resin Type on IB Values; 45.17 lbs/cu.ft. Adjusted Board Density.

Resin Type	Resin Rate (%)	IB (psi)	Change from Control (psi)
PF-320 cps-10.4	3.0	92.2	
	4.5 6.0	124.1 148.9	- -
ALS-PF-84 cps-12.1	3.0	112.2 122.1	+20.0 -2.0
	4.5 6.0	151.3	+2.4
ALS-PF-270 cps-12.3	3.0	112.9	+20.7
	4.5 6.0	125.6 126.7	+1.5 -22.2
ALS-PF-850 cps-12.2	3.0	109.9	+17.7
	4.5 6.0	136.6 140.0	+12.5 -8.9
ALS-PF-100 cps-10.6	3.0	69.4	-22.8
	4.5 6.0	91.5 115.1	-32.6 -33.8
ALS-PF-300 cps-10.6	3.0	78.2	-14.0
	4.5 6.0	90.0 116.1	-34.1 -32.8
ALS-PF-540 cps-10.6	3.0	84.0	-8.2
	4.5 6.0	110.5 105.6	-13.6 -43.3

Cutoff value for significant differences = 18.95 psi.

rate, and density show significant differences (.95 and .99 levels) and that the interaction terms of viscosity × rate and viscosity × rate × density are significant at the .95 and .99 levels respectively. As previously stated the three way interaction is the most meaningful and will contain all the information available. The significance of this interaction indicates that MOR values obtained using a given resin viscosity will depend not only upon the resin application rate employed, but upon board density as well. Table 7 contains the adjusted MOR values for the viscosity × rate × density interaction. Figure 5 is a graphical representation of the test results for MOR, and illustrates the effect of resin viscosity and resin application rate at a given board density.

Investigation of Table 7 and Figure 5 indicates a general increase in MOR values with an increase in percent resin content, however, these increases are only significant in the case of the 100 cps and 300 cps ALS-PF resins in the high density boards. The cutoff value for significant differences was 688 psi, as determined using Tukey's method of multiple comparisons. These results follow with those reported by Post (1958, 1961), Lehmann (1970), and Talbott and Maloney (1957), as discussed earlier. The authors found that increasing percent resin content had little effect on bending strength properties, particularly in the range of 4 to 8 percent.

Increasing nominal board density from 40 to 45 lbs/cu.ft. resulted in a significant increase in bending strength values as can be seen in Figure 5. As was mentioned in the IB discussion, higher board densities result in better particle to particle contacts, and higher quantities of

Table 7. Modulus of Rupture Values for the Viscosity \times Rate \times Density Interaction.

	Adjusted	1000	MOR (psi)	
	Board Density	Resin Application Rate		
Resin Type	(lbs/cu.ft.)	3.0%	4,5%	6.0%
PF-320 cps-10.4	40.39	2579	2726	3230
ser sps err	45.41	3650	3943	4221
ALS-PF-84 cps-12.1	40.39	2568	2596	3010
ALS-II-04 Cps-12.1	45.41	3731	3373	3834
ALS-PF-100 cps-10.6	40.39	2112	2309	2383
•	45.41	2925	3337	3756
ALS-PF-270 cps-12.3	40.39	2190	2836	2866
	45.41	3691	3673	3852
ALS-PF-300 cps-10.6	40.39	2474	2571	2780
	45.41	2938	4170	3931
ALS-PF-850 cps-12.2	40.39	2347	2651	2651
11 030 cpc 11.1	45.41	3238	3583	3588
ALS-PF-540 cps-10.6	40.39	2190	2356	2538
1110 11 5 to opo 1010	45.41	3409	3475	3671

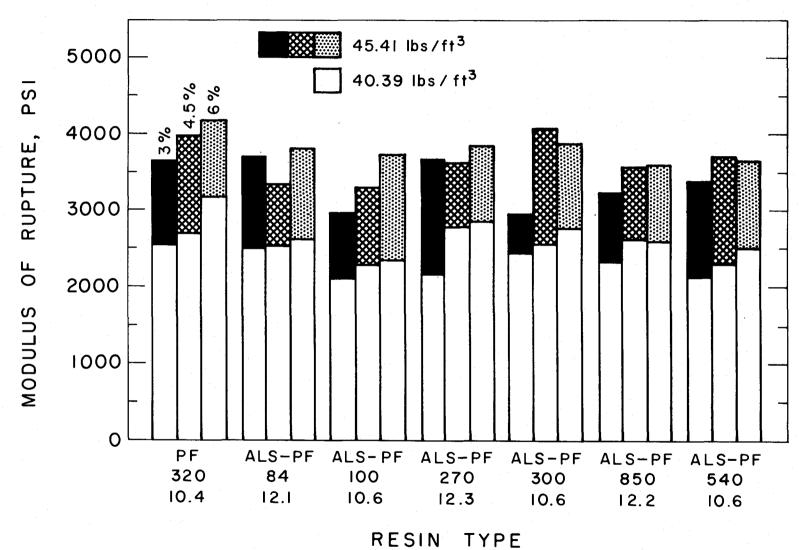


Figure 5. Modulus of rupture values as affected by resin viscosity, resin rate, and board density.

resin, both increasing board strength.

The effect resin viscosity had on MOR values can be observed in Figures 5 and 6. It must be pointed out, however, that Figure 6 considers only the effect of resin viscosity on MOR values. The effect of resin rate and board density would also have to be considered (Figure 5) for a true picture to result due to the significance of the interaction term. Resin viscosity appeared to exhibit little effect on MOR properties. These results agree with those reported by Snyder, Rice, and Hart (1967) in which the authors stated that resin viscosity was not found to have a significant effect on static bending properties.

Tables 8 and 9 help to point out the effect resin type had on bending strength values. The results would indicate that the substituted resins can be successfully used without detrimental effects to board properties.

Modulus of Elasticity

Observation of the analysis of variance table for MOR (Appendix Table B-3) reveals that the main effects show significant differences at the .95 and .99 levels, and that only the three way interaction of viscosity × rate × density is significant (.95 level). As was the case for IB and MOR properties, this interaction indicates that MOE values are dependent not only upon resin viscosity, but upon resin application rate and board density as well. Table 10 contains the adjusted MOE values for the viscosity × rate × density interaction. Figure 7 is a graphical representation of the data contained in Table 10, helping to illustrate the effect resin viscosity and resin application rate have on

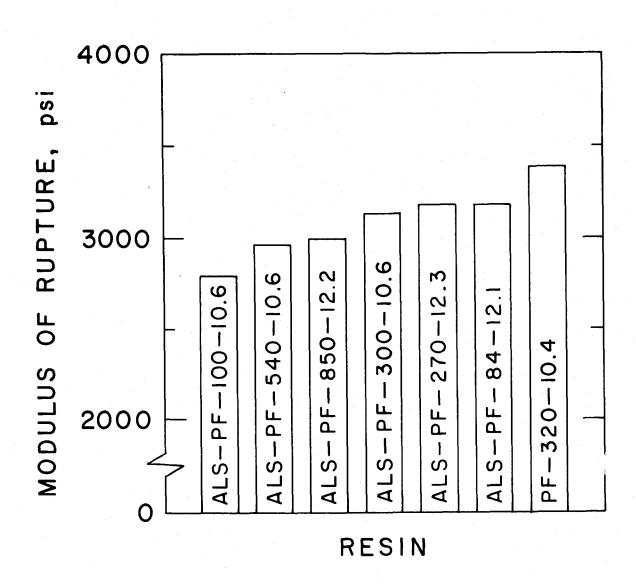


Figure 6. Effect of resin viscosity on modulus of rupture properties.

Table 8. Effect of Resin Type on MOR Values; 40.39 lbs/cu.ft. Adjusted Board Density.

Resin Type	Resin Rate (%)	MOR (psi)	Change from Control (psi)
PF-320 cps-10.4	3.0	2579	-
	4.5	2726	
•	6.0	3230	
ALS-PF-84 cps-12.1	3.0	2568	-11.0
	4.5	2596	-130.0
	6.0	3010	-220.0
ALS-PF-270 cps-12.3	3.0	2190	-389.0
ALD-11 270 Cps 12.5	4.5	2837	+111.0
	6.0	2866	-364.0
ALS-PF-850 cps-12.2	3.0	2347	-232.0
MED-11 030 CPS 12.2	4.5	2651	-75.0
	6.0	2651	-579.0
ALS-PF-100 cps-10.6	3.0	2112	-467.0
And II 100 Cps 10.0	4.5	2309	-417.0
and the second of the second o	6.0	2383	-847.0
ALS-PF-300 cps-10.6	3.0	2474	-105.0
ALS-11-300 Cps-10.0	4.5	2571	-155.0
	6.0	2780	-450.0
ALS-PF-540 cps-10.6	3.0	2190	-389.0
ADD-11-240 Ch2-10:0	4.5	2356	-370.0
	6.0	2538	-692.0

Cutoff value for significant differences = 688 psi.

Table 9. Effect of Resin Type on MOR Values; 45.41 lbs/cu.ft. Adjusted Board Density.

Resin Type	Resin Rate (%)	MOR (psi)	Change from Control (psi)
		0.050	
PF-320 cps-10.4	3.0	3650	
	4.5	3943	
	6.0	4221	
ALS-PF-84 cps-12.1	3.0	3731	+81.0
•	4.5	3373	-570.0
	6.0	3834	-387.0
ALS-PF-270 cps-12.3	3.0	3691	+41.0
MED 11 270 CPS 12.3	4.5	3673	-270.0
	6.0	3852	-369.0
ALS-PF-850 cps-12.2	3.0	3238	-412.0
ALD 11 030 Cp3 12.2	4.5	3583	-360.0
	6.0	3588	-633.0
ALS-PF-100 cps-10.6	3.0	2925	-725.0
ABB II 100 CPB 1010	4.5	3337	-606.0
	6.0	3756	-465.0
ALS-PF-300 cps-10.6	3.0	2938	-712.0
WF9-11-200 ch2-1010	4.5	4170	+227.0
	6.0	3931	-290.0
ALS-PF-540 cps-10.6	3.0	3409	-241.0
МДВ 11 Э40 СРВ 1010	4.5	3748	-195.0
	6.0	3671	-550.0

Cutoff value for significant differences = 688 psi.

Table 10. Modulus of Elasticity Values for the Viscosity \times Rate \times Density Interaction.

	Adjusted		MOE (psi)	
	Board Density	Resi	n Application	Rate
Resin Type	(lbs/cu.ft.)	3.0%	4.5%	6.0%
PF-320 cps-10.4	40.39	409105	429970	455629
	45.41	509077	578999	575853
ALS-PF-84 cps-12.1	40.39	380243	371982	431424
ALS-11-04 Cps-12.1	45.41	520092	460567	503685
ALS-PF-100 cps-10.6	40.39	348057	356061	394633
	45.41	466773	491901	554038
ALS-PF-270 cps-12.3	40.39	330594	399416	405122
er en	45.41	503165	495005	495138
ALS-PF-300 cps-10.6	40.39	401395	410551	431518
-	45.41	471261	572527	554612
ALS-PF-850 cps-12.2	40.39	348483	383622	367572
•	45.41	450273	445344	472720
ALS-PF-540 cps-10.6	40.39	374807	386705	416445
	45.41	520106	551014	544135

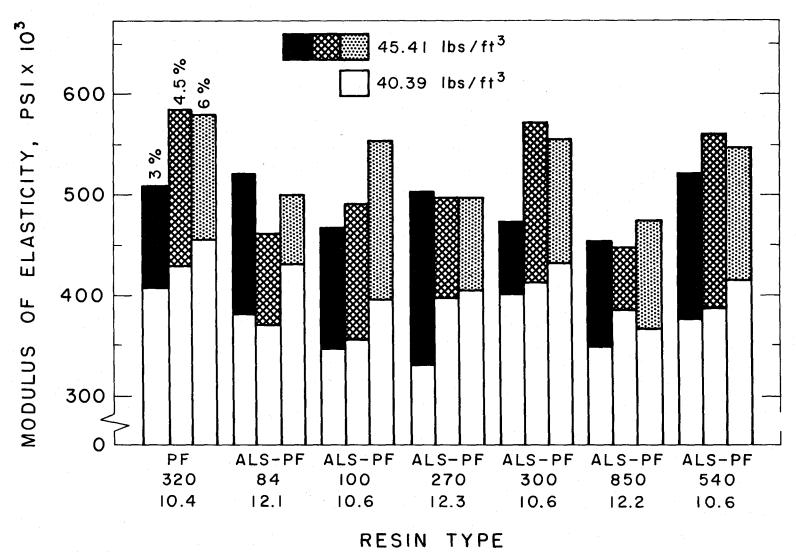


Figure 7. Modulus of elasticity values as affected by resin viscosity, resin rate, and board density.

MOE values for a given board density.

Table 10 and Figure 7 indicate that a general increase in MOE values will result with an increase in resin content from 3 to 6 percent. However, as was the case for MOR test results, this increase in strength is significant in only two cases; the high density boards using the 100 cps and 300 cps ALS-PF resins with resin application rates of 3 and 6 percent and 3 and 4 1/2 percent respectively. The cutoff value for significant differences was 83,944 psi. Post (1958, 1961) found that bending stiffness was not materially influenced by resin content in his studies with oak flakeboard. He found flake size and orientation to be the controlling factors.

Figure 7 illustrates the significant effect increasing board density had on MOE values. The results are in keeping with those found for IB and MOR properties.

The effect resin viscosity had on MOE values is comparable to that found for bending strength values. Resin viscosities falling in the middle and upper end of the range studied (270 cps, 300 cps, and 540 cps) appear to have been optimum in the development of MOR and MOE strength properties. Figure 8 helps to illustrate this point. It should again be noted, however, that the illustration considers only the effect resin viscosity had on MOE values.

The effect of resin type on MOE values is summarized in Tables 11 and 12. As was the case for MOR test results, use of the substituted resins (ALS-PF) resulted in bending stiffness values inferior to those obtained using the commercial phenolic resin. However, these differences were not significant.

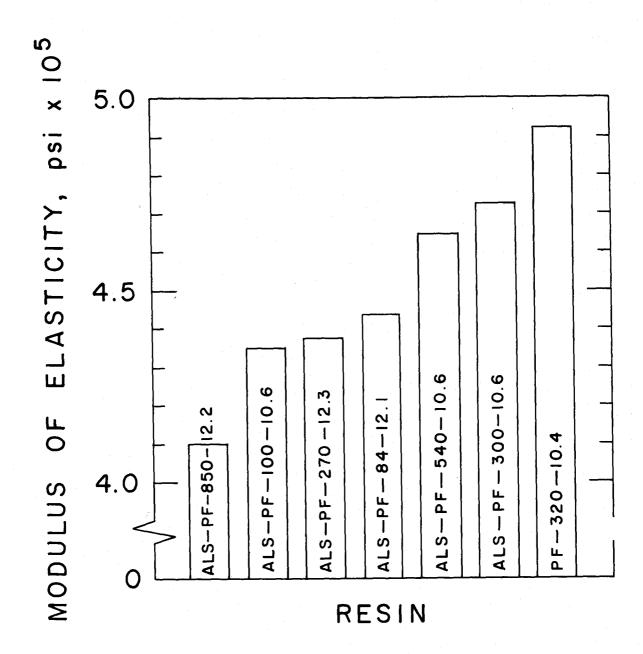


Figure 8. Effect of resin viscosity on modulus of elasticity properties.

Table 11. Effect of Resin Type on MOE Values; 40.39 lbs/cu.ft. Adjusted Board Density.

Resin Type	Resin Rate (%)	MOE (psi)	Change from Control (psi)
		/00±05	
PF-320 cps-10.4	3.0	409105 429970	. -
	4.5 6.0	455629	-
ALS-PF-84 cps-12.1	3.0	380243	-28862
THE OF CPU II.I	4.5	371982	-57988
	6.0	431424	-24205
ALS-PF-270 cps-12.3	3.0	330594	-78511
•	4.5	399416	-30554
	6.0	405122	-50507
ALS-PF-850 cps-12.2	3.0	348483	-60622
-	4.5	383622	-46348
	6.0	367572	-88057
ALS-PF-100 cps-10.6	3.0	348057	-61048
	4.5	356061	-73909
	6.0	394633	-60996
ALS-PF-300 cps-10.6	3.0	401395	-7710
-	4.5	410551	-19419
	6.0	431518	-24111
ALS-PF-540 cps-10.6	3.0	374807	-34298
	4.5	386705	-43265
	6.0	416445	-39184

Cutoff value for significant differences = 83,944 psi.

Table 12. Effect of Resin Type on MOE Values; 45.41 lbs/cu.ft. Adjusted Board Density.

Resin Type	Resin Rate (%)	MOE (psi)	Change from Control (psi)
PF-320 cps-10.4	3.0	509077	
	4.5	578999	-
	6.0	575853	
ALS-PF-84 cps-12.1	3.0	520092	+11015
	4.5	460557	-118442
we'r	6.0	503685	-72168
ALS-PF-270 cps-12.3	3.0	503165	-5912
-	4.5	495005	-83994
	6.0	495138	-80715
ALS-PF-850 cps-12.2	3.0	450273	-58804
· · · · · · · · · · · · · · · · · · ·	4.5	445344	-133655
	6.0	472720	-103133
ALS-PF-100 cps-10.6	3.0	466773	-42304
	4.5	491901	-87098
	6.0	554038	-21815
ALS-PF-300 cps-10.6	3.0	471261	-37816
•	4.5	572527	-6472
	6.0	554612	-21241
ALS-PF-540 cps-10.6	3.0	520106	+11029
· ·	4.5	551014	-27985
	6.0	544135	-31718

Cutoff value for significant differences = 83,944 psi.

Dimensional Stability Properties

Linear Expansion

Observation of the analysis of variance table for LE (Appendix Table B-4) indicates that only the main effects of resin viscosity and resin rate show significant differences (.95 and .99 levels). The significance of these terms means that LE values are dependent on resin viscosity and resin application rate respectively. Figures 9 and 10 help to illustrate these effects.

Resin viscosities in the mid-range of those studied gave reductions in LE, while resin viscosities below 270 cps and above 540 cps were associated with increasing LE values. Tukey's method of multiple comparisons indicated no significant differences in LE properties using the commercial phenolic resin and the 270 cps, 300 cps, and 540 cps substituted resins. These results help to further emphasize the potential of these resins for future use.

Figure 10 illustrates the effect resin application rate had on LE values. There were no significant reductions in LE associated with increased resin application rates.

Thickness Swell

The analysis of variance table for TS (Appendix Table B-5) shows that the main effects as well as the interaction terms exhibit significant differences at the .95 level or above. The significance of the three way interaction of viscosity × rate × density indicates that board TS values are not only dependent upon resin viscosity, but upon

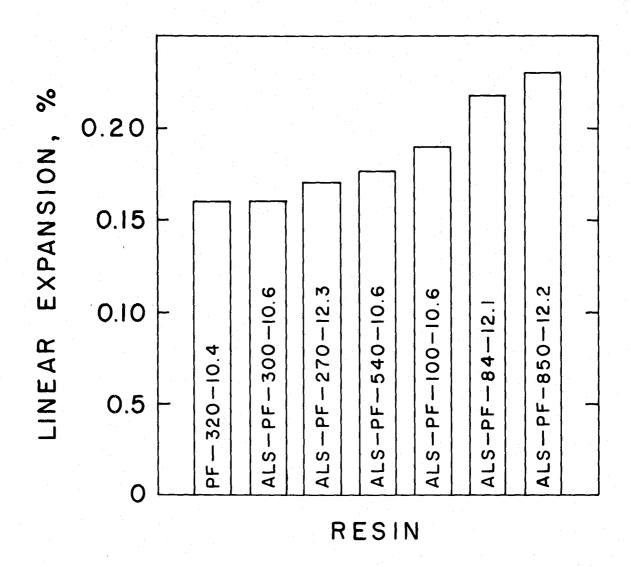


Figure 9. Effect of resin viscosity on linear expansion properties.

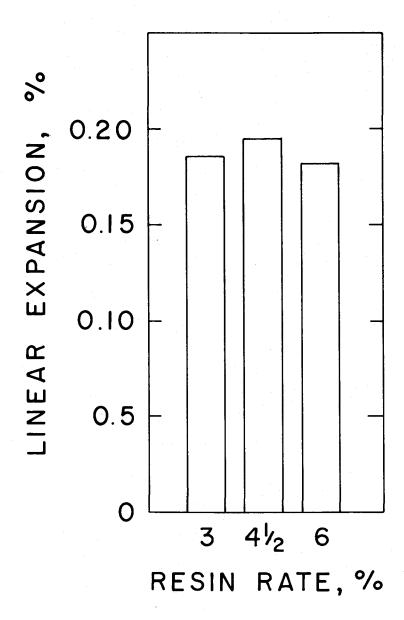


Figure 10. Effect of resin rate on linear expansion properties.

resin application rate and board density as well. Table 13 contains average TS values for the viscosity \times rate \times density interaction. TS values are illustrated in Figures 11 and 12.

A general reduction in thickness swelling was associated with increasing resin content, as would be expected. These results follow with those found by Lehmann (1970), Jorgensen and Odell (1961), and Larmore (1959) as discussed earlier. All authors reported reduced thickness swell values with increasing resin spreads.

An increase in nominal board density from 40 to 45 lbs/cu.ft. resulted in a general increase in TS values as can be observed from a comparison of Figures 11 and 12. Higher density boards possess greater compressive set than lower density boards made of the same wood furnish. Release of these compressive stresses is readily triggered by moisture gain in the particleboard panel. Thickness swelling is also related to the swelling properties of the wood particles themselves.

The effect of resin viscosity on TS values is illustrated in Figure 13. It must be noted here that the effect of resin rate and board density have not been considered. The results indicate that the 84 cps ALS-PF resin gave the lowest thickness swell values. The results also indicate that the higher pH ALS-PF resins exhibit reductions in TS as compared to the lower pH ALS-PF resins. High pH resins improve bond quality in some undefined manner.

Tables 14 and 15 compare the TS values obtained using the commercial phenolic resin to those obtained using the ammoniated-lignosulfonate-phenol-formaldehyde resins. The high pH ALS-PF resins gave TS values comparable to or better than the phenolic resin. On the other hand, the lower pH ALS-PF resins were generally inferior.

Table 13. Thickness Swell Values for the Viscosity \times Rate \times Density Interaction.

	Avorago		TS (%)	
	Average Board Density	Resin Application Rate		
Resin Type	(lbs/cu.ft.)	3.0%	4.5%	6.0%
DE 220 10 /	40.41	13.03	14.33	12.67
PF-320 cps-10.4	45.66	16.63	14.97	16.17
ALS-PF-84 cps-12.1	40.41	14.88	13.67	12.72
•	45.66	15.23	13.68	14.73
ALS-PF-100 cps-10.6	40.41	18.93	15.83	14.17
	45.66	18.70	18.73	16.77
ALS-PF-270 cps-12.3	40.41	15.62	14.93	15.08
•	45.66	16.63	14.70	14.08
ALS-PF-300 cps-10.6	40.41	16.83	16.30	13.03
	45.66	18.63	16.03	13.63
ALS-PF-850 cps-12.2	40.41	15.95	15.45	14.15
	45.66	16.97	14.93	16.63
ALS-PF-540 cps-10.6	40.41	20.47	18.40	15.50
	45.66	22.77	18.63	14.83

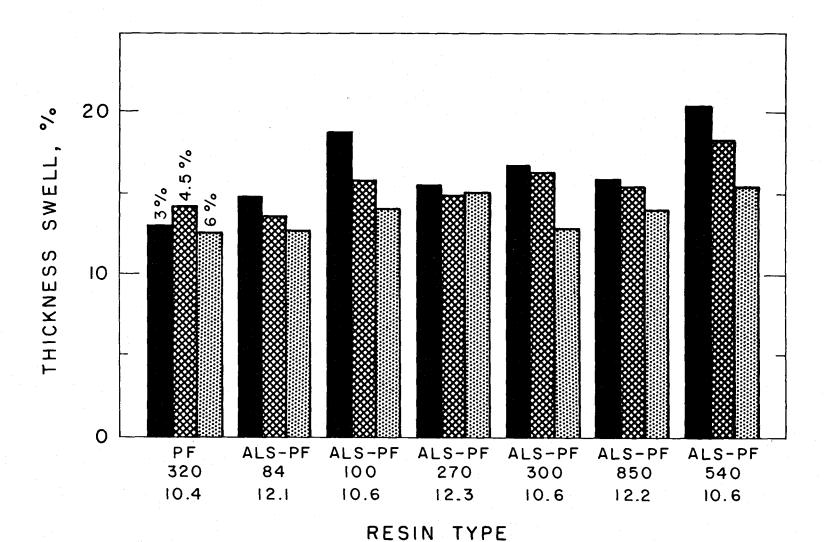


Figure 11. Thickness swell values as affected by resin viscosity and resin rate; 40 lbs/cu.ft. nominal board density.

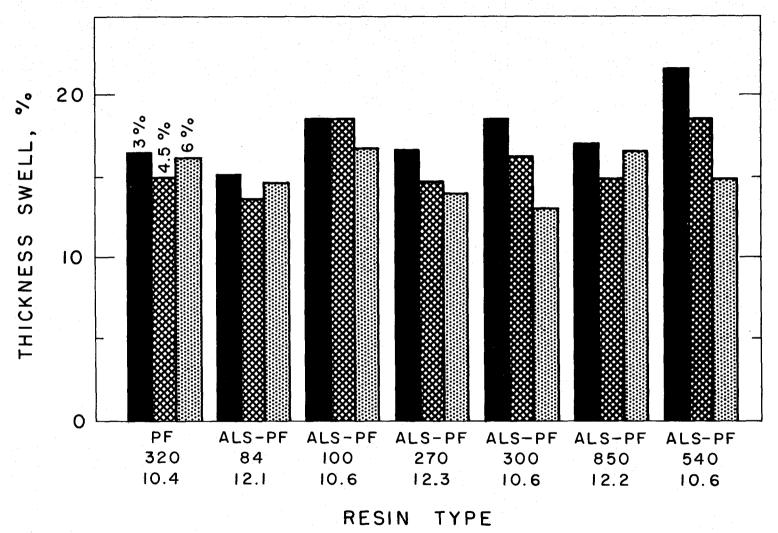


Figure 12. Thickness swell values as affected by resin viscosity and resin rate; 45 lbs/cu.ft. nominal board density.

THICKNESS SWELL, %

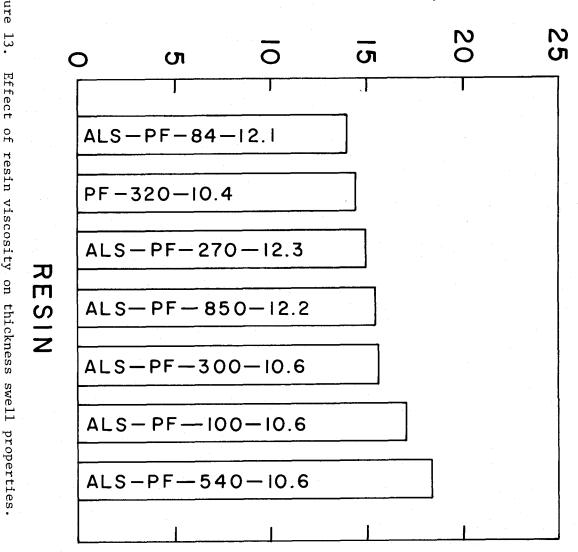


Table 14. Effect of Resin Type on TS Values; 40 lbs/cu.ft. Nominal Board Density.

Resin Type	Resin Rate (%)	TS (%)	Change from Control (%)
PF-320 cps-10.4	3.0	13.03	· ·
	4.5	14.33	<u> </u>
	6.0	12.67	
ALS-PF-84 cps-12.1	3.0	14.88	+1.85
	4.5	13.67	-0.66
	6.0	12.72	+0.05
ALS-PF-270 cps-12.3	3.0	15.62	+2.59
	4.5	14.93	+0.60
	6.0	15.08	+2.41
ALS-PF-850 cps-12.2	3.0	15.95	+2.92
	4.5	15.45	+1.12
	6.0	14.15	+1.48
ALS-PF-100 cps-10.6	3.0	18.93	+5.90
	4.5	15.83	+1.50
	6.0	14.17	+1.50
ALS-PF-300 cps-10.6	3.0	16.83	+3.80
	4.5	16.30	+1.97
	6.0	13.03	+0.36
ALS-PF-540 cps-10.6	3.0	20.47	+7.44
•	4.5	18.40	+4.07
	6.0	15.50	+2.83

Cutoff value for significant differences = 2.59%.

Table 15. Effect of Resin Type on TS Values; 45 lbs/cu.ft. Nominal Board Density.

Resin Type	Resin Rate (%)	TS (%)	Change from Control (%)
Resili Type	(%)	(/8/)	
PF-320 cps-10.4	3.0	16.63	-
11 320 cps 10.4 ·	4.5	14.97	waterja.
	6.0	16.17	_
ALS-PF-84 cps-12.1	3.0	15.23	-1.40
	4.5	13.68	-1.29
	6.0	14.73	-1.44
ALS-PF-270 cps-12.3	3.0	16.63	0
	4.5	14.70	-0.27
	6.0	14.08	-2.09
ALS-PF-850 cps-12.2	3.0	16.97	-0.34
	4.5	14.93	-0.04
	6.0	16.63	+0.46
ALS-PF-100 cps-10.6	3.0	18.70	+2.07
•	4.5	18.73	+3.76
-	6.0	16.77	+0.60
ALS-PF-300 cps-10.6	3.0	18.63	+2.00
	4.5	16.03	+1.06
	6.0	13.63	-2.54
ALS-PF-540 cps-10.6	3.0	22.77	+6.14
•	4.5	18.63	+3.66
	6.0	14.83	-1.34

Cutoff value for significant differences = 2.59%.

Weight Gain

Observation of the analysis of variance table for WG (Appendix Table B-6) indicates that the main effects of resin viscosity and resin rate, as well as the interactions show highly significant differences (.95 and .99 levels). The density term is not significant. Again, the three way interaction will be considered because it contains all the information available. The significance of this term indicates that weight gain values are dependent upon resin viscosity, as well as resin application rate and board density. Table 16 contains the average WG values for the viscosity × rate × density interaction, and this data is illustrated in Figures 14 and 15.

Figures 14 and 15 indicate a general increase in WG values as resin content increases from 3 to 6 percent. However, these increases are generally not significant.

As was the case for TS values, boards with 84 cps ALS-PF resin exhibited the smallest percent gain in weight as can be seen in Figure 16, although the differences between this viscosity and the 270 cps and 850 cps ALS-PF resins were small. It should be noted again that this figure only takes into account the effect of resin viscosity on WG values. Figure 16 also shows that the higher pH ALS-PF resins exhibited better WG values than the lower pH ALS-PF resins.

Observation of Tables 17 and 18 points out the effect resin type had on WG values. The 84 cps, 270 cps, and 850 cps ALS-PF resins gave weight gain values better than those obtained with the commercial phenolic resin. These results help to reinforce the suitability of such substituted resins for manufacturing processes.

Table 16. Weight Gain Values for the Viscosity \times Rate \times Density Interaction.

	Average		WG (%)		
	Board Density	Resin Application Rate			
Resin Type	(lbs/cu.ft.)	3.0%	4.5%	6.0%	
PF-320 cps-10.4	40.41	12.07	13.07	12.97	
11 320 Cp3 10.4	45.66	12.67	12.77	14.63	
ALS-PF-84 cps-12.1	40.41	11.63	11.78	11.78	
•	45.66	11.38	11.33	12.25	
ALS-PF-100 cps-10.6	40.41	14.17	13.87	13.73	
··· •	45.66	13.40	14.27	14.03	
ALS-PF-270 cps-12.3	40.41	11.53	12.62	12.38	
	45.66	11.52	11.77	11.78	
ALS-PF-300 cps-10.6	40.41	13.07	13.90	13.07	
· ·	45.66	13.00	12.70	12.70	
ALS-PF-850 cps-12.2	40.41	11.43	11.68	12.02	
·	45.66	11.68	11.38	12.27	
ALS-PF-540 cps-10.6	40.41	13.27	13.43	13.50	
	45.66	13.43	13.70	13.47	

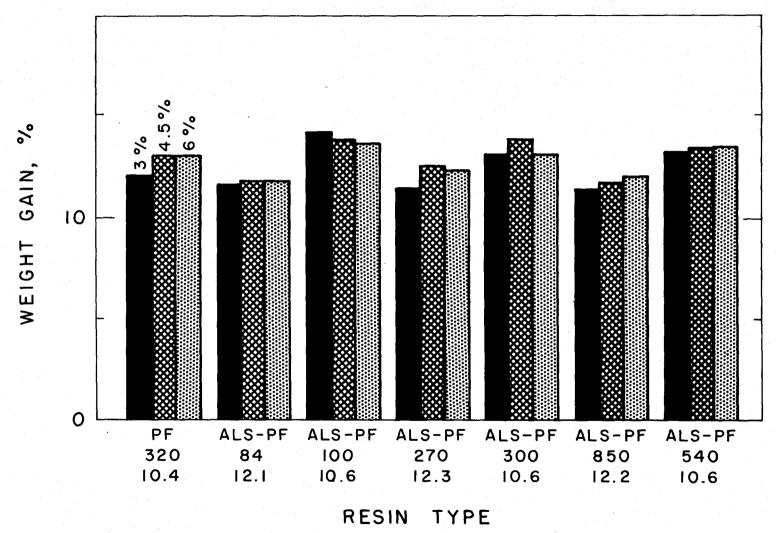


Figure 14. Weight gain values as affected by resin viscosity and resin rate; 40 lbs/cu.ft. nominal board density.

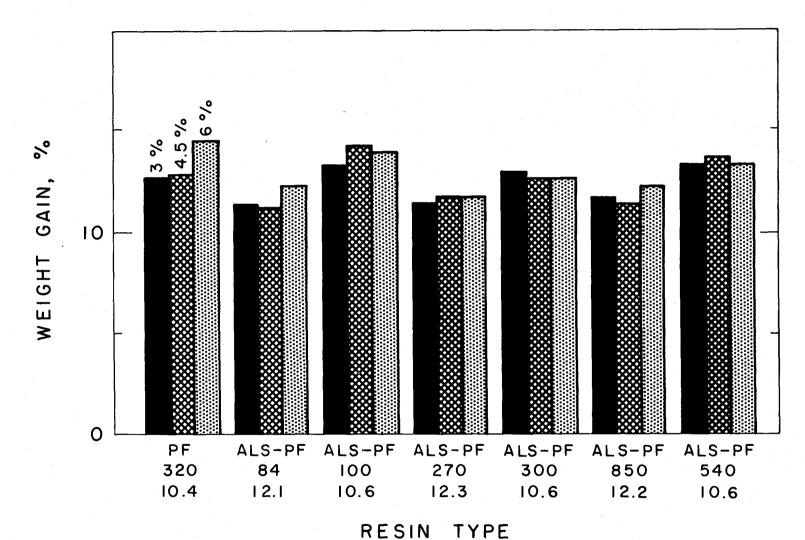


Figure 15. Weight gain values as affected by resin viscosity and resin rate; 45 lbs/cu.ft. nominal board density.

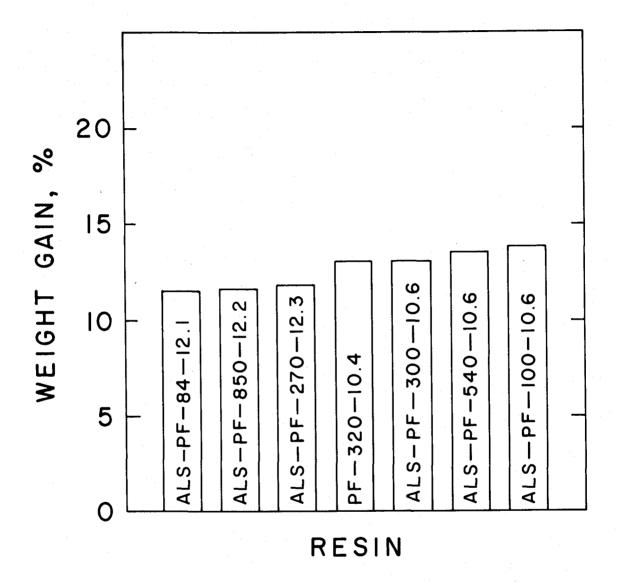


Figure 16. Effect of resin viscosity on weight gain properties.

Table 17. Effect of Resin Type on WG Values; 40 lbs/cu.ft. Nominal Board Density.

Resin Type		Resin Rate (%)		WG (%)	ange from Control (%)
					
PF-320 cps-10.4		3.0		12.07	_
		4.5		13.07	·
		6.0		12.97	_
ALS-PF-84 cps-12.1		3.0		11.63	-0.44
		4.5		11.78	-1.29
		6.0	e	11.78	-1.19
ALS-PF-270 cps-12.3		3.0		11.53	-0.54
•		4.5		12.62	-0.45
		6.0		12.38	-0.59
ALS-PF-850 cps-12.2		3.0		11.43	-0.64
		4.5		11,68	-1.39
		6.0		12.02	-0.95
ALS-PF-100 cps-10.6		3.0		14.17	+2.10
		4.5		13.87	+0.80
		6.0		13.73	+0.76
ALS-PF-300 cps-10.6		3.0		13.07	+1.00
		4.5		13.90	+0.83
		6.0		13.07	+0.10
ALS-PF-540 cps-10.6		3.0		13.27	+1.20
		4.5		13.43	+0.36
		6.0		13.50	+0.53

Cutoff value for significant differences = 0.66%.

Table 18. Effect of Resin Type on WG Values; 45 lbs/cu.ft. Nominal Board Density.

Resin Type	Resin Rate (%)	WG (%)	Change from Control (%)
Resin Type	(/6/	(70)	
PF-320 cps-10.4	3.0	12.67	· _
21 020 aps 200	4.5	12.77	
	6.0	14.63	
ALS-PF-84 cps-12.1	3.0	11.38	-1.29
	4.5	11.33	-1.44
	6.0	12.25	-2.38
ALS-PF-270 cps-12.3	3.0	11.52	-1.15
	4.5	11.77	-1.00
	6.0	11.78	-2.85
ALS-PF-850 cps-12.2	3.0	11.68	-0.99
The second of th	4.5	11.38	-1.39
	6.0	12.27	-2.36
ALS-PF-100 cps-10.6	3.0	13.40	+0.73
	4.5	14.27	+1.50
	6.0	14.03	+0.60
ALS-PF-300 cps-10.6	3.0	13.00	+0.33
	4.5	12.70	-0.07
	6.0	12.70	-1.93
ALS-PF-540 cps-10.6	3.0	13.43	+0.76
	4.5	13.70	+0.93
	6.0	13.47	-1.16

Cutoff value for significant differences = 0.66%.

V. SUMMARY AND CONCLUSIONS

A study was conducted to determine the effect of resin type, resin viscosity, and resin application rate on the physical properties (IB, MOR, MOE, LE, TS, and WG) of a 1/4 inch randomly oriented oak flakeboard to be used as a core-stock in veneer-particleboard composite panels. A commercial phenol-formaldehyde resin and six ammoniated-lignosulfonate-phenol-formaldehyde resins of varying viscosity were used with resin application rates of 3, 4 1/2, and 6 percent resin solids based on ovendry wood weight. Nominal board densities were 40 and 45 lbs/cu.ft.

Based on the results of this study it may be concluded that:

- (1) Ammoniated-lignosulfonate-phenol-formaldehyde resins can be effectively used in the laboratory with resultant board physical properties comparable to those obtained using a commercial phenolic resin.
- (2) In the range of resin viscosities studied, increasing resin viscosity resulted in increasing IB properties, and decreasing MOR, MOE, TS, and WG properties. In the case of IB, TS, and WG properties, the higher pH ALS-PF resins gave physical properties better than those obtained using the lower pH ALS-PF resins.
- (3) An increase in board physical properties can be associated with an increase in percentage of resin content. IB properties continued to increase with an increase in resin content from 3 to 6 percent, however, MOR and MOE properties exhibited a leveling off tendency, particularly in the range from 4 1/2 to 6 percent. LE properties showed no statistically significant differences between resin rates, while TS properties exhibited a general improvement with increasing resin content. Percent WG generally increased with percent resin.

BIBLIOGRAPHY

- Bryant, B. S. and J. M. Garcia. 1967. Factors affecting bleedthrough of phenolic resin adhesive in hardwood plywood. Wood Science and Technology 1:304-320.
- Forss, K. G. and A. F. Fuhrmann. 1976. KARATEX the lignin-based adhesive for plywood, particle and fibre board. Paperi ja Puu 59(11):817-824.
- Holderby, J. M., H. S. Olson and W. H. Wegener. 1967. Thermosetting adhesives from electrodialyzed lignosulfonates. TAPPI 50(9): 92A-94A.
- Johansen, T. A. 1971. Particleboard bonded with sulphite liquor. Fifth Particleboard Sympos. Proc. Washington State University, Pullman. Pp. 11-26.
- Jorgensen, R. N. and R. L. Odell. 1961. Dimensional stability of oak flake board as affected by particle geometry and resin spread. Forest Products Journal 11(10):463-466.
- Larmore, F. D. 1959. Influence of specific gravity and resin content on properties of particle board. Forest Products Journal 9(4): 131-134.
- Lehmann, W. F. 1970. Resin efficiency in particleboard as influenced by density, atomization, and resin content. Forest Products Journal 20(11):48-54.
- Post, P. W. 1958. Effect of particle geometry and resin content on bending strength in oak flake board. Forest Products Journal 8(10):317-322.
- . 1961. Relationship of flake size and resin content to mechanical and dimensional properties of flake board. Forest Products Journal 11(1):34-37.
- Rice, J. T. 1965. The effect of urea-formaldehyde resin viscosity on plywood wood bond durability. Forest Products Journal 15(3): 107-112.
- Roffael, E. and W. Rauch. 1971. Production of particleboards using black liquor as binder. II. A new and fast method for the production of particleboards. Holzforschung 25(5):149-155.
- . 1973. Production of particleboard using black liquor as binder. IV. Use of sulfite liquor in combination with alkaline phenolic resins. Holzforschung 27(6):214-217.

- Shen, K. C. 1974. Modified powdered spent sulfite liquor as binder for exterior waferboard. Forest Products Journal 24(2):38-44.
- . 1974. Acidified spray-dried spent sulfite liquor as binder for exterior waferboard. Eighth Particleboard Sympos. Proc., Washington State University, Pullman. Pp. 231-246.
- Snyder, J. L., J. T. Rice and C. A. Hart. 1967. Influence of selected bonding factors on flakeboard properties. Forest Products Journal 17(8):49-57.
- Sullivan, J. D. and W. L. Harrison. 1965. Rate of viscosity change for a urea formaldehyde resin. Forest Products Journal 15(11):480-484.
- Talbott, J. W. and T. M. Maloney. 1957. Effect of several production variables on modulus of rupture and internal bond strength of boards made of green Douglas-fir planer shavings. Forest Products Journal 7(10):395-398.
- U. S. Forest Products Laboratory. 1977. Critical literature review of relationships between processing parameters and physical properties of particleboard. Myron W. Kelly. U.S.D.A. Forest Service General Technical Report FPL-10. Madison, Wisconsin. 66 p.

STUDY TWO: The Effect of Resin Wettability and Resin Molecular Weight Distribution on Bonding in Oak Flakeboard

I. INTRODUCTION

The means by which an adhesive system anchors itself to a wood substance is an obscure and complex process. A complete theory of bond formation remains to be developed, however, there have been two possibilities debated at length. The first involves the flow of the adhesive into the pores and cavities of the wood and upon setting, locking itself into place. This phenomenon is known as mechanical adhesion. The second theory involves the chemical forces in the adhesive molecules and those in the surface molecules of the wood. These chemical forces attract each other with great power. This theory has been termed chemical or specific adhesion due to the chemical forces brought into action. Chemical adhesion is considered to contribute the major strength to an adhesive bond. Mechanical adhesion is necessary to achieve anchorage below the wood surface, reinforcing the adhesive joint.

The action of chemical adhesion is associated with the wetting properties of both the wood and the adhesive, while the action of mechanical adhesion can be associated with the effective void size of the wood substrate, and the size of the resin molecules. This study deals with the effect resin wetting and resin molecular weight properties exhibit on bond quality in oak flakeboard.

Statement of Objective

The objective of this study was to determine the relationship of resin wetting and resin molecular weight properties with the quality of adhesive bonds in a randomly oriented oak flakeboard to be used as a core-stock in veneer-particleboard composite panels.

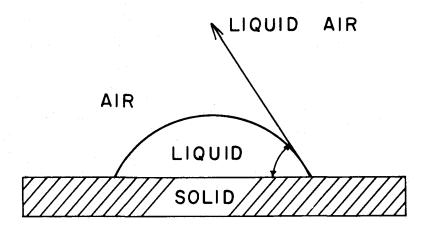
II. REVIEW OF LITERATURE

Wettability as Related to Bond Quality

If liquid and solid surfaces come into contact and the molecules of the solid surface attract the molecules of the liquid surface more strongly than the liquid molecules attract each other, true contact may be established and the liquid will "wet" the solid. The degree of wettability is dependent on the magnitude of the forces involved (Bodig, 1962).

Wettability can be measured by the contact angle between the solidliquid interface and the liquid-air surface (Figure 17). When the work of adhesion of liquid to solid equals the work of cohesion of the liquid, the contact angle is zero; that is, complete wettability has been established. On the other hand, if the contact angle is 90° there is no wettability at all. The wettability can be negative when the contact angle is between 90° and 180° (Bodig, 1962).

The study of the wetting of solids by contact angle measurements has become an important tool in the study of adhesion and the related phenomena of spreading and bond formation. Freeman (1959), in a study involving 22 species of hardwoods of varying specific gravity and pH, found that wettability as measured by the contact angle method was related to glue bond quality. Shear strength, percent wood failure, and shear ratio were used as glue bond evaluators. Freeman found that below 0.80 specific gravity (50 lbs/cu.ft. density), shear strength increased with increasing wettability. In the region above 0.80 specific gravity he found wettability to have a positive relationship with



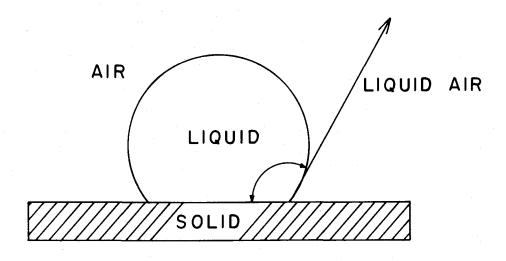


Figure 17. Measurement of contact angle.

all glue bond evaluators; glue bond quality increased as wettability increased. Freeman concluded wettability was closely related to glue bond quality.

Bodig (1962) examined the relationship between wettability and glue bond quality of plywood panels constructed of five species of Philippine luan. Wettability was measured using the cosine contact angle and corrected water-absorption height methods. He found the corrected water-absorption height value to be a better indicator of gluability than the cosine contact angle value, although both methods exhibited a good degree of correlation between wettability and gluability expressed in terms of shear load (psi). In both cases the relationship was positive. Bodig concluded that the degree of correlation between wettability and gluability reinforces the validity of the specific (chemical) adhesion theory; it is possible to predict relative gluability values of an unknown species by measuring its wettability.

In a study involving the interaction of wood and adhesive variables, Bryant (1968) found that bond quality is influenced by the wettability of wood by the resin, and by chemical interactions between the resin and the wood surface. Flakes dried in a commercial drum dryer were found to produce boards with higher physical properties than those produced from air dried flakes. Greater resin efficiency was obtained using the mill dried flakes, the surfaces of which were rendered less wettable due to the drastic drying conditions used. Bryant felt that the resin binder may have wetted too well on the gently dried material. He concluded that while good wetting is essential to good bonding, in particleboard good wetting when coupled with too much flow results in a

weakened bond. The adhesive spreads as a film and is no longer available as a discrete droplet for "spot welding" together adjacent surfaces.

Although the previous studies would indicate that wettability is a good predictor of bond quality, Wellons (1968) points out that many of the contact angles reported for liquids on wood do not match the gluing conditions they are supposed to predict. Many researchers have used distilled water. When water is used as the wetting medium it is absorbed by the wood, resulting in swelling of the wood structure and a change in wettability with time. For adequate prediction of the performance of an adhesive, the wetting liquid certainly needs to match the adhesive in its ability to solubilize components on the wood surface, swell the wood surface, and so on.

These difficulties with wettability data are illustrated in the research of Hse (1972). He reported on the wettability of southern pine veneer by measuring the contact angles made by 36 exterior-grade phenol-formaldehyde resins having a wide range of physio-chemical properties. Discussed were the effects of resin formulation on wettability as measured by contact angles as well as the relation between contact angle and bond quality. Hse found that contact angles decreased with increasing amounts of NaOH in the adhesive (increased swelling power of the adhesive); that contact angles increased with increasing formaldehyde content (higher resin molecular weights); and that glue line performance as evaluated by shear strength, percent wood failure, and percent delamination increased with increasing contact angle. These results appear opposite to the general opinion that a low contact angle is desirable because of its favorable effect on wetting and spreading.

Hse explained these results by noting the high permeability of southern pine, and that excess penetration is one of the most important causes of poor glue bond quality. A resin with a high contact angle does not penetrate excessively, and may give the best compromise conditions of wetting, spreading, and penetration.

In light of the above contradictions, results of studies of this nature should be interpreted with caution and care. We need to keep in mind that although good wetting is a necessary prerequisite to good bonding, good wetting does not always mean that a bond of high strength will result.

Molecular Weight Distribution as Related to Bond Quality

After the adhesive system has spread on the wood surface, it must penetrate the wood substance. Wood substance behaves as a three-dimensional network of polymeric materials. This network expands to maximum size at 100 percent relative humidity. The swollen network in turn contains submicroscopic voids in which water is condensed. Tarkow, Feist, and Southerland (1966) state that some average or effective void size probably imposes an upper limit to the size of molecule of a water-borne solute capable of diffusing into or through the cell walls. Although the interaction of adhesives with wood substance is generally believed to be a surface adsorption phenomenon, the actual interaction may occur within a "surface region". In such a case effective void size would be important in determining degree of penetration.

Tarkow, Feist, and Southerland looked at the effective void size of green Sitka spruce heartwood cross sections. The 2- by 2- by 0.020-

inch sections were first extracted with boiling water and then equilibrated in aqueous polyethylene glycol solutions (1 to 4 percent concentration) of various molecular weights as shown in Table 19. The specimens were soaked for 2 to 5 days at 22°C.

If the value for nonsolvent-water content (Δ) was zero, the solute would have complete access to the wood substance; it would distribute itself in the cell wall in the same ratio with water as it does in the external solution. As the value of Δ increases, solute access decreases. The authors found nonsolvent-water content to increase with molecular weight of solute and then become constant when the molecular weight exceeded 3,000. The authors concluded that the limiting size for the penetration of a water soluble material into green wood substance is that of a PEG-3000 molecule. It must be recalled, however, that the work was performed at 22°C. At elevated temperatures, the resistance offered by the cell wall may diminish, allowing solutes of higher molecular weight to penetrate.

Stamm (1964) looked at the effect polyethylene glycol solutions of varying molecular weight had on the bulking of Englemann spruce cross sections of 2 by 2 by 1/8 inches. The specimens were oven dried to determine their weight and tangential and radial dimensions. They were then immersed in distilled water, evacuated to remove the air, and allowed to soak overnight to become completely saturated. The tangential and radial dimensions were again measured followed by soaking for 20 hours at 25°C in 30 percent by weight solutions of three methoxy polyethylene glycols, and three regular polyethylene glycols having average molecular weights ranging from 350 to 58,000 as shown in Table

Table 19. Relationship Between Molecular Weight of Polyethylene Glycol and the Nonsolvent-Water Content of Green Wood (Tarkow, Feist, and Southerland, 1966).

PEG Number	, Molecular Weight	Δ ¹ (gram of water/ gram of wood)	Standard Deviation (gm/gm)
200	200	0.09	0.01
1000	1000	0.23	0.02
2000	2500	0.27	0.02
4000 USP	3100	0.41	0.03
4000	4800	0.39	0.03
9000	10500	0.40	0.03
20000	18800	0.37	0.03

 $^{^{1}\}Delta$ = nonsolvent-water content.

20. The polymers with molecular weights up to and including 1,000 gave degrees of bulking (reductions in cross sectional shrinkage compared to controls) of at least 90 percent, as shown. The two high molecular weight polyethylene glycols gave considerably less bulking.

In order to account for the low degree of bulking obtained with the high molecular weight polymers, Stamm performed another series of experiments. Water-saturated specimens were immersed in the two high molecular weight methoxy polyethylene glycols and the three regular polyethylene glycols and soaking was carried out in sealed canning jars in an oven at 90°C for three days. Measurements were then taken each day over the next 12 days. The data is given in Table 21. Specimens containing the polymers with a molecular weight of 1000 or less gave similar bulking values to those in Table 20. The two higher molecular weight polymers gave only slightly less bulking than the lower molecular weight polymers. These results are quite different than those reported by Tarkow, Feist, and Southerland, as discussed earlier, but the two experimental procedures are radically different. Stamm attributed the increased bulking at the higher temperatures to the greatly increased solubility of the polymers, and the large decrease in their viscosity. The results may also be due to the micromotions in the polymer network at the elevated temperatures. In any event, effective void size would appear to exhibit a definite influence on the penetration of materials.

Table 20. Bulking of Cross Sections of Englemann Spruce at 25°C (Stamm, 1964).

Molecular Weight	PEG Take-up (%)	Cross Sectional Shrinkage (%)	Average Bulking (%)
Methoxy PEG 350 350	57 58	0.79 0.74	92
550 550	66 65	0.89 0.84	91
750 750	63 62	0.94 0.94	90
Regular PEG 1,000 1,000	68 67	0.94 0.94	90
30,000 30,000	51 50	5.60 5.60	41.3
58,000 58,000	35 34	6.98 7.03	26.6
Water Water	<u> </u>	9.48 9.58	_

Table 21. Bulking of Cross Sections of Englemann Spruce at 90°C (Stamm, 1964).

Chemical	 PEG Content (%)	Cross Sectional Shrinkage (%)	Bulking (%)
Methoxy PEG			
550	51	1.33	91
750	57	1.08	92
Regular PEG		* •••	
1,000	56	0.84	94
30,000	50	2.70	81
58,000	51	3.15	77
Water		13.93	-

III. EXPERIMENTAL DESIGN

A study was designed to relate resin wetting and resin molecular weight properties to the quality of adhesive bonds in a randomly oriented oak flakeboard. The commercial phenol-formaldehyde resin (PF) and the six ammoniated-lignosulfonate-phenol-formaldehyde resins (ALS-PF) used and discussed in the previous study were also used here.

A completely randomized experimental design was employed. Internal bond and modulus of rupture values obtained in the previous study using a resin application rate of 4 1/2 percent were used as bond evaluators. This particular resin application rate was chosen as it represented an average value between the lower and higher rates studied.

Contact angle measurements were used as indicators of resin wetting properties. Contact angle measurements were made after 5-second, 30-second, 1-minute, 5-minute, and 10-minute intervals. Ten replications were carried out for each combination of time and resin viscosity variables.

Measurement of Contact Angle

Contact angle measurements were carried out under controlled conditions of 73°F and 50 percent RH according to TAPPI Standard T458os-70.² The individual oak flakes used in the study were obtained from a random sampling of the oak furnish. Larger sized flakes were selected for the wetting measurements.

²TAPPI Standard T458os-70. A standard published by the Technical Association of the Pulp and Paper Industry. Revised 1970.

Contact angles were observed using a horizontally oriented Bausch and Lomb light microscope equipped with a camera attached for photographing the resin droplet. The flake specimens rested on a stage which allowed the sample to be oriented so that its surface was parallel to the axis of the microscope and the camera.

A droplet of resin was applied to the flakes using a microsyringe with needle sizes ranging from #22 to #26. This range in needle size allowed for a more uniform droplet size when working with the various viscosity resins. The resin droplet was photographed after 5 seconds, 30 seconds, 60 seconds, 300 seconds, and 600 seconds had elapsed.

Contact angle, width of the resin droplet, and maximum height of the resin droplet were measured from the photographs. Contact angles were measured to the nearest degree using a protractor with a movable hairline. The contact angle was measured on both sides of the resin droplet, and the average of these two readings was taken as the wetting contact angle for that replication.

Resin Molecular Weight Distribution

The molecular weight distributions for the resins used in this study were secured through the Weyerhaeuser Company. Resin molecular weights were calibrated using a standard polyethylene glycol solution. The molecular weight distributions are contained in Appendix C. The molecular weight values shown are not the molecular weights of the resins as such, but are the molecular weights of the standards having the same volume as the resin molecules.

IV. RESULTS AND DISCUSSION

Wettability versus Glue Bond Quality

Contact angle values ranged from 87° to 116° at the 5-second interval, and from 53° to 95° at the 600-second interval. Increasing resin viscosity generally resulted in increasing contact angle values (decreasing wetting properties) at any given time interval.

The relationship between resin wetting properties and bond quality (IB, MOR) was investigated statistically using regression analysis. Contact angle measurements taken at the ten-minute interval were averaged over the ten replications, and these average values were used in the analysis. Contact angle measurements taken at this time interval were felt to best approximate wetting properties attained during board fabrication as at least ten minutes elapsed between blending and pressing operations. This averaging procedure was also carried out for the resin droplet width and height measurements as well as for the ratio of droplet width to height.

Regression analysis indicated a significant relationship between internal bond and resin droplet width (.95 level), but there was no significant relationship between MOR properties and droplet width, or bond strength (IB, MOR) and contact angle, droplet height, or the droplet width to height ratio.

The relation between resin droplet width and IB strength, although significant, was not felt to be completely valid due to the fact that there was no means of controlling the amount of resin applied to each flake, and droplet width would be influenced by this fact.

In a further attempt to relate resin wetting properties to bond quality, regression equations were fit to the contact angle measurements relating contact angle to time. The equations were in the form Y=a+blnX where the dependent variable (Y) was contact angle in degrees and the independent variable (X) was time in seconds. The values for the slope of each line (b) were averaged over the ten replications as were values for the ratio of y-intercept to slope (a/b). An attempt was made to relate these values to bond quality, however, analysis indicated that neither relation was significant.

It would appear from the results that an adhesive must do more than just wet and spread on the wood surface for a strong bond to result. An adhesive system must perform five "life processes" for the formation of a strong adhesive joint. The adhesive must flow to a continuous film; transfer to the opposite unspread surface; penetrate the pores of the wood; wet (adsorb to) the wood substance; and solidify to a strong mass. After surface contact has been established the adhesive must continue functioning, penetrating the wood substance and bonding to the wood cell walls. Only after all five "life processes" have taken place to an optimum degree will a strong adhesive bond result.

Molecular Weight Distribution versus Glue Bond Quality

The molecular weight distribution curves for the resins used in this study were broken into areas of low, medium, and high molecular weight (MW). The break points were arbitrarily placed at log 10 relative molecular weights of 3.0 and 4.0 respectively (Figure 18). The left and middle peaks represent a fractionation of the resin molecules,

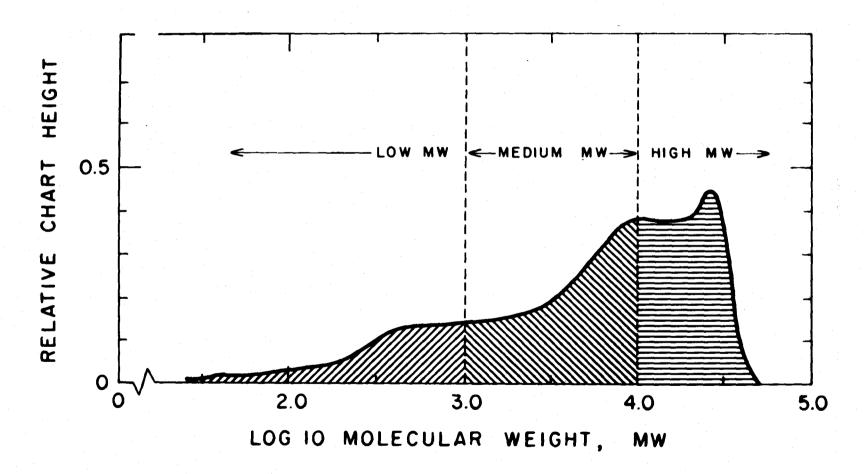


Figure 18. Molecular weight distribution break points.

while the right peak represents an exclusion point and is a collection of the larger resin molecules.

Because total MW area under the distribution curve tended to vary from resin to resin, the low, medium, and high MW areas under the curve were divided by the total MW area under the curve, the result being fractional MW area values. This procedure helped to normalize the data, minimizing differences in area from sample to sample.

Regression analyses relating internal bond strength (psi) to the fractional MW areas under the distribution curves indicated significant correlations between IB and low, medium, and high fractional areas.

These results are summarized in Table 22, and are illustrated in Figures 19, 20, and 21. There was no significant relation between IB and total MW area under the distribution curves.

The negative slopes for the regression equations relating IB quality to the low and medium fractional MW areas under the distribution curve indicate decreasing IB properties with increasing quantities of resin molecules in these areas. On the other hand, the positive slope for the relation between IB properties and the high fractional MW area under the curve indicates an increase in strength values with increasing quantities of resin molecules in this area.

Explained in terms of wood and adhesive variables, the negative slope indicates a tendency toward overpenetration of the resin molecules into the wood substrate, resulting in a starved glue line. The larger resin molecules do not penetrate the wood substrate as easily, increasing the amount of adhesive retained at the glue line and available for bonding. While resin penetration is a necessary part of bond formation,

Table 22. Regression Equations Relating IB Properties (psi) to Fractional Molecular Weight Areas Under the Distribution Curve.

	R	R ²	Significance	
40 lbs/cu.ft. nominal density				
$IB = 88.137 - 99.320 (L/T)^{1}$	-0.81	0.65	.01 <p<.05< td=""></p<.05<>	
IB = 96.360 - 75.010 (M/T)	-0.90	0.82	P<.01	
IB = 44.125 + 53.900 (H/T)	+0.86	0.74	.01 <p<.05< td=""></p<.05<>	
45 lbs/cu.ft. nominal density				
IB = 152.060 - 178.999 (L/T)	-0.75	0.57	NS ²	
IB = 171.000 - 145.260 (M/T)	-0.90	0.82	P<.01	
IB = 71.080 + 101.350 (H/T)	+0.83	0.70	.01 <p<.05< td=""></p<.05<>	

¹Low (L), medium (M), high (H), and total (T) molecular weight area under the distribution curve.

 $^{^2}$ NS = not significant.

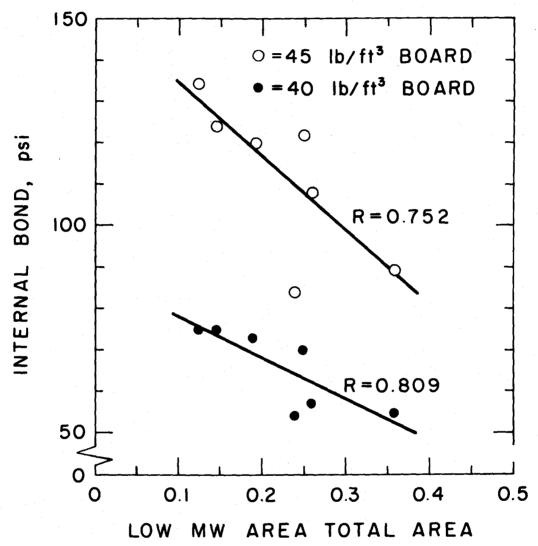


Figure 19. Internal bond strength versus LOW/TOTAL molecular weight area.

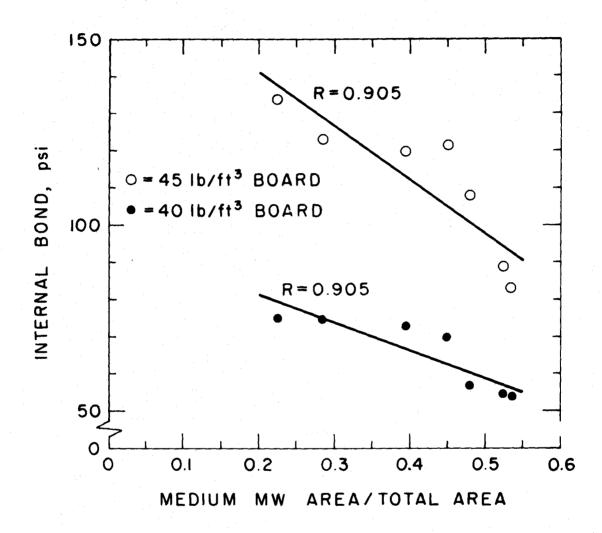


Figure 20. Internal bond strength versus MEDIUM/TOTAL molecular weight area.

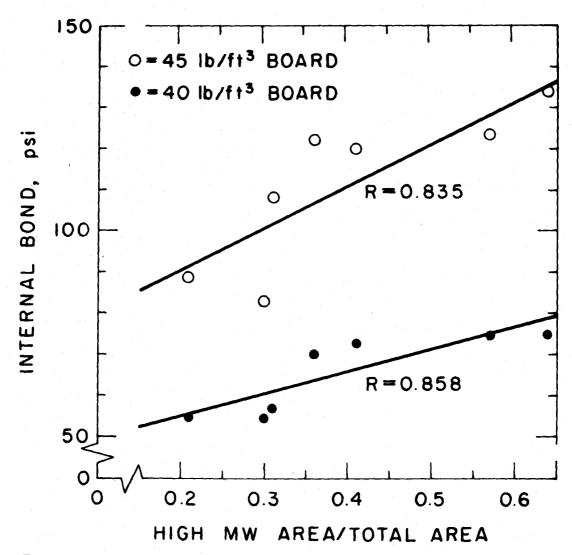


Figure 21. Internal bond strength versus HIGH/TOTAL molecular weight area.

too much penetration can result in reduced bonding.

The results appear consistent with those reported by Tarkow, Feist, and Southerland (1966) as discussed earlier. The authors found through their studies that the limiting size for the penetration of a water soluble material into green wood was that of a PEG-3000 molecule. This value corresponds with the medium fractional MW area under the distribution curve. Resin molecules having molecular weights below 3000, some in the medium MW area and all those in the low MW area, would be capable of penetrating the wood substrate with relatively little difficulty. Resin molecules with molecular weights of 3000 or greater, those in the medium and high fractional MW areas, would experience increasing difficulty in penetration.

It should be recalled here that the study by Tarkow et al. was performed using completely saturated wood specimens whereas this study dealt with relatively dry wood (an in press moisture content of 10 percent). The cell wall structure was not in a completely swollen state. However, phenol molecules are somewhat smaller than polyethylene glycol molecules, moisture content having a rather small effect on the size of the molecule capable of penetrating the cell wall. It should also be recalled that the PF and ALS-PF resins had a solids content of 47 and 40 percent respectively. This means that approximately 60 percent water was associated with each resin. This water might be capable of exhibiting a swelling influence in the immediate vicinity of the resin droplet, allowing for increased chances of resin penetration.

The results indicate that the maximum size of a resin molecule capable of penetrating wood cell walls might well correspond to a

molecular weight of 3000. This value falls in the area of medium molecular weight resin molecules, the area exhibiting the highest correlation with IB quality.

There were no significant relationships found between resin molecular weight distribution and MOR (psi) properties.

The relationship between resin pH and bond quality (IB, MOR) was also investigated using regression analysis. Results indicated a significant relationship (.95 level) between resin pH and IB values for the 40 lbs/cu.ft. nominal density boards, however, a scatter plot of the data indicated a clustering of the data points in two groups. This grouping of data points at the low and high pH values (10.6 and 12.2) resulted in the significant correlation. There were no significant correlations found between IB properties for the high density boards and pH, or between MOR properties for either the low or high density boards and pH. Also, there were no correlations found between resin viscosity and bond quality.

V. SUMMARY AND CONCLUSIONS

A study was conducted to relate resin wetting and resin molecular weight properties of a commercial phenol-formaldehyde resin and six ammoniated-lignosulfonate-phenol-formaldehyde resins with the bond quality of a randomly oriented oak flakeboard.

Resin wetting properties were not found to exhibit any significant correlation with bond quality. However, low, medium, and high fractional areas under the resin MW distribution curves were found to exhibit significant relationships with IB strength properties. There were no significant correlations found for MOR strength properties (psi). A significant relationship was observed between resin pH and IB properties for the low density boards, however, this was due to a clustering of the data points in the two pH areas.

Based on the results obtained in this study it may be concluded that:

- (1) The occurrence of resin wetting and spreading of the wood surface alone is not sufficient for the formation of a strong adhesive bond. Although good wetting is a necessary prerequisite to the formation of a strong bond, good wetting alone does not insure that a quality bond will result.
- (2) Within the range of resin molecular weight distributions employed in this study, fractional areas under the distribution curve of low, medium, and high molecular weight exhibit significant correlations with IB strength. Internal bond strength was found to increase with increasing quantities of higher molecular weight resin molecules. No significant relationships were found between MOR board properties (psi) and resin molecular weight distribution.

- (3) There were no significant relations found between resin pH and IB or MOR board properties.
- (4) There were no significant relations found between resin viscosity and IB or MOR board properties.

BIBLIOGRAPHY

- Bodig, J. 1962. Wettability related to gluability of five Philippine mahoganies. Forest Products Journal 12(6):265-270.
- Bryant, B. S. 1968. Interaction of wood surface and adhesive variables. Forest Products Journal 18(6):57-62.
- Freeman, H. G. 1959. Relation between physical and chemical properties of wood and adhesion. Forest Products Journal 9(12):451-458.
- Hse, C. Y. 1972. Wettability of southern pine veneer by phenol formaldehyde wood adhesives. Forest Products Journal 22(1):51-56.
- Stamm, A. J. 1964. Factors affecting the bulking and dimensional stabilization of wood with polyethylene glycol. Forest Products Journal 14(9):403-408.
- Tarkow, H., W. C. Feist, and C. F. Southerland. 1966. Interaction of wood with polymeric materials penetration versus molecular size. Forest Products Journal 16(10):61-65.
- Wellons, J. D. 1968. Adhesion to wood substrates. A.C.S. Symposium Series, No. 43. Pp. 150-168.

APPENDICES

APPENDIX A

Table A-1. Regression Equations Relating Internal Bond Properties to Board Density.

Resin Type	Resin Rate (%)	$Y^1 = a + b X^2$	R	R ²
PF-320 cps-10.4	3.0 4.5 6.0	IB = -196.07 + 6.3893 X $IB = -288.05 + 9.1244 X$ $IB = -276.05 + 9.3820 X$	+0.90 +0.94 +0.95	0.81 0.89 0.90
ALS-PF-84 cps-12.1	3.0 4.5 6.0	IB = -268.76 + 8.4225 X $IB = -228.63 + 7.7291 X$ $IB = -411.97 + 12.450 X$	+0.94 +0.95 +0.98	0.88 0.90 0.96
ALS-PF-100 cps-10.6	3.0 4.5 6.0	IB = -128.26 + 4.3752 X $IB = -185.72 + 6.1423 X$ $IB = -283.49 + 8.7604 X$	+0.95 +0.92 +0.95	0.90 0.85 0.90
ALS-PF-270 cps-12.3	3.0 4.5 6.0	IB = -268.07 + 8.3812 X $IB = -231.97 + 7.8664 X$ $IB = -184.87 + 6.9331 X$	+0.93 +0.94 +0.82	0.86 0.88 0.67
ALS-PF-300 cps-10.6	3.0 4.5 6.0	IB = -160.46 + 5.2500 X $IB = -167.67 + 5.6727 X$ $IB = -241.09 + 7.8626 X$	+0.89 +0.92 +0.94	0.79 0.84 0.88
ALS-PF-850 cps-12.2	3.0 4.5 6.0	IB = -230.60 + 7.5306 X $IB = -305.08 + 9.7259 X$ $IB = -301.13 + 9.8405 X$	+0.95 +0.93 +0.91	0.90 0.87 0.82
ALS-PF-540 cps-10.6	3.0 4.5 6.0	IB = -180.92 + 5.8584 X $IB = -227.22 + 7.3655 X$ $IB = -211.65 + 7.0313 X$	+0.97 +0.93 +0.92	0.94 0.86 0.85

 $l_{Y} = IB \text{ (psi)}$ 2X = Density (1bs/cu.ft.)

Table A-2. Regression Equations Relating Modulus of Rupture Properties to Board Density.

Resin Type	Resin Rate (%)	$Y^1 = a + b X^2$	R	R ²
PF-320 cps-10.4	3.0	MOR = $-5837 + 208.89 X$	+0.89	0.79
	4.5	MOR = $-7353 + 249.13 X$	+0.88	0.77
	6.0	MOR = $-6170 + 230.67 X$	+0.83	0.69
ALS-PF-84 cps-12.1	3.0	MOR = $-5503 + 201.69 \text{ X}$	+0.82	0.68
	4.5	MOR = $-4536 + 174.66 \text{ X}$	+0.82	0.67
	6.0	MOR = $-4765 + 190.83 \text{ X}$	+0.63	0.40
ALS-PF-100 cps-10.6	3.0	$MOR \approx -5357 + 183.57 X$	+0.76	0.58
	4.5	$MOR \approx -5391 + 191.45 X$	+0.91	0.83
	6.0	$MOR \approx -8030 + 258.91 X$	+0.89	0.80
ALS-PF-270 cps-12.3	3.0	MOR = -8862 + 275.11 X	+0.90	0.81
	4.5	MOR = -4953 + 191.32 X	+0.85	0.72
	6.0	MOR = -5508 + 206.68 X	+0.82	0.68
ALS-PF-300 cps-10.6	3.0	MOR = -2504 + 121.43 X	+0.72	0.51
	4.5	MOR = -9405 + 297.80 X	+0.86	0.73
	6.0	MOR = -7628 + 256.04 X	+0.95	0.90
ALS-PF-850 cps-12.2	3.0	MOR = -4557 + 171.46 X	+0.76	0.59
	4.5	MOR = -5553 + 201.97 X	+0.79	0.63
	6.0	MOR = -6784 + 231.06 X	+0.87	0.75
ALS-PF-540 cps-10.6	3.0	MOR = -7252 + 234.30 X	+0.85	0.73
	4.5	MOR = -7036 + 235.15 X	+0.85	0.73
	6.0	MOR = -6789 + 230.61 X	+0.85	0.72

 $^{^{1}}Y = MOR \text{ (psi)}$ $^{2}X = Density \text{ (1bs/cu.ft.)}$

Table A-3. Regression Equations Relating Modulus of Elasticity Properties to Board Density.

Resin Type	Resin Rate (%)	$y^1 = a + b X^2$	R	R ²
PF-320 cps-10.4	3.0	MOE = -412540 + 20338 X	+0.77	0.60
	4.5	MOE = -736480 + 28927 X	+0.86	0.74
	6.0	MOE = -691330 + 28141 X	+0.86	0.74
ALS-PF-84 cps-12.1	3.0	MOE = -566270 + 23687 X	+0.84	0.70
	4.5	MOE = -438670 + 19929 X	+0.81	0.66
	6.0	MOE = -289260 + 17641 X	+0.73	0.53
ALS-PF-100 cps-10.6	3.0	MOE = -713420 + 26130 X	+0.82	0.68
	4.5	MOE = -687420 + 25903 X	+0.81	0.66
	6.0	MOE = -867390 + 31214 X	+0.88	0.77
ALS-PF-270 cps-12.3	3.0	MOE = -851420 + 29564 X	+0.84	0.71
	4.5	MOE = -435140 + 20569 X	+0.76	0.58
	6.0	MOE = -495630 + 22046 X	+0.69	0.47
ALS-PF-300 cps-10.6	3.0	MOE = -274730 + 16575 X	+0.68	0.46
	4.5	MOE = -788500 + 29838 X	+0.85	0.72
	6.0	MOE = -660320 + 26886 X	+0.89	0.80
ALS-PF-850 cps-12.2	3.0	MOE = -464480 + 20156 X	+0.70	0.49
	4.5	MOE = -378170 + 18494 X	+0.72	0.52
	6.0	MOE = -698530 + 26101 X	+0.87	0.75
ALS-PF-540 cps-10.6	3.0 4.5 6.0	MOE = -775450 + 28506 X $MOE = -805490 + 29711 X$ $MOE = -770300 + 29157 X$	+0.86 +0.83 +0.91	0.74 0.69 0.82

 $^{^{1}}Y = MOE (psi)$ $^{2}X = Density (1bs/cu.ft.)$

APPENDIX B

Table B-1. Analysis of Variance for IB.

Source	SS	DF	MS	F-Ratio	Significance
Resin viscosity	18818.28	6	3136.38	68.24	P<.01
Resin rate	16965.88	2	8482.94	184.57	P<.01
Density	71809.60	1	71809.60	156.24	P<.01
Viscosity × Rate	1781.16	12	148.43	3.23	P<.01
Viscosity × Density	1217.16	6	202.86	4.41	P<.01
Rate × Density	949.94	2	474.97	10.33	P<.01
Viscosity × Rate × Density	2021.76	12	168.48	3.67	P<.01
Error	3860.64	84	45.96	: <u>-</u>	

Table B-2. Analysis of Variance for MOR.

Source	SS	DF	MS	F-Ratio	Significance
Resin viscosity	3789954	6	631659	10.43	P<.01
Resin rate	4281040	2	2140520	35.34	P<.01
Density	35519400	1	35519400	586.42	P<.01
Viscosity × Rate	1410648	12	117554	1.94	.01 <p<.05< td=""></p<.05<>
Viscosity × Density	349453	6	58242	0.96	NS
Rate × Density	47164	2	23582	0.39	NS
Viscosity × Rate × Density	1767108	12	147259	2.43	P<.01
Error	5087897	84	60570	· ,	—

Table B-3. Analysis of Variance for MOE.

Source	SS	DF	MS	F-Ratio	Significance
Resin viscosity	81406800000	6	13567800000	15.03	P<.01
Resin rate	34735200000	2	17367600000	19.25	P<.01
Density	447484000000	1	447484000000	495.87	P<.01
Viscosity × Rate	20120040000	12	1676670000	1.86	NS
Viscosity × Density	10409820000	6	1734970000	1.92	NS
Rate × Density	436374000	2	218187000	0.24	NS
Viscosity × Rate × Density	21928560000	12	1827380000	2 . 02	.01 <p<.05< td=""></p<.05<>
Error	75803280000	84	902420000	-	<u> </u>

Table B-4. Analysis of Variance for LE.

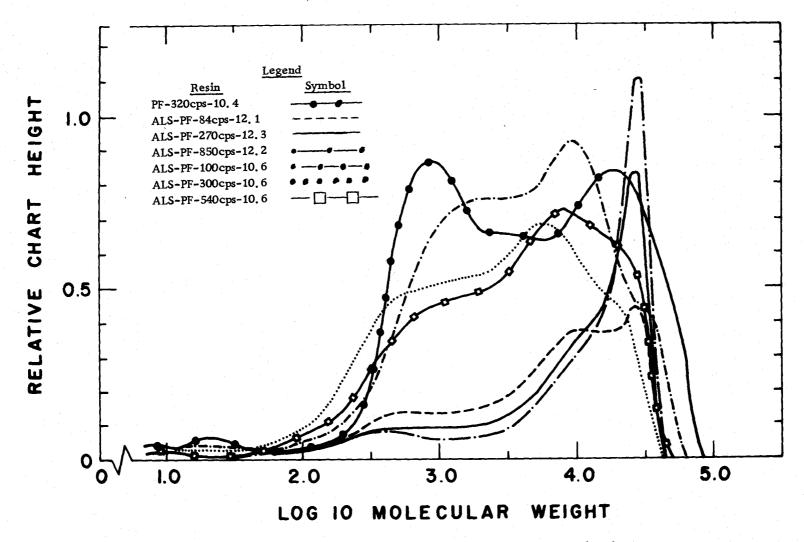
Source	SS	DF	MS	F-Ratio Significance	
Resin viscosity	. 08643	6	.01441	39.80	P<.01
Resin rate	。003916	2	.001958	5.41	P<.01
Density	.0008127	1	.0008127	2.24	NS
Viscosity × Rate	.006229	12	。0005190	1.43	NS
Viscosity × Density	.002954	6	.0004923	1.36	NS
Rate × Density	.001392	2	.006960	1.92	NS
Viscosity × Rate × Density	.004841	12	.0004034	1.11	NS
Error	. 03040	84	。0003619	-	

Table B-5. Analysis of Variance for TS.

Source	SS	DF	MS	F-Ratio	Significance
Resin viscosity	239.80	6	39.97	46.66	P<.01
Resin rate	148.25	2	74.13	86.54	P<.01
Density	35.04	1	35.04	40.92	P<.01
Viscosity × Rate	91.81	12	7.65	8,93	P<.01
Viscosity × Density	20.26	6	3.34	3.90	P<.01
Rate × Density	6.86	2	3.43	4.01	.01 <p<.05< td=""></p<.05<>
Viscosity × Rate × Density	34.10	12	2.84	3.31	P<.01
Error	71.95	84	0.86		· _

Table B-6. Analysis of Variance for WG.

Source	SS	DF:	MS: An area	F-Ratio	Significance
Resin viscosity	86.62	6	14.44	275.10	P<.01
Resin rate	4.40	2	2.20	39.19	P<.01
Density	0.05	1	0.05	0.88	NS
Viscosity × Rate	6.70	12	0.56	9.94	P<.01
Viscosity × Density	4.42	6	0.74	13.13	P<.01
Rate × Density	1.83	2	0.91	16.28	P<.01
Viscosity × Rate × Density	4.99	12	0.42	7 . 40	P<.01
Error	4.72	84	0.06	_	. -



APPENDIX C. Resin Molecular Weight Distributions