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| Title: | BONDING WOOD PART | IC LE COMPOSITES WITI | H FOAMED |
| | ADHESIVES | | |
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The quality and cost of wood particle composites are substantially determined by how the adhesive that bonds the composite is distributed on the particles. Distribution of the adhesive is controlled by the method of application. Currently the adhesive is applied by spraying it onto the particles. It was hypothesized that applying the adhesive as a foam would lead to improved adhesive distribution.

An experimental system was developed for foaming a urea formaldehyde adhesive with liquid freon-12 and for blending the foam with wood particles. This system was compared to systems for applying the adhesive as a spray and bulk liquid.

Strength properties of wood particle composites made with foamed adhesive were found to be equivalent to those with adhesive applied as a spray. Applying foamed adhesive was superior to mixing particles with unfoamed adhesive.

Strength properties were not correlated to adhesive distribution according to particle size. Foam and bulk liquid application produced adhesive distributions proportional to particle surface area. The spray application resulted in adhesive distributed disproportionately heavy on the larger particles relative to their surface area. Other factors, such as the number of particles not receiving adhesive and the uniformity of adhesive coverage on a particle surface, may explain the differences that occurred in strength properties.

Bonding Wood Particle Composites with Foamed Adhesive by

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BONDING WOOD PARTICLE COMPOSITES WITH FOAMED ADHESIVE

I. ADHESIVE DISTRIBUTION IN WOOD PARTICLE COMPOSITES

Wood particle composites such as particleboard, fiberboard, and hardboard are usually made from residues generated in the processing of logs into lumber and plywood. These wood residues are reduced by milling machines such as hammermills, flakers or disc refiners to particles of specified sizes, called furnish. An adhesive polymer is then applied to the particle surfaces. This mixture of particles and adhesive is formed into a mat containing the desired weight of material per square foot of surface area. Finally, the mat is consolidated under heat and pressure to polymerize the adhesive and form a board. Of all the steps in this process, the blending of adhesive with the wood particles has the greatest effect on both the cost and the quality of the composite.

Economics, to a large part, dictate the amount of adhesive that can be applied to the particles. Urea formaldehyde synthetic polymers are most widely used as the adhesive for particleboard. At present these adhesives cost about 13 cents per pound of dry solids. Wood residues cost between one-half and one cent per pound. Particleboard adhesives are generally applied at a rate of five to seven percent solids based on dry wood weight. Thus, costs for the adhesive account for 50-60 percent of the raw material cost. Recently, scarcity and price

increases have raised even higher the binder's proportion of the product costs.

Therefore, technological efforts have been directed at maximizing board properties at a given level of adhesive application.

Physically, applying five to seven weight percent of adhesive solids means a small volume of adhesive is applied to a large volume of particles - approximately one part adhesive to 140 parts wood. The problem is to find a way to adequately distribute this small amount of adhesive on a large particle surface area.

A number of commercial blenders are available to particle-board manufacturers - Bison, Keystone, Grinco. In these blenders, the wood particles are blown or thrown past sprayers which deposit a mist of adhesive on the particles. The dwell time of a particle in most commercial blenders is no more than one to two minutes, and often as little as a few seconds. Thus some particles may be missed, and others may receive more than their share of adhesive. In some mills particles leave the blender through a screw conveyor which rubs one particle against another. The intent of this "post blending" is to redistribute the adhesive. A recently introduced particleboard blender incorporates this principle of frictional transfer into the main blending step. The Littleford blender screw feeds particles through the blending chamber, rubbing them together as adhesive is extruded from rotating arms. At present, there is only partial understanding of what

really occurs when adhesive is blended with wood particles in any of these blenders.

Adhesive Distribution and Composite Quality

Adhesive distribution includes several concepts: whether a particle receives adhesive or not, and what proportion of the particles receive no adhesive; when all particles receive some adhesive, whether the adhesive is distributed proportional to surface area of the particles or not; on a particular particle, whether the adhesive is distributed as a film or as individual droplets and whether the surface is uniformly covered. All of these factors interact to determine the quality of a wood particle composite.

The importance of adhesive distribution can be thought of in terms of the potential bonding surface area between particles. If particles receiving little or no adhesive during blending are mixed with particles receiving adhesive, there is a potential for contact between two particle surfaces with no adhesive. This represents a weak zone in the board. The effect of such non-uniform adhesive distribution is to lower the strength of the board because overall strength is controlled largely by the areas of lowest strength.

Commercially produced particleboards have been shown to have inadequate adhesive distribution (Meinecke, 1960, Carroll and McVey, 1962). Carroll and McVey (1962) compared adhesive distribution in a

board from a commercial blender to that of laboratory boards made with varying amounts of particles receiving no adhesive. The commercial board was equivalent to laboratory boards made with 20-30 percent of the particles having no adhesive deposited on their surfaces. Studies of blender loading consistently suggest that a significant proportion of the wood particles receive little or no adhesive.

Christensen and Robitschek (1974) found that post-blending of a commercial particleboard furnish improved board properties. They attributed improvements to interparticle transfer of adhesive solids, which probably means that the proportion of particles receiving adhesive increased.

Interparticle Adhesive Distribution

Several other investigators have studied how adhesive is distributed in particleboard (Burrows, 1961, Carroll and McVey, 1962, Kamrath, 1963, Lehman, 1965, 1968, 1970, Maloney, 1970, Meinecke, 1960, Schwarz et al., 1968, Duncan, 1974). A wood particle furnish consists of particles of various sizes (see Table 1). Adhesive could be distributed proportionally to the surface area of each particle fraction or unbalanced in favor of large or small particles. If bonding is to be uniform throughout the board, the first alternative should be the optimum. The optimum adhesive distribution has not been clearly defined.

| Table 1. | Average Adhesive Solids Distribution on Furnish from Te | n |
|----------|---|---|
| | West Coast Particle Board Plants. | |

| Mesh No. 1 | % Screen Weight ² with Adhesive | % Total Adhesive Solids | Approximate % 3 Surface Area |
|------------|--|-------------------------|------------------------------|
| +10 | 20-25 | 5-10 | 6 |
| -10 +20 | 30-35 | 15-20 | 25 |
| -20 +32 | 20-25 | 15-20 | 20 |
| -32 +42 | 5-10 | 10-15 | 13 |
| -42 | 10-15 | 35-40 | 37 |
| | 100 | 100 | 100 |

⁺ means material retained on a screen

Data by Schwartz et al. (1968) on adhesive solids distribution from ten West Coast particleboard mills are shown in Table 1.

Particles larger than 32 mesh size accounted for 70-85 percent of the total furnish weight but received only 40-50 percent of the total adhesive solids applied. In contrast, fines (smaller than 32 mesh) comprised 15-25 percent of the furnish weight and received 45-55 percent of the total adhesive solids. The fines had an adhesive solids content of 20 percent in a board with a nominal adhesive content of eight percent. Results of this type have led to the conclusion that fines receive more than their share of adhesive. This effect is ascribed to the high surface area per gram of the smaller particles.

⁻ means material that passes through a screen

From data by Schwarz, et al. (1968)

³Calculated based on data by Maloney (1970)

Because many researchers have believed that fines receive excessive adhesive, much research has attempted to develop systems for putting more of the adhesive on the larger particles.

Duncan (1974) has developed a "Normal Resin Distribution" theory which predicts based on probabilities that adhesive distribution will parallel particle surface area distribution. He did not directly measure particle sizes and calculate surface areas. Duncan's calculations were based on the questionable assumption that the volume of a particle is proportional to the first power of the mesh opening on which the particle was retained. He demonstrated with that assumption that adhesive distribution is highly correlated to the relative numbers of particles (RNP) of each size and their relative surface areas (RA). Adhesive solids distribution was found to be poorly correlated to fraction weights as determined by screen analysis. Duncan's results showed, in contrast to Schwarz, that coarse particles tend to receive more resin than area contributed and that a higher adhesive content on fines is not sufficient to offset their higher surface area. Cox (1974) used Duncan's assumptions and found similar results on furnish sprayed in a commercial blender (See Table 2). Using data by Maloney (1970) on the surface area per gram of the fractions in Table 1, I calculated by Duncan's method the approximate surface area of Schwarz's furnish. It can be seen that relative surface area more or less parallels adhesive solids distribution, and that fines did not

really receive more than their share of the adhesive, but the errors resulting from Duncan's assumptions are unknown. The practical significance of Duncan's research is that spread rates must be adjusted whenever particle surface area is changed, if board properties are to be maintained.

Table 2. Adhesive Distribution by Duncan's Method.

| Mes | h No. | % Screen Weight with Adhesive | % Total Adhesive Solids | % R.A. ² | % R.N.P. |
|-----|-------|-------------------------------|-------------------------|---------------------|----------|
| | +8 | 13.33 | 8.49 | 4.77 | 4.75 |
| -8 | +10 | 20.14 | 13.05 | 10.22 | 10.16 |
| -10 | +14 | 17.41 | 13.75 | 11.92 | 11.41 |
| -14 | +20 | 8.44 | 9.45 | 9.75 | 9.66 |
| -20 | +28 | 16.32 | 19.14 | 19.99 | 19.83 |
| -28 | +40 | 8.84 | 14.63 | 15.27 | 15.58 |
| -40 | +60 | 7.71 | 19.57 | 23.75 | 24.06 |
| -60 | | 7.81 | 1.92 | 4.33 | 4.73 |
| | | 100.00 | 100.00 | 100.00 | 100.00 |

Data from Cox (1974)

Adhesive distribution exactly proportional to surface area may be difficult to attain with a spray system because the surface area of the adhesive spray is substantially less than that of the particles. For

Relative surface area calculated based on Duncan's assumptions (1974)

Relative number of particles calculated based on Duncan's assumptions (1974)

example, an adhesive spray with an average droplet diameter of 40 microns applied at seven percent solids to one gram of particleboard furnish has a surface area of approximately 82 cm^2 . The gram of particleboard furnish has a surface area of approximately 260 cm^2 .

Intraparticle Adhesive Distribution

Several investigators have suggested that optimum intraparticle adhesive distribution involves spreading the adhesive to form continuous interparticle films of adhesive rather than "spot welds" between the particles (Klauditz and Ulbricht, 1958, Kollman, 1957, Lehman, 1968, 1970, Marion 1958, Meinecke, 1960). Marion (1958) states that "the bonding strength... increases with chip surface area and diminishing spread per unit of surface. This is sufficiently explained by the fact that more intimate contacts are formed between thinner chips with thinner gluelines as a consequence." Thinner wafers mean greater subdivision and increased surface area. Less adhesive (i.e. continuous films) is spread per unit of wafer surface area to take advantage of the greater contact area for bonding.

Suchsland (1959) demonstrated that continuous films of adhesive solids spread as low as 3.2 grams per square meter produce joints stronger than wood for woods of specific gravity less than 0.5.

Higher spread rates did not increase bonding strength because the strength of the wood was the limiting factor. A spread rate of 3.2

grams per square meter would produce a uniform adhesive film of only 3.9 microns or about 0.00015 inches thick. Klauditz and Ulbricht (1958) calculated an approximate surface area of 1.43 square meters for 100 grams of spruce wafers of 0.04 centimeter average thickness. If six percent adhesive solids were applied uniformily, this resulted in a spread rate of 4.2 grams per square meter or a glueline thickness of 4.9 microns (0.00020 inch). This is a greater spread rate than Suchsland found necessary to produce adhesive bonds stronger than the wood.

Continuous gluelines are not formed directly by spraying the adhesive as droplets. Klauditz (1957) and Marra (1960) suggested that adhesive be applied as small droplets to uniformly cover the chip surface so that the entire contact area is acted upon. The adhesive droplets flow under heat and pressure to form continuous interparticle gluelines. Studies have demonstrated that a fine degree of dispersion is necessary to form continuous interparticle gluelines. Meincke (1960) found that when droplets of 8-35 microns in diameter are distributed uniformly, continuous gluelines are formed and result in maximum board properties at a given density. However, commercial boards had strength properties only 60 percent of those of laboratory boards formed under these optimum conditions of resin dispersal.

Bonding occurs as "spot welds" of discrete droplets of adhesive in commercial particleboards. Lehman (1968, 1970) utilized a coarse

spray (diam. 50-60 microns) to produce boards of this type. Their properties were inferior to boards made with a fine spray (diam. 30-40 microns) at a constant adhesive level and board density. This difference was attributed to greater surface coverage due to the formation of continuous films of adhesive between particles with the fine spray.

Carroll and McVey (1962) have produced evidence contrary to the thin film theory. They made boards with a high flow phenolic adhesive and noted reduced internal bond strength when compared to "spot weld" type boards. They postulated that the adhesive penetrated into the wood leaving the glueline between particles starved. Bryant (1968) confirmed these results but did not find a similar effect with urea adhesives.

Optimum droplet sizes of sprayed adhesives encountered under carefully controlled laboratory conditions are not necessarily obtainable in commercial production. Applying a fine spray is not practical because additional blending time is needed to achieve an optimum distribution. A coarse spray ranging from 7-80 microns is used to speed the blending operation (Maloney, 1970). A continuous process leads to high flow rates which results in poor atomization. In addition, sufficient maintenance of spray equipment is not possible in most production situations.

Dilution of the adhesive with water is a method which can be utilized to achieve a fine atomization and an optimum distribution (Carroll and McVey, 1962). In practice, any advantages are offset by increased drying costs to avoid increases in mat moisture content. Also, increased throughput of liquid due to its increased volume overloads the spray system and results in poor atomization. Dilution may also have adverse effects due to penetration of adhesive into the wood.

Trends have been toward using rapid cure adhesives in order to diminish press times. Precure of these "fast" adhesives is another potential cause of lost bonding area. Applying the adhesive as droplets requires pressure to increase bonding surface area. Loss of potential bonding surface area occurs if the adhesive cures before maximum bonding contact area is established (Carroll, 1963). This phenomena probably occurs more readily in surface layers which are exposed to higher temperatures early in the press cycle. Thus, it seems that the ideal is to disperse the adhesive as fine droplets to create continuous films, provided dryout and overpenetration can be avoided. But commercial processes are unable to achieve this optimum with sprayed adhesives.

A final aspect of inefficient intraparticle distribution relates to density. Adhesive in areas where interparticle contact does not occur is wasted. This was noted by Maloney (1970) in a study of adhesive

distribution in 3-layer boards. When a coarse fraction was placed in the core a low density core was formed and low internal bonds resulted.

In summary, adhesive distribution affects both the cost and quality of composites made from wood residues. Inadequacies in adhesive distribution with current technology exist in (1) the failure to apply the adhesive to a portion of the particles, (2) the failure to distribute the adhesive according to the surface area contributed by each particle fraction, and (3) the inability to distribute the adhesive over the entire contact surface of the particle. These inadequacies are compensated for commercially by raising adhesive level and/or pressing to a higher density. Both of these measures are contrary to efficient utilization of the adhesive binder.

Improvements in Adhesive Distribution

Improvements in adhesive distribution could have two effects (Lehman, 1965):

- 1. Improve board quality at present adhesive levels;
- 2. Maintain present levels of quality and lower adhesive consumption, thus lowering costs of manufacture.

Carroll and McVey (1962) have shown that board quality can be maintained by lowering adhesive content if there is adequate distribution.

They estimate that adhesive consumption could be reduced by 20 percent on a commercial level. Lehman (1968, 1970) found that at a constant density and resin level, board properties improve with better adhesive distribution.

One approach to better distribute adhesive is to develop a new method for applying the adhesive. Applying the adhesive as a foam is an alternative method for blending. Zeigler (1958) and Bornstein (1958) mention potential advantages of foam application but no specific research results were reported. Adhesive applied as a foam has the advantage of a large surface area, thus fewer particles should be missed; the adhesive should be applied proportionally to the area contributed by each particle fraction; and continuous films should result on the particle surfaces. These potential advantages of a foam blending system are discussed in detail in Chapter II.

Study Objectives

This study intends to determine the effect of applying a urea formaldehyde adhesive as a foam on the adhesive distribution and strength properties of wood particle composites. This evaluation will be made by conducting a series of experiments with objectives as listed below:

 to develop a method for foaming a urea formaldehyde adhesive to 50-100 times its bulk liquid volume,

- to develop an apparatus for blending the foamed adhesive with wood particles,
- 3. to form wood particleboards with the foamed adhesive and evaluate strength properties of these boards using seven percent adhesive. (Strength tests are utilized as a measure of board quality) and,
- 4. to describe adhesive solids distribution by determining nitrogen contents on particle fractions segregated according to size.

II. DEVELOPING A FOAM BINDER

A foam is a suspension of a gas in a liquid. The gas phase is subdivided by spherical films of the liquid. By distributing the liquid as thousands of "bubbles," its surface area and volume may be extended greatly. Because of this increase in surface area, foams are never in a state of thermodynamic equilibrium. Formation of a foam increases the liquid's surface free energy, which must be at a minimum for the system to be stable. Surface free energy is reduced by destroying surface area, so reduction in surface area, i. e. breakdown of the foam, continues until the liquid phase returns to its original volume.

Blending Foam with Particles

The principle advantages of foamed adhesive application for wood composites were stated in Chapter I to be (1) a large surface area, thus fewer particles receive no adhesive; (2) application of adhesive proportional to surface area contributed by each fraction, and (3) deposition of continuous adhesive films on particle surfaces.

Effects of Surface Area and Volume

As stated previously, the formation of a foam results in a large increase in surface area compared with the bulk liquid. If the foam

surface area is greater than that of the particle surface area and if this surface can be transferred effectively, few particles should be missed and adhesive should be distributed "normally" or proportionally to the surface area of each particle fraction.

The surface area potential of a foam is presented in Figure 1, a graph of the surface area per gram of a foam as a function of bubble diameter. Surface areas per gram are shown at four levels of volume expansion. The curves demonstrate that (1) greater subdivision (smaller diameter spheres) results in higher surface areas per gram, and that (2) surface area per gram increases proportionally to volume expansion. The range of bubble diameters used in Figure 1 was based on actual measurements on a foamed urea formaldehyde adhesive. The curves in Figure 1 are useful in determining if a foam surface area is potentially greater than particle surface areas.

Figures 2 and 3 relate the surface areas per gram of wood particles and fibers to their sizes. Surface areas were derived from models for particle and fiber shapes. A rectangular solid with length: width:thickness ratios of 12:2,75:1 was used to calculate approximate wood particle surface areas. The range of particle sizes represented is approximately that of a wood particleboard furnish. Similarly, a cylinder with a length to diameter ratio of 100 to 1 was used for calculating wood fiber surface areas.

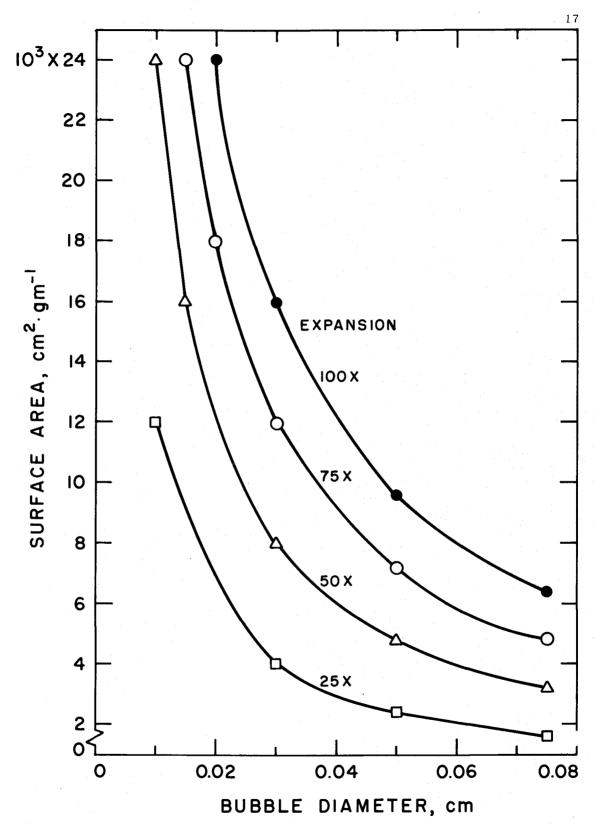


Figure 1. Surface area per gram of a foam as a function of bubble diameter.

Comparison of Figure 1 to Figure 2 shows that a foam expanded 50-100 times and applied at seven percent solids will have a greater surface area per gram than the surface area per gram of wood particles in a particleboard furnish. For example, 0.07 grams of foam expanded 75 times with a bubble diameter of 0.04 cm has a surface area of 630 cm². The smallest particles in a particleboard furnish have surface areas per gram less than 630 cm². They represent only 10-15 percent of the total surface area in a gram of furnish. On the average, a gram of particleboard furnish has less than half the surface area of these fine particles.

In contrast, wood more finely divided as fibers has a greater surface area per gram than that of a foam applied at seven percent solids. A wood fiber of four millimeters in length has a surface area per gram of approximately 1800 cm². This is nearly three times that of the surface area of the foam used in the previous comparison (630 cm²). Therefore, it appears that the potential advantage of the increased surface area of a foam is limited to particle composites.

The potential gain in surface area of a foam compared to a spray system is substantial (see Table 3). An adhesive foam (75X) applied at seven percent solids has a surface area more than twice that of a gram of a typical wood particle furnish. The adhesive spray surface area is far less than the particle surface area.

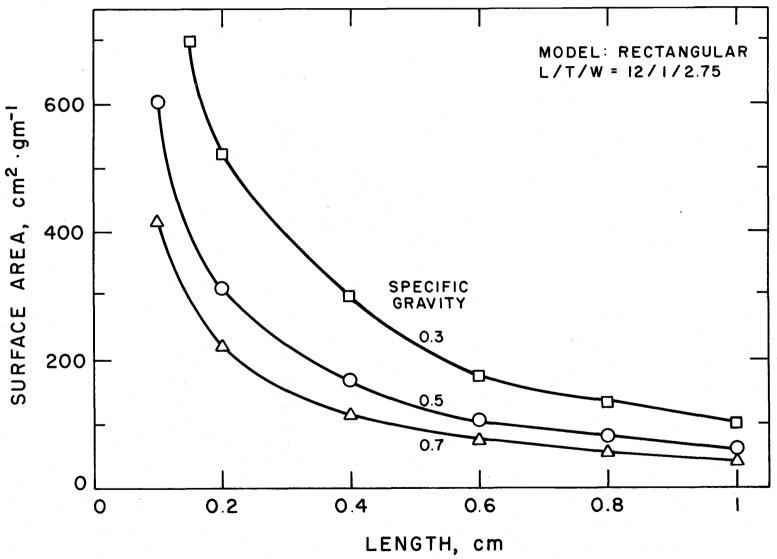


Figure 2. Surface area per gram of a rectangular wood particle as a function of length.

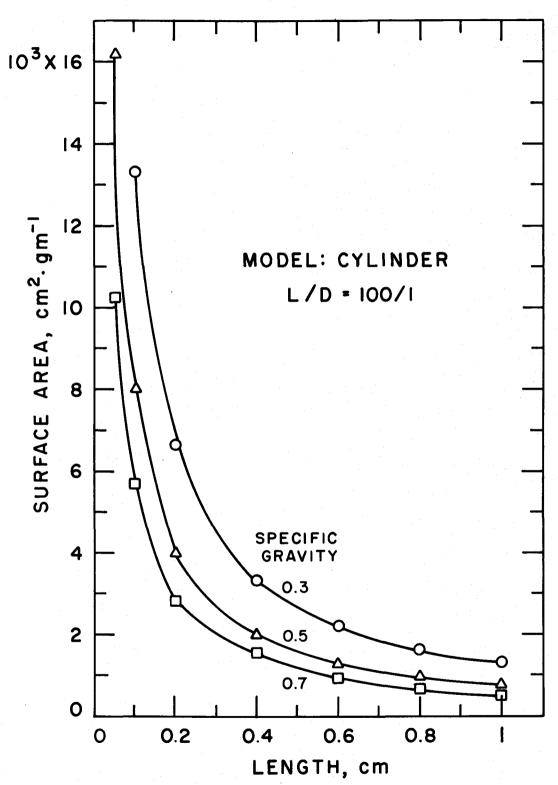


Figure 3. Surface area per gram of a cylindrical wood fiber as a function of length.

Table 3. Comparison of Adhesive Foam and Spray Surface Areas and Volumes.

| | | S.A. (cm ²) | Vol. (cm ³) |
|----------|-----------------------------|-------------------------|-------------------------|
| l gm. | Particleboard furnish | 256 | 5.5 |
| 0.07 gm. | Adhesive spray (40 microns) | 82 | 0.06 |
| 0.07 gm. | Adhesive foam (75X) | 630 | 6.3 |

Calculation based on screen analysis and surface areas given by Maloney (1970) for a typical particleboard furnish.

The volume of the adhesive is also increased by foaming. If the volume of foam and particles are similar, blending can be simplified to mixing of two components of equal proportions. Thus, few particles should be missed. Table 3 illustrates that a foam volume equal to or greater than the particle volume is feasible.

Continuous Adhesive Films

Several investigators (Klauditz and Ulbricht, 1958, Lehman, 1968, 1970, Meinecke, 1960) have found that adhesive distributed as a continuous film on particle surfaces resulted in composites with superior strength to those bonded with adhesive droplets. This effect is probably due to a greater bonding surface area.

It is hypothesized that a continuous adhesive film will be deposited when a foam is blended with wood particles. The internal structure of a foam is comprised of spherical films of adhesive. When spread on a surface the foam structure is destroyed and is deposited as a continuous film.

In contrast to spray droplets, heat and pressure are not necessary to extend bonding surface area and produce continuous interparticle gluelines. A greater surface area is immediately available for bonding, eliminating any effects of precure before maximum interparticle contact is established.

Several new processes are being developed for forming particle-board mats with oriented fibers or particles. These processes require a low tack in the mat. Adhesive sprayed as droplets produces sufficient tack to make these new forming processes difficult because the adhesive is highly concentrated at one point or points on the particle surface, and retains its moisture layer. Foamed adhesive might result in a blended furnish of low tack because the adhesive is spread as a film allowing water to be adsorbed more readily by the wood particles. However, if any of the foam structure is retained after blending, the foam will help prevent dryout, and result in tack equivalent to sprayed adhesive (Bornstein, 1958).

Low tack might be caused by adhesive penetration into the wood structure. Objection to adhesive distribution as a thin film is based on the belief that "starved" interparticle gluelines are formed.

Applying adhesive foams as designed in this study will help resolve this issue.

Past work on foamed adhesives for wood has emphasized benefits other than those discussed above. The idea of foaming adhesives for bonding wood materials was conceived before World War II.

Interest has centered on their use as a binder system for plywood (Bornstein, 1958, Cone, 1969, Freeman and Sorsa, 1961, Sevando, 1968, Ziegler, 1958, Zurowski, 1959).

Particle composite research related to foams has dealt with producing low density materials (< 35 pounds per cubic foot). As the density of a composite is lowered there is less interparticle contact and an increasing amount of void space. Therefore, strength properties drop substantially. Attempts have been made to incorporate foam structure into void areas to improve strength properties. Several patents have been issued for particleboard processes utilizing plastic foams (i. e. polystyrene, polyurethane) as binders (Deppe et al., 1966, Friebel and Kubel, 1971, Gaiten, 1967, Himmelheber, 1962, 1963, 1965).

The quantities of adhesive necessary to produce desired properties have made these prohibitively expensive. Deppe (1969) demonstrated the feasibility of producing low density wood particleboard with a foamed polyurethane, but concluded that adhesive cost makes the process uneconomical.

Developing a Foam Binder System

Information on a foamed binder using standard condensation adhesives is lacking. As previously stated, Ziegler (1958) and Bornstein (1958) mention potential advantages but no specific research results are reported. Bushbek (1964, 1968, 1969) has done a series of studies with a foamed urea adhesive applied by spraying.

To evaluate foam application a system for foaming the adhesive and blending it with particles must be developed and tested. Particle composites made with a foam system then can be compared to those made by standard spray systems. But to make this comparison, it is important to manipulate only the blending step of the process, so that changes in board properties are affected only by adhesive distribution. This fact is the basis for developing criteria for a system of generating and applying a foam.

Criteria for a Foam System

The desired characteristics of the foaming system are listed below.

- (1) The ability to control the polymerization properties of the adhesive should not be altered by the foaming method.
- (2) A volume expansion of 50 to 100 times is desirable, based on the surface area and volume relationships presented earlier.

- (3) The volume expansion must be reproducible.
- (4) The foam must be stable for a sufficient period of time to allow its application to wood particles.
- (5) The method must be compatible with particle blending after, or as, the foam is produced (See discussion in Chapter III).

To meet these criteria variables affecting foam volume, reproducibility, and stability must be controlled. The quantity of gas generated must be controlled if foam volume is to be reproducible.

Foams of high expansions are produced only when gas generation is spontaneous and rapid so that the liquid is supersaturated with gas.

This excess gas pressure is relieved by the formation of spherical films of liquid. Foam stability is affected by uniformity of bubble sizes. Pressure differences between bubbles of varying sizes results in coalescence and decrease of foam volume with time. Uniform bubble sizes are produced by rapid, spontaneous nucleation of gas bubbles. The surface tension and viscosity of the foaming mixture affects the ease of bubble formation, the thickness of interfacial films, and their resistance to thinning and rupture.

Components of a Foamed Adhesive

The variables discussed in the preceeding section are controlled by manipulating the components in the adhesive foam mixture. These are a blowing agent, surface-active compound, water, and adhesive solids.

The blowing agents function is to produce and/or introduce the gas phase. Under ideal conditions, foam expansion should be predictable from the quantity of blowing agent added to the system. Methods of generating foams are categorized by the type of blowing agent and will be discussed later.

A surface-active compound is added to the adhesive to lower its surface tension. The surfactant forms a monolayer on the surface of a liquid and lowers the energy necessary to expand the surface of the adhesive, lessening resistance to formation of bubbles. Bubble size can be controlled by varying the concentration of surfactant. At a given expansion, bubbles will be larger if the surfactant concentration is low. Less surfactant means less monolayer surface area which in turn means less subdivision of the liquid to produce a given volume of foam, in other words, fewer and larger diameter bubbles.

Water and adhesive solids interact to affect the viscosity of the foam mixture. Urea formaldehyde adhesives used for wood particle composites are supplied at 64 - 66 percent solids (partially reacted polymer) with water as the solvent system. Dilution of the adhesive with water reduces its viscosity. A positive effect of dilution is lowered resistance to formation of thin films which increases the potential for foam expansion. Dilution is detrimental to foam stability.

Interfacial films thin and rupture more rapidly at lower viscosities.

The rate of polymerization of the adhesive may also be affected adversely.

Methods of Generating Foams

Several methods of generating foams were studied in a number of preliminary experiments. These methods are categorized as dispersion and chemical.

Dispersion foams are produced by incorporating air into the liquid. Air is introduced by mechanical agitation or by passing compressed air through a capillary or frit. It was not possible to generate foams of 50-100 times expansions by either of these methods at the viscosities of standard urea condensation adhesives. Dilution with water to below 50 percent solids adversely affected the cure speed of the adhesive and also decreased foam stability. For example, foams were made by whipping air into a urea formaldehyde adhesive at 66 percent and 50 percent solids. The 66 percent solids adhesive expanded less than one time; at 50 percent solids, the foam expansion was four times. Presumably, I could not obtain higher foam expansions because air could not be introduced spontaneously. The mechanical process of whipping additional air into the adhesive destroyed the foam as rapidly as it was formed. Volume increases of 10 times have been reported in the literature (Ziegler, 1958). With ultrasonic mixing

equipment even higher expansions may be a possibility.

Expansion of air introduced into the liquid under pressure might also be feasible with proper equipment. The impracticality of this approach with a batch laboratory process is illustrated with the ideal gas law, where:

$$P_1V_1 = P_2V_2$$

For example, take a pressure cylinder with a fixed volume of 100 cm³ of which 20 cm³ is occupied by liquid. This leaves 80 cm³ (V₁) for introduction of air under pressure. P₂ is atmospheric pressure (1 atm.) and V₂ is taken as the desired expansion (75X) to 1500 cm³. Solving for P₁, the air pressure needed to produce the foam is almost 19 atm. or 280 PSI. To be able to work at a reasonable pressure, for example 60 PSI, at an expansion of 75 times only about five cm³ of liquid can be added to the cylinder. The expanded foam volume is then 375 cm³ of which approximately one-third is left behind in the cylinder. Since the cylinder volume is large in relation to the foam volume, it is impractical to build a system to a scale needed for laboratory board making.

Continuous production of urea formaldehyde foam for applications such as insulation is based on mechanical expansion (Schutz, 1968). A mixture of adhesive and surfactant is pumped to a foaming head (nozzle) in which it is aerated to a foam. The foaming head is designed to create a turbulent path so that mechanical expansion can

take place continuously. Typical foam densities range from 0.2-0.8 pounds per cubic foot (>80 times expansion). This type of system could be adapted to continuous blending in a commercial particleboard process.

Chemical foams can be generated with (1) a thermally unstable compound that breaks down with heat releasing a gas; (2) a gas (i.e. CO_2 , N_2) formed by a chemical reaction or (3) a low boiling point organic liquid which volitalizes when brought to its boiling point.

Nitrogen containing azo-compounds are commonly used as blowing agents for rigid plastic foams. They decompose to form N_2 gas at temperatures generally above $200\,^{\circ}\text{C}$. These high temperatures drastically reduce gelation time of urea formaldehyde particleboard adhesives, leaving inadequate time for applying the foams. Thus, this system was not studied.

Foams generated by chemical reactions which have gas byproducts were studied in detail in a series of preliminary experiments.

They were difficult to control because the polymerization of the adhesive and the method of foaming are not independent. I developed and
studied in detail a system using the reaction of sodium nitrite,
ammonium chloride, and formaldehyde in a urea formaldehyde polymer
medium. This system illustrates the difficulty with chemical foams.

The gases which generate the foam are produced by several complex reactions involving nitrous acid. Some of the reactions which

have been postulated to occur are listed below:

(1)
$$NH_4^+ + HNO_2 = N_{2(g)} + 2H_2O + H^+$$

(2)
$$4\text{HNO}_2 + 3\text{HCOH} = 3\text{CO}_{2(g)} + 5\text{ H}_2\text{O} + 2\text{N}_{2(g)}$$

(3) HNO₂ decomposition

(a)
$$3HNO_2 = H^+ + NO_3^- + 2NO_2(g) + H_2O$$

(b)
$$2HNO_2 = NO(g) + H_2O + NO_2$$
 (g)

Because of the complexity of these reactions, the exact role of each component and their interactions were not known. Thus, empirical descriptions of the system were necessary, resulting in data (see Figure 4) which did not delineate an optimum combination of components. For example, the foam with the greatest expansion (32 times) had a pH of less than three. The pH of the foam was directly related to whether the adhesive would gel. If less than pH four, as in this case, the foam gelled almost instantaneously. If the pH was greater than six, the adhesive foam would not gel. A suitable gelation time would have resulted from critical combinations of ingredients making the system very non-reproducible. This system was abandoned because a combination of components which generated a reproducible foam volume and gel time was not encountered.

Blowing agents with boiling points slightly higher than room temperature (i. e. pentane) were tried and rejected because mild heat is necessary to produce the foam. These foams collapsed rapidly

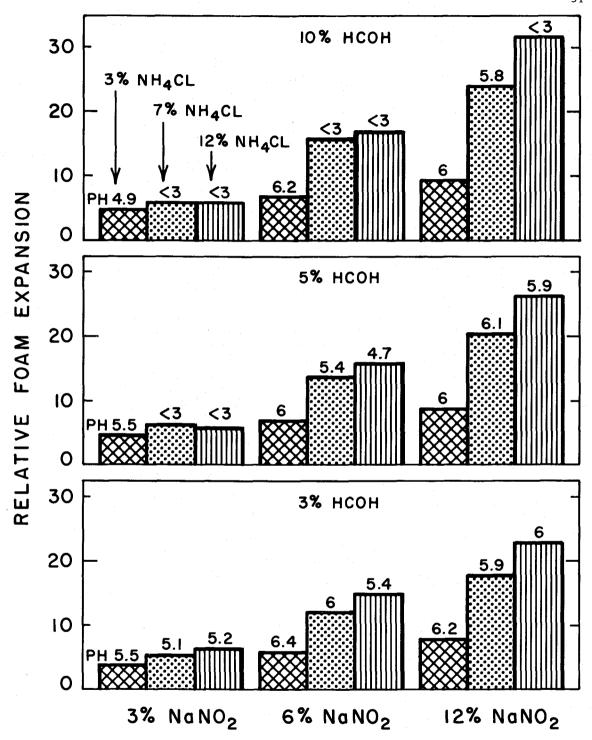


Figure 4. Foam expansion of a urea formaldehyde adhesive using a sodium nitrite - ammonium chloride-formaldehyde blowing agent.

when cooled even slightly. However, a system utilizing freon-12, which boils at room temperature, was developed for experimental application to particles. This system satisfied all of the criteria listed earlier. Foam volume and stability are easily controlled. While this system would not be practical for an industrial operation, it was controllable with simple laboratory equipment and allowed me to test the hypothesis of this study.

The Freon-Foam System

Freon-12, CF₂CL₂, has a boiling point of -22°C and is maintained in a liquid state under pressure. It can be combined with an adhesive - surfactant mixture under pressure. When the pressure is reduced to atmospheric, the freon boils and foams the adhesive.

A pressure cylinder as shown in Figure 5 was constructed for laboratory experiments. Foam was produced by adding adhesive-surfactant mixture to the chamber and sealing it with the cap. The cylinder was weighed and then connected to an inverted freon refrigerant tank via the needle valve. By inverting the tank the freon was injected as a liquid. Freon injection was controlled with a toggle switch located between the tank and the needle valve. A slight excess of the desired freon weight was injected into the pressure cylinder. The excess was bled off through the needle valve until an exact weight

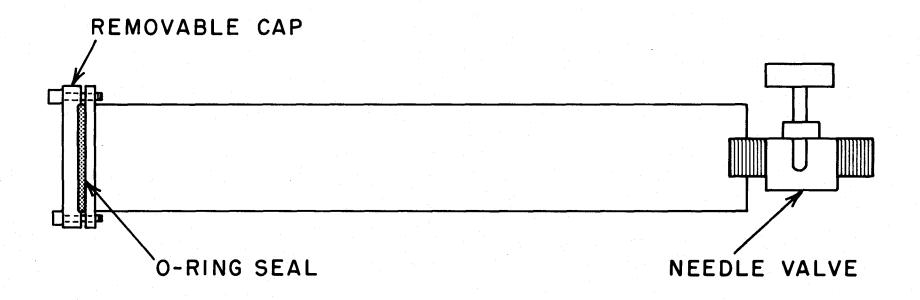


Figure 5. Pressure cylinder for producing urea formaldehyde foams with freon-12.

of freon was attained. Freon has a higher density than the surfactantadhesive mixture, but an emulsion was not formed until the two liquid
phases were agitated. The freon-adhesive emulsion was stable for
approximately five minutes. After emulsification, the needle valve
was opened and the foam extruded as a bead.

Figure 6 is a graph of foam expansion versus grams of freon per gram of adhesive based on expansion of 22 cm³ of adhesive-surfactant mixture in a chamber of 200 cm³. If more than two grams of freon per gram of adhesive were injected, it was not possible to emulsify all of the freon. The unemulsified freon boiled when the needle valve was opened, water condensed and froze the valve regulating the foam extrusion.

A foam expanded 85 times was used in all experiments with wood particles. This represents a foam density of approximately one pound per cubic foot.

Bubble sizes for one pound per cubic foot foams were measured as the foam extruded. They ranged from 0.02 - 0.05 cm in diameter.

The temperature of the interior of the foams immediately after extrusion was two to five degrees centrigrade. As the foam warmed, further expansion of the freon compensated for foam breakdown. This, coupled with the viscosity of the 66 percent solids urea formaldehyde adhesive, resulted in a highly stable foam.

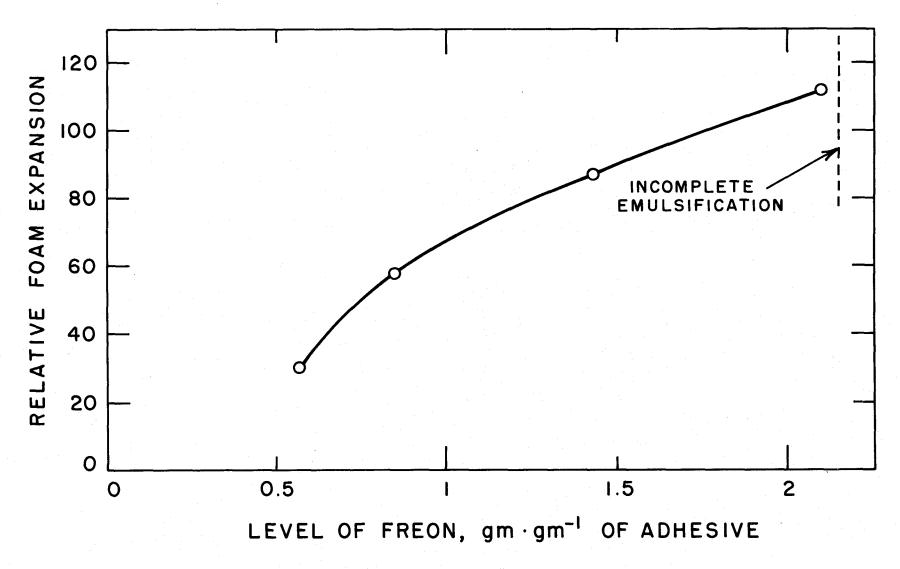


Figure 6. Foam expansion as a function of the ratio of freon-12 to adhesive.

III. DEVELOPING A BLENDING SYSTEM

Initially, two approaches to blending were considered. The first was to use conventional spray blending and foam the adhesive after applying it to the particles. The second was to generate a foam and develop a blender system for applying it to particles. The first alternative was rejected because it did not conform to the criteria established for the foaming and blending systems.

If the adhesive is foamed after blending, components which produce a gas must be present in the adhesive prior to spraying. This would restrict the type of blowing agent to (1) compounds that decompose to form a gas and/or (2) gases formed by chemical reactions.

A catalyst would be necessary to activate the blowing agent. The most functional catalyst would be heat which polymerizes the adhesive during board pressing. Urea formaldehyde adhesives are heat sensitive and are likely to be too advanced in molecular weight before reaching a temperature necessary for gas formation to occur. Other problems related to changes in adhesive behavior on addition of chemicals were discussed in Chapter II.

Even if a gas producing system were feasible, press cycles would have to be modified to prevent "blows" resulting from pressure build-up of entrapped gases. Also foaming would not be uniform throughout the thickness of the board because heat transfers from the surface inward.

Ideally, the system of foaming an adhesive after blending would redistribute the adhesive between the particles. The droplets deposited on particle surfaces would expand and increase bonding area. In practice, water loss from the adhesive into the wood and the resulting increases in viscosity would be variable. The high viscosity in some of these droplets on a wood surface would limit the degree of foaming. To counteract this, spread rates would have to be increased. This would make the process uneconomical. Deppe (1969) developed a method for coating the top of a particle mat with an isocyanate adhesive. As the mat was pressed, an activator foams the adhesive through the mat. The process was uneconomical because the ratio of wood to plastic was 1:2.

Blending the adhesive as a foam incorporates the potential advantages of increased surface area and volume. The foam should break down as it is applied and gases escape before pressing. Adhesive surface area for bonding would be established as the foam is deposited on particle surfaces.

The freon system for foaming was easily adapted to blending with particles. A blender for applying the foam was made by modifying a paddle-type mixer (See Figure 7). Threaded injection ports were soldered at eight staggered locations on the lower half of the mixing chamber circumference. The adhesive foams were prepared in eight pressure cylinders (See Figure 5) which were threaded into the

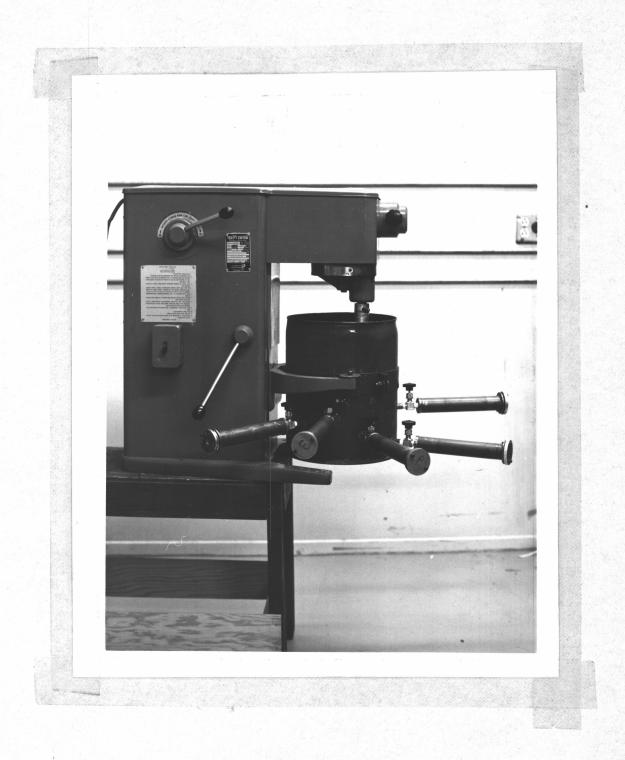


Figure 7. Experimental apparatus for blending of foam with wood particles.

injection ports. A batch of chilled particles (34°F) was placed in the chamber. The particles were chilled because heat generated by friction during mixing might have had adverse affects on adhesive cure rates. Foam was injected into the mixing chamber as the mixer stirred the particles. Injection rate was controlled by opening the valves gradually over a 2 1/2 minute time period.

IV. EVALUATING THE FOAM BINDER SYSTEM

An experiment was designed to compare the strength properties and adhesive distribution of wood particleboards made by three methods of applying the adhesive; (1) spraying, (2) injecting as a foam, and (3) injecting as a bulk liquid. I was interested in comparing foam to spray application, but because each required different kinds of blenders, that comparison would contain both blender type and foam versus spray sources of variation. Thus, the liquid injection was included to give an unconfounded comparison of foam versus non-foamed adhesive application. Boards were made at three target densities for each method of application so that any interaction of blending method with density could be detected. The adhesive level was held constant at seven percent solids. Table 4 is a summary of the experimental design.

Table 4. Summary of Experimental Design.

| Factors | No. of Levels | Levels Studied |
|-----------------------|---------------|--|
| Application | 3 | Spray, Foam (75X), Liquid |
| Densities/Application | 3 | 0.55, 0.75, 0.90 density (g/cm^3) for each application |
| Boards/Density | 3 | Three boards for each density |

Total Boards: $3 \times 3 \times 3 = 27$

Summary of Procedures

Preparation of Boards

All boards were prepared from a mixture of dried Douglas-fir and Hemlock planer shavings, plywood trim, and particles. The furnish was screened (-4 + 16) to produce a narrow particle size distribution (for detailed procedures, see Appendix).

Adhesive was applied as a spray in a rotary drum blender through an air atomizing nozzle. One board was made per batch of material sprayed.

The blending equipment and procedure discussed in Chapter III were used for applying both foamed and liquid adhesive to the particles. The liquid application deviated from the foam application only in that no surfactant was added and the pressure cylinders were connected at a 45° angle to the mixing chamber. Absence of surfactant eliminated foaming. Connecting the pressure cylinders at a 45° angle to the injection ports aided adhesive injection because of gravity flow. The volume expansion of the pressurized freon-adhesive mixture when reduced to atmospheric pressure forced the adhesive into the mixing chamber.

Processing steps for mat formation and pressing were the same for all boards produced. After applying the adhesive, a mat was formed by hand felting the particles onto a 12 inch x 12 inch caul

plate placed inside a deckle box. Board density was controlled by varying the weight of material in the mat. The mat was pressed for nine minutes at 325° F and 250 PSI. Board thickness was 3/4 inch. After pressing, the boards were cooled and conditioned at 70° F. and 65 percent relative humidity for three weeks.

Testing of Boards

Before testing, all boards were planed to 5/8 inch thickness.

In all strength tests, procedures outlined in ASTM standard D-1037-64 were followed as closely as possible.

Internal bond was determined from two inch by two inch specimens glued between metal blocks with an epoxy resin. They were loaded in tension perpendicular to the assembly at a head speed of 0.05 inches per minute.

Modulus of rupture and elasticity were determined for specimens tested on an eight inch span and deflected at a rate of 0.08 inches per minute. The span to thickness ratio (12.8) is less than recommended by the ASTM standard (24). This could not be avoided because board size was determined by the capacity of the blending equipment used for applying the foam.

Adhesive Distribution

Samples of furnish were collected after blending for each of the

three adhesive application methods. Particle distributions were determined by screening samples through selected standard Tyler screens (See Appendix). Samples of unblended furnish were also screened. The weight of material retained on each screen was recorded. All particle fractions were analyzed for nitrogen content with a Hewlett Packard CHN analyzer. The nitrogen contents for fractions to which adhesive had been applied were adjusted for nitrogen inherent in the wood using values determined from the unblended furnish. Adhesive solids content per fraction was calculated based on the nitrogen content per gram of polymerized urea formaldehyde adhesive.

In addition, to evaluate adhesive distribution on a surface area basis, the dimensions of fifty representative particles were measured for each fraction. For calculation of surface areas, the particles were assumed to be rectangular solids with smooth surfaces. A particle weight was determined by assuming a uniform particle density of 0.48. The surface area per gram was calculated from the surface area and weight. These were used to determine the relative surface areas of each fraction.

Results and Discussion

Strength Properties

Strength properties of boards made by applying the adhesive as

spray, foam, and liquid were compared statistically with a one-factor analysis of covariance treating density as the covariate. In this analysis mean strength properties were regressed to a common density and then compared (See Tables 5-7). This analysis was possible because strength properties increased linearly with increasing board density, and the slopes of the regression lines did not vary between application methods. Figure 8 is typical of the data obtained. Figures 9 to 11 are the regression equations for the various strength properties of boards made with different adhesive application methods. The statistical data are summarized in Tables 8 and 9.

Table 5. Summary of Data from Internal Bond Tests,

| Application | No. of Samples | Resin Content (%) | Density gm/cm ³ | I.B. PSI | Adjusted I. B. PSI ¹ |
|-------------|-------------------|-------------------|----------------------------|-------------|---------------------------------------|
| Spray | 36 | 7 | 0.705 | 164 | 175 |
| Foam | 36 | 7 | 0.745 | 171 | 167 |
| Liquid | 36 | 7 | 0.752 | 147 | 141 |
| Spray | 11 | 4 | 0.784 | 140 | • • • • • • • • • • • • • • • • • • • |
| Foam | 11 | 4 | 0.784 | 139 | |

Values adjusted to a density of 0.734 via regression

Table 6. Summary of Data from Modulus of Rupture Tests.

| Application | No. of Samples | Resin Content (%) | Density gm/cm ³ | M.O.R PSI | . Adjusted M.O.R. PSI ¹ | : |
|-------------|-------------------|-------------------|-------------------------------|--------------|--|---|
| Spray | 18 | 7 | 0.721 | 3001 | 3226 | |
| Foam | 18 | 7 | 0.754 | 3260 | 3161 | |
| Liquid | 18 | 7 | 0.757 | 3063 | 2937 | |
| Spray | 6 | 4 | 0.763 | 2680 | - - | |
| Foam | 6 | 4 | 0.762 | 2574 | ************************************** | |

Values adjusted to a density of 0.744 via regression

Table 7. Summary of Data from Modulus of Elasticity Tests.

| Application | No. of Samples | Resin Content (%) | Density (g./cc) | MOE (psi)x10 ³ | Adjusted MOE ¹ (psi)x10 ³ |
|-------------|-------------------|-------------------|--------------------|------------------------------|---|
| Spray | 18 | 7 | 0.721 | 522 | 559 |
| Foam | 18 | 7 | 0.754 | 584 | 568 |
| Liquid | 18 | 7 | 0.757 | 566 | 545 |

¹ Values adjusted to a density of 0.744 via regression

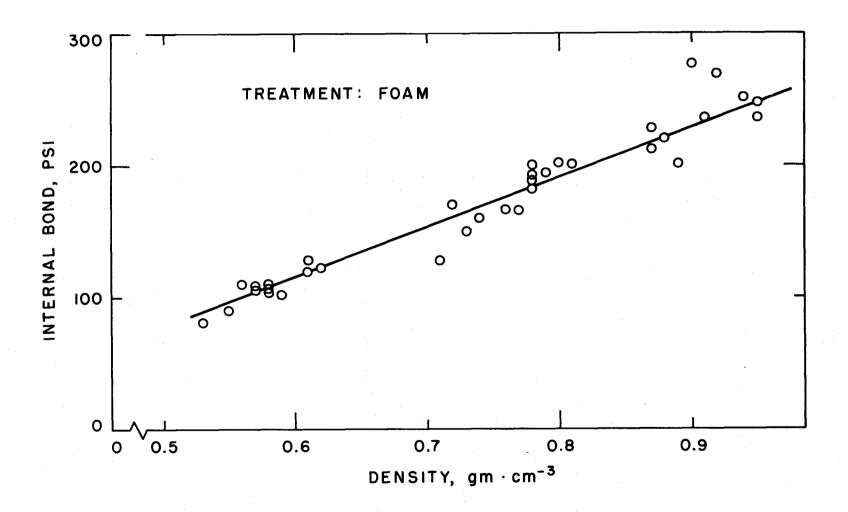


Figure 8. Typical data for internal bond strength as a function of density.

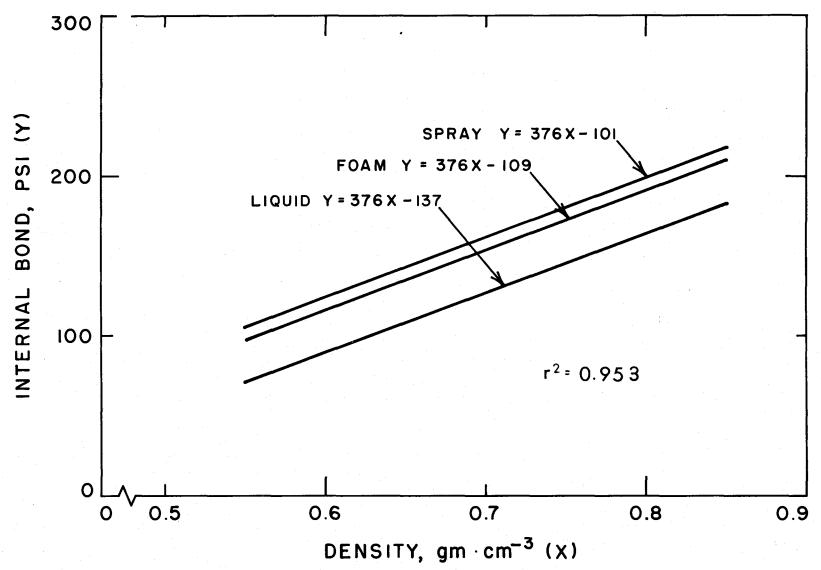


Figure 9. Regression lines of internal bond strength as a function of density.

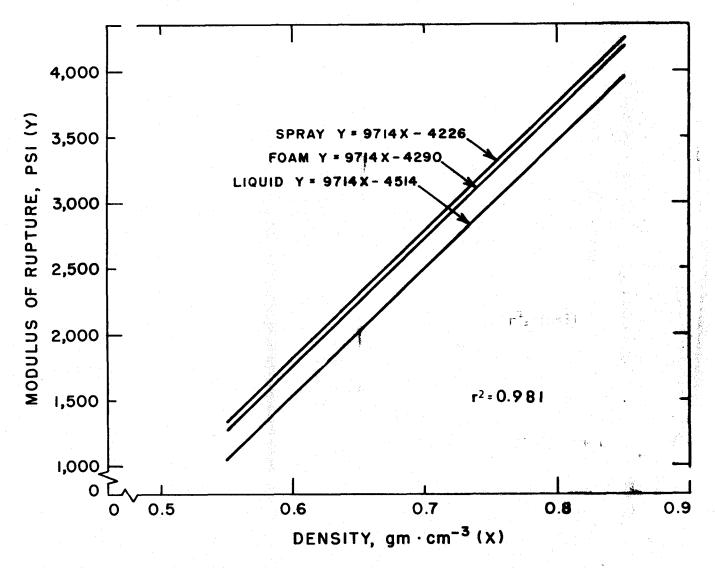


Figure 10. Regression lines of modulus of rupture as a function of density.

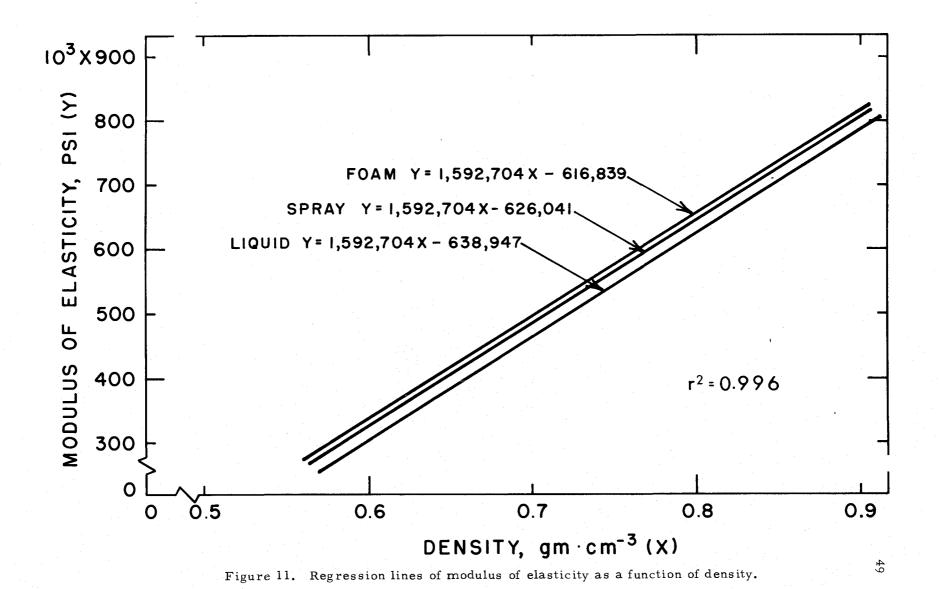


Table 8. Analysis of Covariance for Strength Properties.

| Source | D.f | s. s. | M. S. | F |
|----------------|------------|------------------------|----------------------|-----------|
| | | Internal Bond | | |
| Total Adjusted | 106 | 34,827 | | |
| Applications | 2 | 22,321 | 11,160 | 92.8 ** |
| Error | 104 | 12,506 | 120.2 | |
| | <u>M</u> | odulus of Ruptu | <u>re</u> | |
| Total Adjusted | 52 | 2,340,682 | | |
| Applications | 2 | 816,535 | 408,268 | 13.93 ** |
| Error | 50 | 1,524,146 | 30,483 | |
| | <u>M</u> c | dulus of Elastic | city | |
| Total Adjusted | 52 | 2.587x10 ⁶ | | |
| Applications | 2 | 1.829x10 ¹⁰ | 9.149x10 | 9 61.1 ** |
| Error | 50 | 7.474×10^9 | 1.49x10 ⁸ | |

^{***} Significant difference occur among adjusted means at the 95% level.

Table 9. Probability of Differences in Adjusted Strength Property Means Occurring by Chance Rather Than as a Result of the Application Method.

| Comparison | Adhesive Content (%) | Probability | | | |
|------------------|----------------------|-------------|--------|--------|--|
| | (14) | IB | MOR | MOE | |
| Spray vs. Foam | 7 | 0.025 | 0.29 | 0.03 | |
| Spray vs. Liquid | 7 | <0.001 | <0.001 | 0.0015 | |
| Foam vs. Liquid | 7 | <0.001 | <0.001 | <0.001 | |
| Foam vs. Spray | 4 | 0.80 | 0.52 | | |

Spray and foam application of adhesive produced boards with internal bonds superior to those made by injection of liquid adhesive (See Table 5 and Figure 9). The adjusted mean internal bond strength was 28-35 PSI below foam and spray boards. The bending properties (MOR, MOE) of foam and spray boards were also superior to those made by liquid injection (See Tables 6-7 and Figures 10-11).

Liquid injection involved applying of a small adhesive surface area to a large particle surface area. The probability of a particle receiving adhesive as it is injected was low. A small portion of the particles received all of the adhesive. The excess was then transferred to other particles via the rubbing action of one particle against another as they were stirred. Therefore, the surface of a particle receiving adhesive by transfer was probably not uniformly covered. In addition, a portion of the particles probably received no adhesive. This would have created weak zones in the board and lowered its overall strength. The lower strength properties obtained with liquid injection demonstrates that interparticle transfer of adhesive was less efficient than with the foam application.

Since the foam and liquid were blended in the same experimental mixer, the better strength properties of foamed boards are interpreted to mean better adhesive distribution occurred. The more efficient distribution is attributed to the adhesives increased surface area and therefore a higher probability that more particles received adhesive.

Foam application at seven percent solids resulted in an internal bond strength slightly lower than spray application. The adjusted mean difference was eight pounds per square inch and this small difference was statistically significant at the 97.5 percent level (See Table 9). Assuming that resin distribution becomes more critical at lower application rates, I made additional boards containing four percent adhesive solids. These results are also summarized in Tables 5 and 9. In contrast to the greater adhesive solids spreads, at four percent adhesive solids and at an average density of 0.784 there was no significant difference in the internal bonds between foam and spray application. Internal bond strength at four percent adhesive solids was approximately 40 PSI lower than at seven percent solids for an equivalent density. For modulus of rupture, there was no significant difference between foam and spray applications at either level of adhesive solids (See Table 9). On the other hand, the modulus of elasticity of foam boards was slightly greater than that of spray boards. Thus, it appears that no consistent difference exists between the properties of foam and spray boards.

From an industrial standpoint, the statistically significant differences in internal bond and modulus of elasticity are not important.

For example, an eight to fifteen PSI variability of internal bond probably represents the limits of control in a commercial production

situation. The eight PSI difference detected in this study is marginal or barely within those limits.

It was postulated in Chapter II that application of adhesive as a foam should result in a better distribution than by spraying the adhesive. This was not reflected in the strength property results.

The full potential of the increased surface area of a foam was not realized because of limitations imposed by the design of the blending apparatus. The foam had to be injected from the outer surface of the blender and was expected to penetrate six inches into the interior to reach all the particles. Injecting it from the mixing blade would have been a definite improvement. Also, the foam was injected rapidly because of problems caused by the stirring action of the paddle. Stirring the particles generated heat which had to be minimized so that adhesive cure would not be affected. As stated in Chapter II, this was partially resolved by cooling the furnish to 34°C before blending. Even so, the blending time had to be kept short to eliminate the effects of The same stirring action also caused a reduction in particle Significant changes in the size of particles affect strength The experiment was intended to detect differences in properties. strength properties due solely to the method of adhesive application. Both of these effects were minimized by reducing the foam injection time. But rapid injection caused the foam to form "clumps" which were dispersed by the paddle onto the particles. The foam surface

area per unit of volume exposed to particles was lower than if the foam had been extruded slowly as a cylindrical bead.

As stated previously, because of friction during mixing the particle size distribution of the furnish used in foam boards was slightly different from that used in spray boards. The particle size distributions are summarized in Table 10. To determine if this difference had an effect on strength properties three additional boards were made with the furnish in column five of Table 10. This particle size distribution was generated by stirring unblended furnish in the foam mixer for the standard 2 1/2 minutes. Adhesive was then applied to these particles at seven percent solids in the spray blender. During spray blending, some of the -20 + 32 and -32 fractions were lost (See Column 6). They probably adhered to the larger particles or to the sides of the blender. The strength properties of these boards were compared to those of the original spray boards made with the furnish in column three of Table The 95 percent confidence bands for the average internal bond and modulus of rupture overlap those for the mean values of spray boards made with the altered furnish (See Table 11). Based on these results, the altering of the particle-size distribution of the furnish in the foam blender cannot be considered to have affected the comparison of strength properties between the foam and spray methods of adhesive application. This would also apply to differences between spray application and liquid injection.

Table 10. Particle-Size Distribution for Foam and Spray Application.

| | Origin | nal Design | Altered Sp | ray Finish |
|---------|--------------------------------|-------------------------------|--------------------------------|-------------------------------|
| | % screen before blending | % screen after blending | % screen before blending | % screen after blending |
| Mesh # | | Foam Spray | Spray | Spray |
| + 8 | 11.0 | 8.7 11.7 | 8.6 | 9.4 |
| - 8 +10 | 24.8 | 21.8 25.3 | 23.3 | 25.3 |
| -10 +16 | 49.9 | 46.8 47.8 | 47.4 | 47.0 |
| -16 +20 | 7.0 | 9.3 7.5 | 8.7 | 8.6 |
| -20 +32 | 6.5 | 10.3 6.8 | 9.3 | 7.9 |
| -32 | 0.5 | 2.7 0.5 | 2.7 | 1.7 |
| | 100.0 | 100.0 100.0 | 100.0 | 100.0 |

Particle-size distribution for liquid application varied insignificantly from the particle-size distribution for foam application.

Table 11. Strength Properties of Spray Boards Made from Altered Furnish.

| Furnish | Density (g./cc) | Internal Bond (psi) | Density (g./cc) | Modulus of Rupture (psi) |
|----------------|-----------------|------------------------|-----------------|--------------------------|
| Altered Spray | 0.783 | 204 ± 7.6 ¹ | 0.757 | 3185 ± 298 ¹ |
| Original Spray | 0.783 | 194 ± 8.9^2 | 0.757 | 3350 ± 141^2 |

 $^{^{1}}$ 95% confidence limits of the mean strength value

²95% confidence bands of the regression equation at this strength value

There was a difference in the adhesive tack of a furnish blended with foam versus spray. Tack was not measured quantitatively but was estimated by the feel of the furnish after blending. Furnish with adhesive applied as a foam was tacky when removed from the mixer. This tack diminished during forming and was negligible by the time the mat was placed in the press. Adhesive tack on sprayed furnish diminished only slightly during board assembly. The effect was probably related to spreading the adhesive as a thin film as opposed to deposition of droplets. Less adhesive would be concentrated at a given point on a particle surface with foam application. If the absence of tack influenced strength properties, the effect was negligible as evidenced by the equivalent strengths of foam and spray boards. Whether or not tack influences strength has not been clearly established in the literature. The observed difference in tack is included also because adhesive tack is important in the formation and integrity of the particle mat on an industrial scale. For the traditional forming processes used commercially, tack is necessary to hold the mat together as it is conveyed to the press. It can also be the cause of excessive buildup of furnish on the walls of the blender. Tack is undesirable in new particle alignment technology which is in the developmental stage at present.

Applying the adhesive as thin films has been criticized because it is thought that starved gluelines occur between particles. The

effect of starved gluelines would be to lower strength properties. This was not observed for foam application, which should result in deposition of thin films of adhesive on particle surfaces.

The spray system against which the foam application was compared was equivalent to that used by Lehman (1965, 1968, 1970) to produce optimum laboratory boards. Foam boards had properties equivalent to this optimum. For the reasons already discussed it is felt that the optimum for foam boards was not reached. Further experimentation would be necessary to verify this assertion. It is well established that industrial spray systems do not perform at the optimum level attained under laboratory conditions. In fact, commercially produced boards typically have only 70 percent of the strength of these optimum laboratory boards (Carroll and McVey, 1962, Meinecke, 1960). This difference is attributed to a non-uniform adhesive distribution produced in a commercial blender. Based on the results of this study, a foam application system could not be justified unless it could be shown to perform on an industrial scale closer to its laboratory optimum than is true of spray systems. This may be feasible by incorporating a foam with the new Littleford blender. This blender stirs the particles with a rotating shaft equipped with paddles. Liquid adhesive is injected through ports also located on the rotating shaft. Commercially produced boards made with this blending system are equivalent to those made with conventional spray systems. Instead

of "dripping" the adhesive onto the particles as is now done, it could be pumped in as a foam. The superiority of the foam over a liquid injection was well established in this study. If foam systems can perform better in the field, adhesive consumption could be reduced while maintaining the board property levels at which the industry now operates.

The method for generating foams in this study would not be feasible on an industrial scale because of the high cost of the freon blowing agent. Alternative low cost technology does exist. Urea formaldehyde foams of densities less than one pound per cubic foot can be produced by mechanical expansion (Schutz, 1968). The feasibility of this was discussed in Chapter II. An adhesive surfactant mixture is pumped to a foaming nozzle in which compressed air is emulsified with the liquid by turbulence. The system is designed so that foaming is a continuous process. As stated previously, a system such as this could be adapted to use with the Littleford blender. In summary, existing technology for blending could be converted if further experimentation in the laboratory proved foam application desirable.

Adhesive Distribution

Data for adhesive distribution by the three application methods is summarized in Tables 12 - 14. In addition to the percentage of the

Table 12. Adhesive Distribution by Spray Application.

| Mesh # | % Screen Weight After Blending | % Total Adhesive Solids | % Relative Surface Area ¹ | % Relative Surface Area ² | % Relative Number of Particles 1 |
|---------|-----------------------------------|----------------------------|--|--|-------------------------------------|
| + 8 | 11.7 | 10.1 | 7.5 | 5.3 | 7, 2 |
| - 8 + 9 | 11.0 | 10.1 | 8.4 | 6.7 | 6.6 |
| - 9 +10 | 14.3 | 12.0 | 10.5 | 10.3 | 10.1 |
| -10 +14 | 33.5 | 29.8 | 32.7 | 32.9 | 32.2 |
| -14 +16 | 14.2 | 14.4 | 16.4 | 17.2 | 16.8 |
| -16 +20 | 7.5 | 12.7 | 10.8 | 11.2 | 10.9 |
| -20 +32 | 6.8 | 9.8 | 12.0 | 14.4 | 14.1 |
| -32 | 0.5 | 1.1 | 1.7 | 2.0 | 1.9 |
| | 100.0 | 100.0 | 100.0 | 100.0 | 100.0 |

Based on measurement of particles of a density of 0.48

²Calculated based on Duncan's assumptions (1974)

Table 13. Adhesive Distribution by Foam Application.

| Mesh # | % Screen Weight After Blending | % Total Adhesive Solids | % Relative Surface Area ¹ | % Relative Surface Area ² | % Relative Number of Particles |
|---------|-----------------------------------|----------------------------|--|--|--------------------------------|
| 8 | 8.7 | 4. 2 | 5.1 | 3.4 | 3,6 |
| - 8 + 9 | 9.0 | 6.5 | 6.2 | 4.8 | 4.5 |
| - 9 +10 | 12.8 | 8.1 | 8.5 | 8.1 | 7.6 |
| -10 +14 | 32.3 | 26.6 | 28.4 | 27.8 | 27.2 |
| -14 +16 | 14.5 | 15.7 | 15.1 | 15.3 | 14.5 |
| -16 +20 | 9.3 | 18.6 | 16.3 | 19.1 | 20.6 |
| -32 | 2.7 | 9.5 | 8.4 | 9.4 | 10.8 |
| | 100.0 | 100.0 | 100.0 | 100.0 | 100.0 |

Based on measurements of particles of a density of 0.48

²Calculated based on Duncan's assumptions (1974)

Table 14. Adhesive Distribution by Liquid Application.

| Mesh # | % Screen Weight After Blending | % Total Adhesive Solids | % Relative Surface Areal | % Relative Surface Area ² | % Relative Number of Particles l |
|---------|-----------------------------------|----------------------------|--------------------------------|--|----------------------------------|
| + 8 | 8.0 | 4.3 | 4.5 | 3.1 | 4.2 |
| - 8 + 9 | 8.6 | 4.3 | 5.8 | 4.5 | 4.4 |
| - 9 +10 | 12.2 | 7.4 | 8.0 | 7.6 | 7.6 |
| -10 +14 | 32.2 | 29.9 | 27.7 | 27.0 | 26.7 |
| -14 +16 | 15.7 | 14.3 | 16.0 | 16.2 | 15.9 |
| -16 +20 | 9.7 | 12.6 | 12.2 | 12.3 | 12.1 |
| -20 +32 | 10.6 | 15.7 | 16.6 | 19.3 | 19.1 |
| -32 | 3.0 | 11.5 | 9.2 | 10.1 | 10.1 |
| | 100.0 | 100.0 | 100.0 | 100.0 | 100.0 |

Based on measurements of particles of a density of 0.48

²Calculated based on Duncan's Assumptions (1974)

total furnish weight and adhesive solids per screen fraction, the relative number of particles, and the relative surface area of each fraction were calculated according to a method developed by Duncan (1974).

The relative surface area was also determined from direct measurements of particles. Table 15 is a summary of the average particle size for each screen fraction.

Duncan's calculations are based on the idea that either volume or weight can be used as a measure of wood substance retained on a screen if the density of the material being screened is uniform. get the relative particle volume of a particle in a screen fraction he assumed that the relative particle volume is equal to the mesh opening of the screen in question divided by the coarsest mesh opening in the screen analysis. Duncan used this assumption in deriving formulas for calculating relative number of particles and relative surface areas. The assumption would be valid if a particle is rectangular (assumed by Duncan) and two of its dimensions remain relatively constant as particle size changes. My average particle sizes (See Table 15) showed that this is true only for particle length if the largest (+8, +9) and smallest (-32) fractions are excluded. To calculate relative surface areas Duncan further assumes that "particles retained on a given screen have two dimensions finer than the previous screen (M_1) and two dimensions coarser than the screen retained on (M2). Therefore \cdot . . one can say that one (dimension) equals M_2 and the other

(dimension equals) one-half the sum of the two screens, i.e. M_1 + $M_{2/2}$." My particle sizes in general had width and thickness dimensions smaller than the preceding screen and length and width dimensions larger than the screen on which they were retained. The second part of the quoted statement, when calculated from actual mesh openings, means that two of the particles dimensions are similar in magnitude. For actual particles (See Table 15) this was true only in a relative sense in that the ratios of length to width or thickness were much greater than the ratio of the smaller dimensions to each other. However, relative surface areas based on the average particle dimensions paralleled those calculated from Duncan's assumptions (See Tables 12-14). Thus, the data verified Duncan's derivations even though some of the assumptions seem questionable.

The three application methods produced different adhesive distributions. This was true whether based on surface area or weight. For spray applications the larger particles (+8, +9, +10 mesh) received more adhesive than surface area contributed (See Table 16). The ratio of adhesive solids to relative area was 1.44 on the larger particles. Smaller fractions received slightly less adhesive than area contributed (0.87). Duncan (1974) and Cox (1974) also found that for a sprayed furnish the larger particles received an adhesive portion greater than their relative area. In contrast, for foam and liquid

Table 15. Average Particle Dimensions. 1

| Mesh Number | Mesh Opening (cm) | Length (cm) | Width (cm) | Thickness (cm) |
|----------------|----------------------|----------------|---------------|-------------------|
| + 8 | 0.238 | 0.778 | 0.315 | 0.083 |
| + 9 | 0.200 | 0.659 | 0.243 | 0.070 |
| +10 | 0.168 | 0.532 | 0.206 | 0.074 |
| +14 | 0.119 | 0.492 | 0.149 | 0.058 |
| +16 | 0.100 | 0.490 | 0.098 | 0.050 |
| +20 | 0.084 | 0.403 | 0.097 | 0.039 |
| +32 | 0.050 | 0.382 | 0.067 | 0.035 |
| -32 | 0.025 | 0.204 | 0.040 | 0.016 |

All dimensions are based on measurement of 50 particles.

Table 16. Comparison of Adhesive Distribution on Large and Small Particles.

| Application | Large Particles l | Small Particles ² |
|-------------|-------------------------------|------------------------------|
| Ratio of A | dhesive Solids to Relative Su | rface Area (%:%) |
| Spray | 1.44 | 0.87 |
| Foam | 1.15 | 0.97 |
| Liquid | 1.04 | 0.98 |
| Ratio of Ad | hesive Solids to Relative Fra | ction Weight (%:%) |
| Spray | 0.88 | 1.06 |
| Foam | 0.63 | 1.15 |
| Liquid | 0.56 | 1.17 |

All particles retained on eight, nine, and ten mesh screens

²All particles passing through a ten mesh screen

injection, the adhesive was distributed approximately proportional to the surface area of the large and small particles.

The distribution of adhesive proportional to surface area by foam and liquid injection would be expected. For liquid injection, only a small portion of the particles received the adhesive directly, the remainder receiving adhesive by frictional transfer. Interparticle transfer should not be disproportionate according to particle size. In addition, there is no reason to believe that injecting the adhesive as a foam would discriminate in favor of certain particle sizes. As noted earlier, the rubbing action in the blender used for foam and liquid injection caused a reduction in particle size. This may have had a subtle influence on adhesive distribution since surface area was being created on smaller particles during the blending process.

The disproportionate amount of adhesive on coarse fractions when sprayed is unexplainable. Resin droplets of 30-40 microns should not discriminate between large and small particles as targets since the particles are all substantially larger (See Table 15). It would seem, therefore, as Duncan concluded, that adhesive distribution should parallel the relative area. The coarse particles may have picked up adhesive by interparticle transfer from smaller particles. There are three to four times as many smaller particles. This could not have occurred to any substantial degree because very little "rubbing" action occurs in a rotary drum blender.

Adhesive solids distribution based on weight was different from that based on surface area. The trend for all three application methods was to increased adhesive solids per fraction weight as particle size decreased. Several investigators have reported similar results (Cox, 1974, Duncan, 1974, Maloney, 1970, Schwarz et al., 1968). This was most pronounced for foam and liquid injection. The three coarse fractions had only 55-65 percent of the adhesive solids they should receive based on their weight (See Table 16). Smaller fractions had more adhesive than weight contributed. The magnitude of change with particle size was least dramatic for the sprayed furnish which was nearly proportional to weight.

Strength properties were not correlated to adhesive distribution according to particle size. Boards made by foam and spray applications differed only slightly in strength properties but had dissimilar adhesive distributions. Liquid injection boards were significantly lower in strength but had an adhesive distribution equivalent to those produced with a foam. As stated previously, the strength properties would be influenced by the number of particles that received no adhesive, and the uniformity of coverage on surfaces which received adhesive. These factors must have been more important than the adhesive-particle size distribution in determining board strength.

V. SUMMARY AND CONCLUSIONS

This study evaluated foam application of adhesive for wood particle composites. A system was developed for foaming a urea formaldehyde adhesive and for applying the foam to wood particles. The foam was produced by expanding the adhesive with liquid freon-12 (C F₂ Cl₂) which boils at -22 °C. Foams with a density of less than one pound per cubic foot were produced by this method. Wood particles and foam were blended by injecting the foam from eight pressure cylinders fixed on the circumference of a mixing chamber in which the wood particles were being stirred.

The foam system for applying adhesive was evaluated by comparing it to spray and liquid application. Spray application of adhesive is the method most widely used commercially. Foam and spray application required different blending equipment, so liquid application was included in the design to estimate differences between foam and spray application due to the type of blender. Wood particle composites were made from furnish blended by each application method and their strength properties and adhesive distributions were determined.

The application of adhesive as a foam produced wood particle composites with strength properties equivalent to those where adhesive was applied as a spray, even though the foam system seemed to be

operating below its optimum. Applying the adhesive as a liquid resulted in significantly lower strength properties, suggesting that improvement is possible by foam extending adhesive prior to liquid injecting.

Spray application distributed the adhesive disproportionately heavy on the larger particles, based on surface area. Adhesive distribution on smaller particles was slightly less than proportional to surface area. Injecting the adhesive as a foam or liquid produced an adhesive distribution on each fraction approximately proportional to its surface area. Based on fraction weight, the proportion of adhesive increased with diminishing particle size. The magnitude of change with decreasing particle size was greatest for adhesive applied as a liquid.

Differences in strength properties resulting from the three application methods did not parallel differences in adhesive distribution. Strength properties were probably significantly influenced by the number of particles or surface area receiving no adhesive. The probability of more particles receiving adhesive would be greater for foam and spray application than for liquid injection. This was attributed to the increased surface area of the adhesive. With liquid injection the mechanism for distributing the adhesive was probably by frictional transfer between particles.

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

- 1. Foam application of adhesive for wood particle composites is equivalent to spray application on a laboratory scale.
- 2. Foam application is superior to applying the adhesive as a liquid, which depends on frictional transfer to distribute the adhesive.
- The superiority of foam and spray application cannot be explained by the distribution of adhesive according to particle size.

A foam application system could not be justified on a commercial scale unless it resulted in decreased adhesive consumption while maintaining current board property levels. The new Littleford blending system, now in use commercially, blends adhesive by injecting it as a liquid. This system produces boards with properties equivalent to those produced by commercial spray application. The superiority of foam over liquid injection was shown in this study. Therefore, reduced adhesive consumption may be feasible with such a foam system.

The method used for generating foams in this study would not be feasible on an industrial scale because of the high cost of the blowing agent. Technology does exist however for low cost continuous production of mechanically expanded urea formaldehyde foams with properties equal to those produced in this study.

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EXPERIMENTAL PROCEDURES

Raw Materials

The wood particle furnish used in this study was a mixture of dried Douglas-fir-hemlock planer shavings, plywood trim, and particles. It was obtained from Duraflake, Co. of Albany, Oregon. A urea formaldehyde adhesive, WS-155-11B, at 66 percent solids was supplied by Borden Chemical Company of Springfield, Oregon. The surfactant used in the foam mixture was Emmersol 6462 Sodium Alkyl Sulfate, supplied by Emery, Industries. Freon-12 refrigerant was obtained in pressurized cylinders containing 15 pounds.

Blending Procedures

Spray Application

Adhesive was applied in a rotary drum blender by spraying it through an air atomizing nozzle. The nozzle was situated so that the spray pattern covered the width of the drum. Atomization air pressure was maintained at 60 PSI.

For each board the blender was charged with 2500 grams of screened furnish. The moisture content of the screened furnish had to be adjusted so that after addition of adhesive the moisture content would be that desired for pressing $(10.0 \pm 0.5\%)$. This was done by

spraying on water immediately preceding application of adhesive.

After blending a batch of furnish, its moisture content was determined. A mat was then formed and pressed as follows.

Foam Application

As with the spray system, the moisture content had to be adjusted so that it would be at the proper level for pressing. This was done by spraying water on 3500 gram (oven-dry basis) batches in the spray blender. The furnish was then divided into portions based on the weight needed for a 12 inch x 12 inch board of 55 pounds per cubic foot density. Each portion (1710 grams, oven-dry basis) was placed in a plastic bag and stored in a conditioning room at 34°C until made into a board.

An adhesive mixture for foaming was prepared by adding two percent of Emmersal surfactant (weight basis) to 350 grams of 66 percent solids urea formaldehyde adhesive. After stirring, the mixture was placed in a 250 cm³ buret. Equal aliquots of adhesive mixture were added to each cylinder. This was based on the amount of adhesive needed for the application of seven percent solids to 1710 grams (dry weight basis) of furnish. The cylinders were then sealed and weighed.

To inject freon into the foam cylinder a quick release couple was threaded onto the needle valve of the pressure cylinder, and

connected to an inverted pressurized freon tank through a toggle valve. Liquid freon was injected by inverting the freon tank. A slight excess of the desired weight of freon was obtained by injecting for three seconds. The cylinder was then re-weighed and excess freon was removed by opening the needle valve. Twenty-five grams of freon were added to each cylinder. When all eight cylinders were loaded with freon, it was emulsified in the adhesive by shaking. Emulsification occurs when no evidence of liquid movement can be detected inside the cylinder. This procedure produced eight foams that had nearly identical expansions when released - 85 X or 1800 cc per cylinder.

The loaded cylinders were threaded horizontally into the injection ports on the mixing chamber. A batch of chilled particles was added to the chamber and foam was injected through the needle valves while the mixer stirred the particles. In order to inject the foam at a uniform rate, the valves were opened gradually. Foam injection time was 2 1/2 minutes.

The cylinders were then removed and re-weighed as a check for completeness of injection. The furnish was processed into a board as follows.

Liquid Application

Liquid was applied to the furnish with the same experimental

apparatus used for foam application. No surfactant was added to the adhesive. The procedures for loading the cylinders with adhesive and freon were identical to those used for preparing foams.

Gas released by reducing pressurized liquid freon to atmospheric pressure forced the adhesive out of the cylinders. Gravity flow was also found to assist ejection of the adhesive. The horizontal connection to the mixing chamber was modified by introducing a 45 of elbow.

When all eight cylinders were loaded with freon and adhesive, they were agitated to disperse the freon in the adhesive. Injection onto the particles in the mixing chamber was initiated as soon as possible after agitation. This was necessary because the more dense freon separated rapidly from the adhesive and boiled, leaving the adhesive inside the cylinder.

Chilled particles were used as with the foam system. Injection time was 2 1/2 minutes. The cylinders were re-weighed after injection to check for completeness of adhesive removal. Boards were formed as follows.

Preparation of Boards

Processing steps and variables which were held constant for all boards are listed and discussed below:

Particle Size distribution: -4 +16 mesh

Board size: 12 inches x 12 inches

Thickness: 3/4 inch, planed to 5/8 inch

Moisture content of mat

into press: 10.0 + 0.5 percent

Press closing time: 15-30 seconds

Press cycle: 9 minutes at 325 F

The particle furnish was collected at Duraflake as it left the driers. Its moisture content averaged 4.0 ± 0.5%. All furnish was screened to produce a narrow particle size distribution. Fractions remaining on a 4 mesh screen and passing through a 16 mesh screen were discarded. The screened furnish was thoroughly mixed, sealed in plastic bags and stored in a conditioning room at 90° F and 30 percent relative humidity until board preparation.

Mat forming and pressing were the same for all boards produced. After applying the adhesive, mats were formed by hand felting the furnish onto a 12 inch x 12 inch caul plate placed in a deckle box. The weight of material per board was based on that needed to produce the desired density at the moisture content of the furnish after blending $(10.0 \pm 0.5\%)$.

The formed mat plus caul plates were placed in a hot press equipped with 3/4 inch stops. Stops are metal bars that prevent the platens from closing further than a pre-determined thickness.

Sufficient pressure was applied to close the press to the 3/4 inch thickness in 15 - 20 seconds. The pressure was then gradually reduced to 250 PSI, while maintaining thickness. It was held at this level for nine minutes and was released gradually over the last 30 seconds of the press cycle.

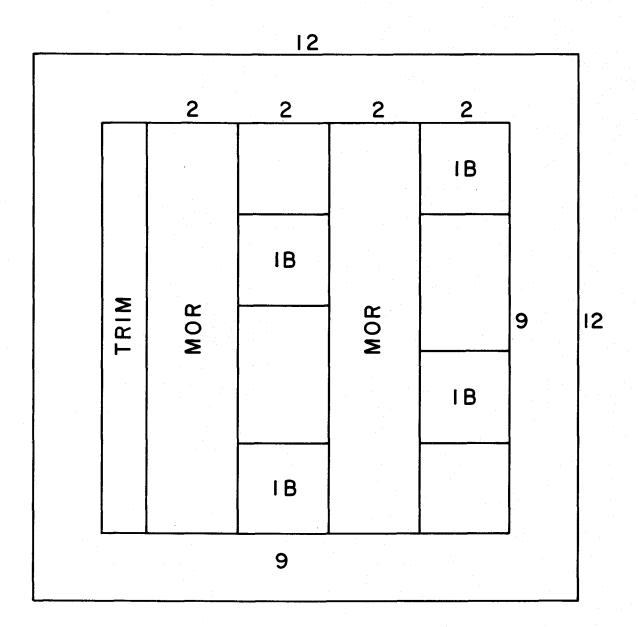
After pressing, the boards were cooled and placed in a conditioning room at 70° F and 65 percent relative humidity for three weeks. All boards were planed to 5/8 inch thickness before testing.

Testing of Boards

After conditioning, each board was planed to 5/8 inch thickness, trimmed to nine by nine inches and cut into four strips (See Figure 12). Two of the strips were used in static bending tests. The remaining two were cut into eight two by two inch internal bond specimens of which four were tested. ASTM standard D-1037-64 test procedures were followed as closely as possible.

For the internal bond tests, the volume and weight of the sample was recorded. Each two by two inch specimen was glued between two metal blocks with an epoxy adhesive. They were tested at a load rate of 0.05 inches per minute. If the sample fractured in the face, the test value was discarded and another sample was tested.

The two by nine inch static bending specimens (eight inch span) were loaded at a rate of 0.08 inches per minute on an Instron testing



CUTTING PATTERN

Figure 12. Cutting pattern for obtaining samples used in strength property tests.

machine. Load-deflection curves were recorded and were used to calculate stiffness values (MOE). Modulus of rupture (MOR) was calculated from the load at which the sample failed. The volume and weight of each specimen was recorded before testing.

Adhesive Distribution

Samples of furnish were collected after blending for each of the three adhesive application methods. Particle distributions were determined by screening samples through selected standard Tyler screens. Fractions were collected at 8, 9, 10, 14, 16, 20, 32 and smaller than 32 screen mesh sizes. Each sample weighed 150 grams and was screened for 15 minutes. Four replications were made for each adhesive application method. Three samples of furnish collected before blending were also screened. The weight of material retained on each of the eight screens was recorded and all fractions were saved for nitrogen analysis.

A sample of each fraction was ground separately to -60 mesh size with a Wiley mill. The sample was then reduced to a fine, homogeneous powder by placing it in a wiggle-bug for 30 seconds.

All samples were oven-dried for two hours at 105°C before nitrogen analysis.

A sample of thermoset urea formaldehyde adhesive was prepared according to the West Coast Adhesive Manufacturers Association (1957) procedure for determination of adhesive solids. The adhesive was reduced to a powder in a wiggle-bug and oven-dried for one hour at 105° C. The nitrogen content of the adhesive is used to convert the nitrogen contents of the wood particles to adhesive solids.

All nitrogen determinations were made with a Hewlett Packard CHN analyzer at Monsanto Co. laboratory in Eugene, Oregon.

Nitrogen contents of blended fractions were adjusted for inherent wood nitrogen content determined from the unblended furnish. The mean nitrogen content for a given fraction was based on samples from four boards per treatment, each replicated twice for a total of eight determinations.

The dimensions of fifty representative particles were measured for each screen fraction of the furnish used for background nitrogen analysis. These dimensions were approximate because of the irregular shape of the particles. They were measured with a stereoscope equipped with a micrometer eyepiece. Surface areas were calculated assuming the particle was a rectangular solid with a smooth surface.