PENETRATION OF LIQUID UREA-FORMALDEHYDE ADHESIVE INTO BEECH WOOD

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ABSTRACT

The main objective of this research was to evaluate the penetration of a liquid urea-formaldehyde adhesive (UF) into beech (*Fagus sylvatica* L.) wood as influenced by moisture content (MC) and the method of curing. The maximum penetration of the UF adhesive was detected at 9% MC within the MC range of 4 to 13%. Adhesive penetration was greater with samples that were cured in a conventional press when compared with high-frequency pressing. Penetration in the tangential direction was greater than in the radial direction. The application of mechanical pressure to the bondline greatly increased penetration, whereas extended open assembly times did little to increase penetration.

Keywords: Adhesive curing, adhesive penetration, beech, *Fagus sylvatica* L., fluorescence microscopy, high-frequency curing, moisture content, urea-formaldehyde adhesive.

INTRODUCTION

Marra (1992) describes the process of adhesive bond formation in a wood substrate by five steps: flow, transfer, penetration, wetting, and solidification. The flow involves the spreading of the liquid along the external surface. The assembly of the wood elements leads to transfer of the liquid adhesive to the adjacent wood surface. Penetration then occurs as a result of capillary forces within the cell lumens and bulk flow due to applied pressure. Wetting not only applies at the external wood surface, which Marra called flow, but also aids in the movement of the liquid adhesive along the walls of the cell lumens. Finally, solidification occurs, which in the case of thermoset adhesives, signifies polymerization of the adhesive into a rigid polymer with infinite molecular weight.

While not the only factor, penetration of adhesive into the porous network of wood cells is believed to have a strong influence on bond strength (Brady and Kamke 1988; Collett 1972; Jakal 1984; Marra 1992). In this sense, penetration is defined as a spatial distance from the interface of the adjoining substrates. Marra (1992) referred to penetration as a pro-

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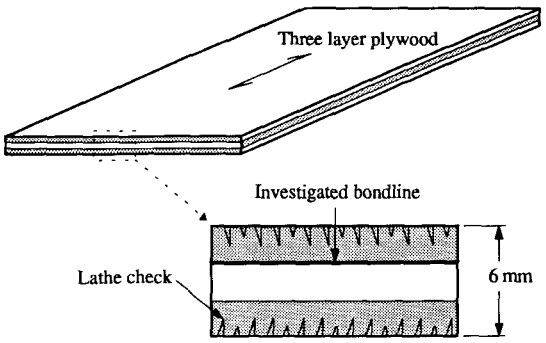

FIG. 1. Specimen manufacture at different MC and method of curing.

cess of fluid movement. The mechanism of adhesion in wood is often debated. Mechanical interlocking, covalent bonding, and secondary interactions have all been proposed as significant mechanisms (Johns 1989; Marra 1992). However, in each case adhesive penetration, and the associated intimate contact with internal surface, will play an important role.

The interphase region of the adhesive bond is defined as the volume containing both wood cells and adhesive (Brady and Kamke 1988). The size of the interphase region is determined by the depth of penetration of the adhesive. The literature is rich with qualitative discussions of adhesive penetration and its influence on bond strength. Damaged wood cells may be reinforced by the adhesive, and stresses may be more effectively disbursed within a larger interphase region. However, excessive penetration may lead to a starved bondline, with insufficient adhesive remaining at the interface (Marra 1992). The optimum depth of penetration is not known.

Fluidity of waterborne thermoset adhesives, such as urea-formaldehyde and phenol-formaldehyde, is largely dependent on the flow properties imparted by the water component. Molecular weight distribution of the resin solids, extender and filler content, and pH will also influence the fluidity of these adhesives. Any wood characteristics or processing variables that interact with water may influence adhesive penetration. The MC of wood is known to influence penetration of phenol-formaldehyde adhesive into aspen wood (Brady and Kamke 1988), Douglas-fir, and southern pine (Smith 1971). High-frequency (HF) heating to cure adhesives is sensitive to moisture content in wood (Resnik et al. 1997). The electromagnetic field energy is preferentially adsorbed by the polar water molecules, thus accelerating the temperature rise in the higher MC regions. Consequently, the method of heating during adhesive bond formation may influence the amount of penetration.

Other wood-related and process-related factors that have an influence on adhesive penetration are: direction of penetration with respect to the wood structure, permeability, porosity, roughness, surface energy, temperature, pressure, and time (Hare and Kutscha 1974; Kedzierski 1986; Marra 1992; Smith and Côté 1971; Tarkow and Southerland 1964). Few quantitative results on the influence of these factors on adhesive penetration have been reported (Brady and Kamke 1988; Furuno et al. 1983; Johnson and Kamke 1992; White et al. 1977).

This research focused on adhesive penetration and some factors that may influence pen-

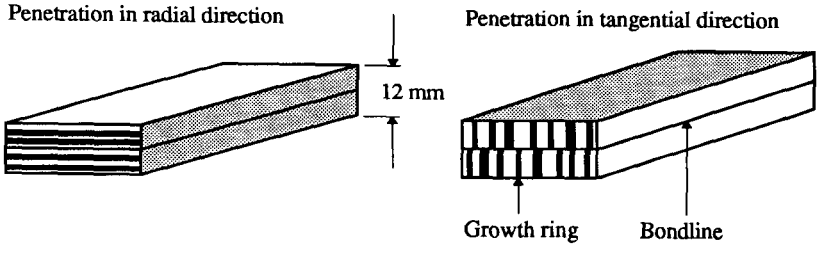

FIG. 2. Specimen manufacture for study of penetration in radial and tangential directions.

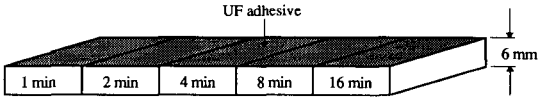

FIG. 3. Specimen manufacture for study of time-dependent penetration.

etration. The main objective of this research was to evaluate the penetration of a liquid urea-formaldehyde (UF) adhesive into beech wood at different levels of moisture content. In addition, the influence of HF heating, direction of penetration, and time on UF adhesive penetration was evaluated.

EXPERIMENTAL METHODS

The experiment was divided into three parts: I. Penetration of a liquid urea-formaldehyde adhesive into veneer at different levels of moisture content and using two methods of curing; II. Penetration of a UF adhesive in the radial and tangential directions; and III. Time-dependent penetration of UF adhesive for both the radial and tangential directions.

Specimen preparation

In Part I, three-ply, cross-laminated plywood was produced from rotary-peeled beech veneer (*Fagus sylvatica* L.). All of the veneer was obtained from the same sheet and was previously dried in a commercial dryer. The veneer had a thickness of 2.2 mm, width 200 mm, and length 400 mm. The veneer was conditioned in an environment chamber to yield five levels of MC (4, 6, 9, 11, and 13%). Before the adhesive was applied, the veneer was lightly sanded with 200-grit sandpaper and cleaned with compressed air. This surface preparation procedure was the same for all parts of the experiment. The adhesive was applied by hand with a hard-rubber roller, first on the bottom veneer and then on the top veneer. The time delay between applying the adhesive on the top veneer and the start of curing was approximately 1 min.

The plywood was pressed in a conventional press at 120°C for 4 min, or in a HF press at 6.3 MHz and approximately 120°C for 2.5

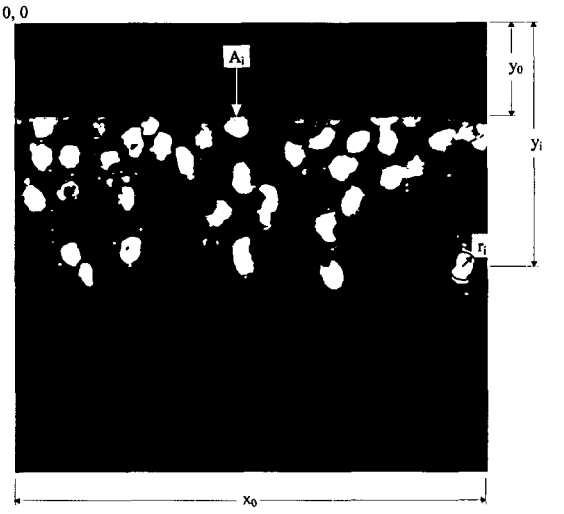

FIG. 4. Graphical explanation of effective penetration and maximum penetration. Photomicrograph illustrates penetration in the radial direction.

min. The pressure applied on the samples was 1.6 MPa (232 psi). Six replicate plywood panels were made for each of the ten treatment combinations (five MC levels and two methods of curing). Five samples (approximately 6 mm by 12 mm by 12 mm) were cut from different locations in the plywood panels for the preparation of microscope slide sections (Fig. 1).

For Part II, two-layer, parallel-laminated specimens of solid-sawn beech wood of thickness 6 mm, width 35 mm, and length 150 mm were prepared. Half of the specimens were prepared for radial penetration and the other half for tangential penetration. The wood was conditioned to an MC of 12%. The time delay between applying the adhesive on the bottom adherend to the start of curing was approximately 1 min. Four replications of two-layer samples, for both directions (Fig. 2), were pressed in a conventional press at 1.6 MPa (232 psi) and 120°C for 9 min. Four samples were taken from different locations of each specimen for preparation of microscope slide sections.

In Part III, solid-sawn beech wood specimens of thickness 6 mm, width 30 mm, and length 300 mm were prepared. The wood was

TABLE 1. *Effective penetration (μm) of UF adhesive for conventional and high-frequency cured samples.*

	Conventional curing					High-frequency curing				
MC (%)	4.1	5.7	9.2	11.0	13.1	4.1	5.7	9.2	11.0	13.1
Mean	42.9	57.1	64.2	61.0	55.4	49.9	44.4	46.7	46.5	51.7
StDev	12.6	20.0	14.0	21.7	19.0	13.7	10.5	19.7	16.1	12.8
COV (%)	29.4	35.1	21.8	35.6	34.3	27.5	23.6	42.3	34.7	24.7
n	30	30	30	30	30	30	30	30	30	30
P-value			0.0002					0.3662		

conditioned to an MC of 12%. After the adhesive was applied, each sample was cut into five sections (Fig. 3). The first section was put into an oven after 1 min, second after 2 min, third after 4 min, fourth after 8 min, and last one after 16 min. The samples were kept in the oven for 10 min at a temperature of 160°C. Four replications were made for both the radial and tangential directions. Microscope slide sections were prepared from two different locations in each specimen.

The UF adhesive mix was applied to all samples at a loading level of 200 g/m^2 for the total bondline (100 g/m^2 for each adherend). The adhesive mix contained 85% neat UF resin (LENDUR™, 66% of solids), 10% wheat flour, and 5% ammonium chloride (20% aqueous solution). The adhesive was applied by hand with a hard-rubber roller.

Preparation of microscope slide sections

All microscope slide sections were prepared using the same procedure. The samples that were obtained from each bondline were immersed in a water-filled beaker, which was placed into a sealed vessel. A vacuum was then pulled for 30 min, followed by an additional 30 min in the sealed vessel. This operation was repeated (usually twice) until the samples absorbed enough water to sink. After vacuum-pressure soaking, slide sections were cut using a sliding microtome. One 60-μm-thick section was cut from each specimen for Part I, exposing a single bondline with a radial surface adjacent to a cross-sectional surface. A 40-μm-thick section was cut from each end of each specimen for Parts II and III, exposing a bondline with a cross-sectional surface.

All microscope slide sections were set in a 0.5% Brilliant Sulphaflavine solution for 21 h and then rinsed in water. The sections were then soaked in a 0.5% Safranin O solution for 3 h. Next, the sections were rinsed several times in water until the water remained clear. This was followed by a rinse in 70% ethanol, and then 100% ethanol. Finally, the samples were fixed between a microscope slide and a cover glass using glycerin.

Measurement of adhesive penetration

The resin penetration in this experiment was measured using an epi-fluorescence microscope (Zeiss Axioskop), a 100 W HBO lamp, and digital image analysis techniques (Johnson and Kamke 1992). The optical filter set used consisted of a 365-nm excitation filter, 395-nm

TABLE 2. *Maximum penetration (μm) of UF adhesive for conventional and high-frequency cured samples.*

	Conventional curing					High-frequency curing				
MC (%)	4.1	5.7	9.2	11.0	13.1	4.1	5.7	9.2	11.0	13.1
Mean	261.9	342.5	385.8	385.2	355.9	311.0	278.1	306.4	322.2	332.2
StDev	98.9	118.7	147.9	114.1	99.2	89.5	106.7	77.3	104.6	120.2
COV (%)	37.8	34.7	38.3	29.6	27.9	28.8	38.4	25.2	32.5	36.2
n	30	30	30	30	30	30	30	30	30	30
P-value			0.0003					0.2537		

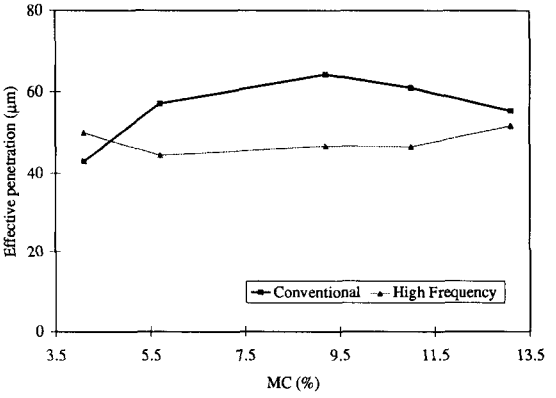

FIG. 5. Effective penetration of UF adhesive vs. MC for conventional and high-frequency cured samples.

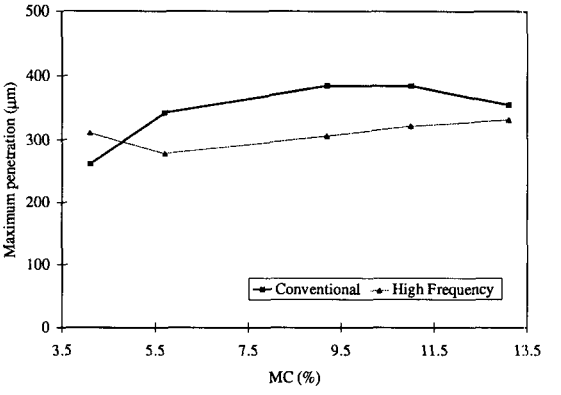

FIG. 6. Maximum penetration of UF adhesive vs. MC for conventional and high-frequency cured samples.

dichromatic mirror, and a 420-nm emission filter. The image analysis system included a Dage MTI CCD-72 black and white video camera, a Pentium 100 personal computer, image processing and analysis software (MetaMorph by Universal Imaging), framegrabber board, and a Sony Trinitron high resolution image monitor.

A random area from a single bondline was used to measure effective penetration (EP) and maximum penetration (MP). The EP is the total area of adhesive detected in the interphase region of the bondline divided by the width of the bondline. The EP excludes the cell walls and the unfilled lumen area. The MP is the average distance of penetration of the five most distant adhesive objects detected within the field of view. The resolution of the spatial area measurement was equal to one pixel (6.8 μm^2). The field of view was 1.66 mm^2. A manual process called thresholding enabled the analyst to separate the bright adhesive objects from the darker image background. Once the individual adhesive objects were highlighted, the program calculated several statistical parameters concerning the selected objects. After the individual objects were analyzed, both the EP and the MP were calculated using Eqs. (1) and (2). The measurement parameters are illustrated in Fig. 4. In Parts II and III only the EP was calculated.

$$EP = \frac{\sum_{1}^{n} A_i}{x_0} \quad (1)$$

$$MP = \frac{\sum_{1}^{5} (y_i + r_i - y_0)}{5} \quad (2)$$

where:

EP = effective penetration (μm)

A_i = area of adhesive object i (μm^2)

n = number of objects

x_0 = width of maximum rectangle defining measurement area (1297 μm)

MP = maximum penetration (μm)

y_i = centroid of adhesive object i, one of five representing the deepest penetration (μm)

r_i = mean radius of adhesive object i (μm)

y_0 = reference y-coordinate of the bondline interface (μm)

Tukey's multiple range analysis and two-sample analysis of variance (at alpha = 0.05 or 95% confidence level) were used to identify significant differences between treatments.

TABLE 3. *Effective penetration (μm) of UF adhesive in radial and tangential directions.*

	Bottom side		Top side	
Direction	Radial	Tangential	Radial	Tangential
Mean	15.3	21.1	11.6	15.7
StDev	5.3	8.6	5.2	9.5
COV (%)	34.8	40.8	44.6	60.2
n	30	30	30	30
P-value	0.0078		0.0146	

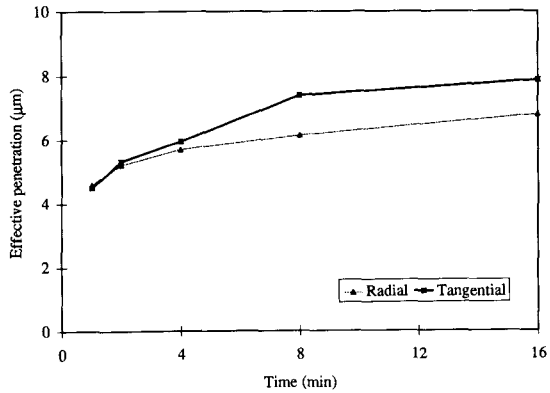

FIG. 7. Effective penetration of UF adhesive vs. time in radial and tangential direction.

RESULTS AND DISCUSSION

Influence of MC and method of curing

The results shown in Tables 1 and 2 indicate that the MC of beech veneer had a significant influence on UF adhesive penetration (EP and MP) for samples that were cured in a conventional press. Adhesive penetration was the lowest at 4% MC, then increased when MC increased. The deepest adhesive penetration was detected at 9% MC. Further increasing MC to 13% decreased adhesive penetration (Figs. 5 and 6), but the penetration was still deeper than at the 4% MC level. The nonlinear behavior is a result of two opposing factors: dry wood rapidly adsorbing water molecules from the adhesive solution; and conversely, wet wood inhibiting the reduction of resin viscosity promoting better mobility of the resin solids. Previous research had only reported a monotonic increase of adhesive penetration (phenol-formaldehyde) with increasing MC because intermediate MC levels were not investigated (Brady and Kamke 1988; Smith 1971).

No significant differences due to MC for the samples cured in the HF press were detected for either EP or MP (Tables 1 and 2). Although the MC effect was not significant in the HF-cured samples, a look at Figs. 5 and 6 would give the impression of a penetration minimum at approximately 6% MC. Apparently the relatively slow heating rate, and associated slower rate of polymerization, at the lowest MC allowed more penetration under pressure in comparison to the intermediate MC levels. Since the heating rate of wood is not proportional to MC in HF heating (Resnik et al. 1997), the 13% MC may have had a greater impact on inhibiting the gain of adhesive viscosity than the viscosity gain due to increased heating rate. Unfortunately, the sample variability masked any differences, if they exist.

Comparisons between conventional curing and HF curing showed significant differences at all MC levels for EP and MP, except at the highest MC. In general the penetration was lower for samples cured in the HF press. Conventional curing was slower than HF curing, allowing the adhesive more time to penetrate during the conventional pressing. High-frequency curing delivers heat energy almost immediately upon application of the electric field. High-frequency heating is also selective, with the heating process especially intensive in the bondline, which has a higher MC when using water-borne adhesives.

Penetration in radial and tangential directions

In Part II a two-sample analysis of variance indicated a significant difference of adhesive penetration between the radial and tangential directions. Penetration of UF adhesive in the tangential direction was greater than in the radial direction (Table 3). Beech contains thin latewood bands with fewer and smaller vessels, thus inhibiting radial penetration. Beech vessels have more pits in the radial surface

TABLE 4. *Effective penetration (μm) of UF adhesive in radial and tangential directions vs. time.*

	Radial					Tangential				
Time (min)	1	2	4	8	16	1	2	4	8	16
Mean	4.6	5.2	5.7	6.1	6.8	4.5	5.3	6.0	7.4	7.9
StDev	1.1	1.7	1.9	2.3	1.7	0.9	1.7	1.9	1.5	2.6
COV	24.8	32.3	32.9	37.2	24.9	19.3	31.1	31.7	20.2	32.9
n	12	12	12	12	12	12	12	12	12	12
P-value			0.0288					0.0001		

than in the tangential surface, which could contribute to greater penetration in the tangential direction. The air permeability of beech (*Fagus sylvatica* L.) has been measured to be nearly 90 times greater in the tangential direction than in the radial direction (Bohner 1977).

A significant difference between penetration in the bottom side (greater) and the top side of the bondline was also indicated (Table 3). This was expected because the adhesive was applied first on the bottom adherend, giving it more time to penetrate.

Time-dependent penetration

In Part III the adhesive was applied to only one surface and no pressure was applied. In this case all penetration was due to capillary flow, wetting, and diffusion. The rate of penetration was fastest at the beginning (first 4 min), then slowly decreased, and was almost negligible after 16 min (Fig. 7). There was no significant difference between penetration in the radial and tangential directions (Table 4). Note that when pressure was applied to the bondline in Part II, a significant difference was detected between the radial and tangential directions. This illustrates the importance of bulk flow to adhesive penetration. The samples made with applied pressure in Part II had 3 to 4 times the effective penetration of the samples with no applied pressure. The application of pressure greatly increases penetration and accentuates the influence of grain direction.

CONCLUSIONS

Moisture content of wood will influence adhesive penetration into beech during conventional hot-pressing. In these experiments with beech, a maximum penetration was detected with a 9% wood MC. Conversely, moisture content has little or no influence on adhesive penetration in beech during HF-pressing. Adhesive penetration is potentially greater using conventional hot-pressing in comparison to HF-pressing due to the rapid heating rate in a HF press. Penetration of UF adhesive into beech is greater in the tangential direction than the radial direction when pressure is applied. When no pressure is applied to the bondline, there are no significant differences in regard to tangential and radial directions. Most of the penetration during lay-up occurs within the first four minutes. However, the penetration during lay-up will likely be small in comparison to the bulk flow that occurs after pressure is applied to the bondline.

ACKNOWLEDGMENTS

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