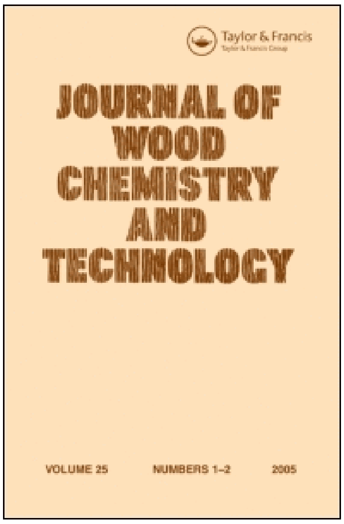

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BONDING WOOD VENEERS WITH CELLULOSE SOLVENTS

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ABSTRACT

Various solvent systems capable of dissolving cellulose have been reported in the literature. Cuene (cupriethylene diamine hydroxide) and FeTNa (ferric sodium tartrate) are well known examples. Preliminary experiments were conducted with Cuene and FeTNa to determine if the cellulose dissolving ability of these systems could be used to bond yellow birch veneers. The results indicate that relatively high dry shear strengths, and low wet shear strengths are obtained.

INTRODUCTION

Wood cellulose is a highly crystalline polymer of β-1,4-linked glucose units. It is insoluble in water and common organic solvents. However, a variety of systems capable of swelling[2] and in some cases of dissolving cellulose[3] [6] have been described in the literature. Several of these systems have been proposed as commercial methods for dissolving cellulose and then forming useful products by regenerating solid cellulose from the solution.[7] Cellulose solvents are also used to prepare cellulose solutions for routine viscosity measurements, e.g., TAPPI and ASTM standard methods.

The best known and most widely used cellulose solvents are those based on copper hydroxide[8] in aqueous solutions of either

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ammonia (cuprammonium hydroxide, often abbrev. cuam, cupram, or complex or hydrogen bonds formed between the surfaces of the two wood panels or both.

The following experiments demonstrate that it is possible to bond wood with cellulose solvents. The bulk of our experiments to determine if wood panels could be bonded with cellulose solvents were conducted with cuene, which is available commercially.

RESULTS

Table 1 shows the dry and wet shear strengths for 2-ply yellow birch panels bonded with cuene, FeTNa, and several other systems. The data presented in this table indicate that fairly high dry shear strengths can be obtained; but that wet shear strengths are low and in some cases nonexistent. No attempts were made to optimize bonding with these systems. However, the data show that the shear strengths of cuene bonded panels depend on the pressing temperature and pressing time. On pressing at 125°C and 965 kPa (140 psi), both the dry and wet shear strengths of the cuene bonded panels increased with pressing time. On pressing at 965 kPa for 5 min over the temperature range of 125° to 185°C, the dry shear strengths of cuene bonded panels continued to increase with temperature. The wet shear strength under these pressing conditions also increased initially, but then declined over the interval from 160° to 185°C.

In experiments conducted for comparative purposes, 2-ply panels bonded with ethylenediamine (EDA) (known to swell cellulose) and with ethylenediamine containing hydroxides other than cupric hydroxide (Table 1) had low to high dry shear strength but no wet shear strengths. Ethylenediamine bonded panels had low dry shear strength. Ethylenediamine in combination with ammonium hydroxide (EDA + NH_4OH) was also a poor bonding system. Ethylenediamine plus sodium hydroxide (EDA + NaOH) gave good dry shear strengths but no wet shear strength. Indeed, sodium hydroxide alone can be used to bond panels with high dry shear strength but no wet shear strength.[12] A question arose whether water, cellulose solvent, cuoxam) or ethylenediamine (cupriethylenediamine hydroxide, often abbrev. cuen, cuene, cuprien, or ced). By light absorption methods Gralen and Linderot[8] showed that in these solutions cellulose is dissolved as a cellulose-copper complex.

Jayme and Verburg[9] discovered that a ferric sodium tartrate complex in sodium hydroxide solution possessed a high dissolving power for cellulose. The term Eisen-Weinsäure-Natrium Komplex (EWNN) is used for this cellulose solvent in much of the European literature, while in most English language publications it is generally referred to as FeTNa. FeTNa is a green complex formed in solution from $Fe(OH)_3$, tartaric acid, and NaOH in the molar ratio of 1:3:6.[3] Franke[10] had described this complex prior to its use as a cellulose solvent. Bayer et al.[11] found that the molar ratio of iron to tartrate in the FeTNa complex could be varied from 1:1 to 1:4.5. They found that acetate grade cotton linters were not soluble in FeTNa at the molar ratio of 1:4.5, but were readily soluble at the molar ratio of 1:3. The 1:3 molar FeTNa complex reacts with the C-2 and C-3 hydroxyl groups of cellulose to complete the unfilled glycol-binding sites of the 1:3 complex. Thus, the fibrous structure of cellulose is first loosened and then the cellulose polymer is brought into solution as a cellulose-FeTNa complex.

Because cellulose solvents have the ability to disrupt the crystalline structure of cellulose and then to dissolve the cellulose, it was of interest to conduct preliminary experiments to determine if cellulose solvents could be used to bond wood panels. Thus, one could envision bonding taking place as the result of several steps. First, the cellulose solvent would be applied to the surface of wood panels to disrupt the crystalline structure and presumably dissolve the cellulose at the surface. Second, the panels would be pressed to bring the two surfaces into intimate contact. Third, the panel would be heated while pressing to remove the volatile components of the cellulose solvent. And fourth, bonding would occur from crosslinking caused by a cellulose-metal

TABLE 1
Shear Strengths for 2-Ply Yellow Birch Panels Bonded with Cellulose Solvents and Related Systems

Bonding agent	Pressing time	Pressing temperature	Shear strength[a]	
			Dry	Wet
	Min	°C	MPa (Psi)	MPa (Psi)
Cuene	2.5	125	4.6±0.5 (657±69)	0.3±0.3 (43±41)
Cuene	5.0	125	5.4±0.3 (766±50)[b]	0.9±0.1 (134±18)[b]
Cuene	7.5	125	8.3±1.1 (1186±150)	0.9±0.4 (129±60)
Cuene	10.0	125	12.9±2.9 (1836±419)[c]	2.0±0.4 (290±62)[c]
Cuene	5.0	160	6.6±0.6 (935±84)	1.1±0.4 (153±49)
Cuene	5.0	185	7.7±1.1 (1095±160)	0
Cuene	16.0 h	25	6.3±0.1 (890±19)	0
Cuene[d]	7.5	125	3.3±0.5 (462±77)	0
Cuene[e]	7.5	125	2.9±0.8 (406±109)	0
EDA[f]	7.5	125	0.9±1.3 (127±179)	0
EDA+NH$_4$OH[g]	7.5	125	1.3±1.9 (188±266)	0
EDA+NaOH[h]	7.5	125	7.7±2.7 (1092±378)	0
NaOH[i]	5.0	150	4.4±1.2 (631±163)	0
FeTNa[j]	7.5	125	5.6±0.9 (792±131)	0
Phenol-formaldehyde[k]	5.0	150	13.7±0.3 (1955±49)	8.3±0.3 (1182±39)

[a] Shear strength ±s.e. (standard error) in all cases is the average of four lap shear specimens from one bonded panel except as noted.

[b] Shear strength ±s.e. is the average of four lap shear specimens from each of four bonded panels.

[c] Shear strength ±s.e. is the average of four lap shear specimens from each of two bonded panels.

[d] Cuene was allowed to dry for 20 min prior to assembling panel.

[e] Cuene was allowed to dry for 60 min prior to assembling panel.

[f] EDA = ethylenediamine. 0.8 ml of EDA was spread on the surface of each veneer used to form the 2-ply panel.

[g] 0.8 ml EDA + 2.0 ml 30% NH$_4$OH was spread on the surface of each veneer used to form the 2-ply panel.

[h] 0.8 ml EDA + 6.0 ml 3N NaOH was spread on the surface of each veneer used to form the 2-ply panel.

[i] 6.0 ml 3N NaOH was spread on the surface of each veneer used to form the 2-ply panel.

[j] 6.0 ml FeTNa was spread on the surface of each veneer used to form the 2-ply panel.

[k] 5 g of a commercial resin was spread on the surface of one veneer and used to form a 2-ply panel. Pressed at 1.23 MPa (175 Psi).

heat and pressure induced thermoplastic lignin bonding. Control veneers coated with 7 ml of water or 7 ml of 2 M EDA and pressed under conditions similar to the other panels failed to bond. Therefore it is unlikely the bonds we obtained with cuene are due to thermoplastic lignin bonding.

Panels bonded with cuene after the coated surfaces of the veneers are dried at ambient temperature for 20 and 60 min prior to pressing had diminished dry shear strength as compared with panels pressed immediately after coating with cuene, and had no wet strength. This is presumably due to the fact that cellulose "dissolved" by the cuene at the wood surface did not remain dissolved as the volatile components in the cuene evaporated. Thus, these conditions limited the formation of crosslinks between the wood panels on pressing.

Birch panels bonded with FeTNa (Table 1) at 125°C and 965 kPa for 7.5 min gave dry shear strengths comparable to those for cuene panels bonded at 125°C, 965 kPa for 5 min; however, no wet shear strength was observed. Because the results with FeTNa were not significantly different from those with cuene, bonding with this cellulose solvent was not studied further.

In cuene and FeTNa bonded panels failure of all the shear specimens occurred in the bond line. It is interesting to note, however, that the dry shear strength of the best bonded specimens was equal to 95% of that obtained with a commercial phenol-formaldehyde and tested under the same conditions.

CONCLUSIONS

Bonding with the cellulose solvents cuene and FeTNa, can produce wood panels with substantial dry shear strengths and minimal wet shear strengths. Bonding presumably occurs via a cellulose-metal complex formed between the "solubilized" cellulose at the surfaces of the wood panels or via hydrogen bonds formed between the bonded surfaces or both. However, because cellulose is known to slowly degrade oxidatively in cuene solution, it is possible that bonding may be due in part to oxidative wood surface reactions of the type recently investigated by Kelley et al.[13] for wood bonding. Further studies with these and newer cellulose solvents, not based on metal complexation, should be conducted and might provide new insight into the mechanisms of wood bonding.

EXPERIMENTAL

Cupriethylenediamine Hydroxide (Cuene)

This cellulose solvent (2 M in ethylenediamine and 1 M in copper) was obtained from American Scientific and Chemical, and was used as received.

Iron Sodium Tartrate (FeTNa)

This cellulose solvent was prepared by the method of Malm et al.[14] as described by Browning.[15]

Veneer

Rotary cut yellow birch veneer (3 mm thick) was conditioned to equilibrium moisture content at 27°C and 30% relative humidity. Pieces 150 x 150 mm^2 were cut and bonded into panels.

Bonding

Approximately 7 g of the cellulose solvent or other bonding system was spread as evenly as possible with a spatula on one side of each piece. Two pieces were immediately assembled into a panel so that the grain in both plies was parallel. The panel was placed into a heated press and pressed at 965 kPa (140 psi) for various times and temperatures.

Determination of Shear Strength

Each panel was conditioned at 27°C, 30% relative humidity for approximately 1 to 2 weeks before cutting into 12 2-ply lap shear specimens. Four specimens from each panel were tested for dry shear strength using a universal testing machine at a loading rate of 1 cm/min. Another four specimens from each panel were subjected to a standard vacuum-pressure soak.[16] A vacuum of 85 kPa (25 in. of mercury) was drawn on the specimens while in water and held for

30 minutes. The vacuum was broken and a pressure of 450 to 480 kPa (65-70 psi) was applied to the specimen still in water and held for 30 minutes. The shear strength was determined on the wet specimens. The four remaining specimens were held in reserve.

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