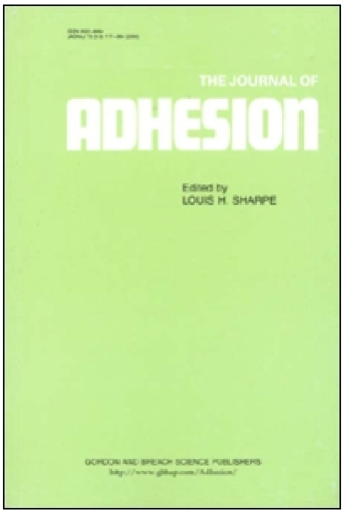

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Some Aspects of Bond Formation and Failure in Adhesive Wood Joints

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Some Aspects of Bond Formation and Failure in Adhesive Wood Joints

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An investigation has been made of the effect of varying glue-spread on the bond strength of holly (*Ilex aquifolium*) using three adhesives and three different wood sections. The glue-spreads are lower than those normally used, and it has been found with edge-grain joints that 100% cohesive wood failure can occur with a glue-spread as low as 2.7 mg/cm^2. Scanning electron microscopy shows that interlocking between adhesive and adherend does not occur. Factors leading to delamination and joint failure are discussed.

Lignin, without further addition, has been shown to be a useful wood adhesive. It has also been shown that it is possible to make end-grain joints without the use of an adhesive; the lignin present in the wood specimens is considered to be responsible for such joints.

INTRODUCTION

The quantity of adhesive used in a wood joint is important in that it influences both the bond quality and final cost of the finished product. The quantity of adhesive applied to a joint is termed the glue-spread, and is expressed as the weight of adhesive per unit area of joint. A number of investigations have been made of the effect of varying glue-spread on the strength of wood joints.[1-6] These have shown that the bond strength either increases slightly or remains constant with increasing glue-spread. Suchsland[6] found that the effect of glue-spread on shear strength was influenced by the specific gravity of the wood, with those of high specific gravity being more sensitive to varying glue-spread.

The present communication reports an investigation of the effect of varying glue-spread on the strength of adhesive joints made between specimens of holly (*Ilex aquifolium*). Holly was chosen since it is a dense hardwood with a very fine and even texture, in which the absence of distinct heartwood avoids the differences in chemical content that occur between sapwood and heartwood. Lower glue-spreads than those in the published studies[1-6] have been used, and with one adhesive (bovine plasma albumen) multilayers were deposited by the Blodgett technique[7] from a film spread on water.

EXPERIMENTAL

Apparatus

The bond testing apparatus used was a modification of the torque testing apparatus used by Holloway and Walker[8] to measure the adhesion of surface coatings under service conditions, and subsequently by Shen and Carroll[9, 10] to measure the torsion-shear strength of wood particle board. The modified apparatus consisted of a jig which held the glued wood joint tightly in place so that no other force than torsional shear was exerted during testing. The load required to break the joint was applied manually through a recording torque spanner. This type of test eliminates the possibility of the cleavage component which may occur in a standard plywood testing machine. However, it is not the usual kind of test used in assessing the bond strength of plywood or laminated wood, and the bond strength is expressed in kg/cm rather than the conventional kg/cm^2.

The hot press used consisted of aluminium platens 13 cm in diameter which were electrically heated and thermostatically controlled. The pressure was applied manually by a torque spanner through a bolt attached to the upper platen.

Materials

The wood specimens used were supplied and machined by the Forest Products Research Laboratory, Princes Risborough. They were accurately cut to 0.125 × 0.5 × 2.0 inches from a single log of holly using a bench circular saw with a fine tooth, carbide tipped blade. The surfaces were very smooth and no further preparation was required. Three types of specimen were prepared, namely, radial, tangential, and cross or transverse sections. The first two are referred to as side- or edge-grain and the third as end-grain wood. In the transverse section the growth rings ran across the width of the specimen. The specimens had an average moisture content[11] of 8.8% and a density[12] of 0.81. Extracted specimens were prepared by treating the wood slides in a soxhlet apparatus with a 2 : 1 mixture of benzene-methanol. They were then washed several times with methanol-distilled water and stored under distilled water until required for use.

The adhesives used were bovine plasma albumin (B.P.A.), lignin and Melolam 295. The B.P.A. was a Fraction V (moisture 6 %) specimen obtained from Armour Pharmaceutical Co. Ltd. The lignin was provided by the Forest Products Research Laboratory, and had been extracted from wood with methanol–HCl (moisture content 15.8 %). The Melolam 295 is a commercial adhesive from CIBA (ARL) Ltd., and is a plasticised melamine formaldehyde resin in powder form.

Methods

Two methods were used to apply adhesive to the wood specimens. The first was restricted to B.P.A. where multilayers were built up on extracted wood slides using the Langmuir-Blodgett technique.[7] A film of B.P.A. was spread on water at 20°C in a Langmuir-Adam trough and brought to a surface pressure of 10 dyne/cm. A pair of wood slides were lowered and raised through the film at a speed of 6.9 cm/min. The surface pressure was maintained during film deposition by adjusting the movable barrier, and the B.P.A. associated with the decrease in the area of the film was taken to be deposited on the wood. If a multilayer was to be built up, it was found that the wood had to be dried for a few minutes in an oven at 40°C after each cycle of dipping and withdrawal. B.P.A. could not be spread on unextracted wood in this way due to contaminants from the wood spreading on the water. The second method was by spreading adhesive solution directly and as evenly as possible on the wood slides from an Agla micrometer syringe. The technique of double spread was used. The solvents used were distilled water for B.P.A., chloroform-methanol (85 : 15 parts by volume) for lignin and ethanol-distilled water (65 : 35 parts by volume) for Melolam 295. To minimize differences in the amount of solvent applied to the wood the volume of solution was maintained between 0.2 and 0.3 ml, whereas the solution strength varied between 0.005 and 8.6 % weight/volume. Following adhesive spreading the wood slides were assembled to form joints. With wet wood slides a closed assembly time of 10–15 minutes was followed by hot pressing at 95°C and 110 kg/cm^2 for 30 minutes. With unextracted, dry wood slides the open assembly time was sufficient for the solvent to evaporate or diffuse into the wood, and this was followed by hot pressing at 140°C and 110 kg/cm^2 for 20 minutes. The joints were stored at room temperature for 24 hours before testing.

RESULTS AND DISCUSSION

Wet wood

When B.P.A. was applied either by the dipping technique or by direct spreading on wet wood, no bond was formed with side-grain wood sections with glue-spreads up to 1.4 mg/cm^2. This is probably due to the very high moisture content of the wood (~80%) which washes the adhesive away from the glue line during pressing. However, with end-grain wood a bond was formed, the strength of which was independent of glue-spread up to 1.4 mg/cm^2 with an average value of 7.76 ± 0.24 kg/cm. It is evident that something inherent in the wood, rather than the adhesive, is responsible for joint formation. Blank experiments in which slides of wet end-grain wood without adhesive were hot pressed gave comparable bond strengths of 7.85 ± 0.35 kg/cm. No bond was formed in blank experiments with two side-grain wood slides or a combination of side-grain and end-grain wood slides. It may be argued that the bond formed was due to the mechanical interlocking of the fibres in end-grain wood under the applied pressure. However, no bond was formed when two end-grain dry wood slides were pressed at 550 kg/cm^2 and 20°C.

A significant difference between end-grain and side-grain wood is the much higher concentration of cut cells exposed in the end-grain wood. The cell walls, together with the middle lamella between the cells, are the locations of the lignin content of wood.[13] It has been reported[14] that the softening temperature of lignin ranges over 127–193°C, hemicellulose 167–181°C and cellulose >230°C, and that the sorption of water by lignin and hemicellulose causes a pronounced decrease in their softening temperatures. Moreover, the softening points of lignin and hemicellulose correlate with the temperatures at which they develop adhesive properties.[14] Thus with wet wood slides pressed at 95°C the substance responsible for bond formation is most probably lignin. The flow of lignin from the wood to the "glue-line" would be favoured by end-grain wood, and presumably does not occur in effective amounts with side-grain wood.

Dry wood

The effect of varying glue-spread on bond strength was determined for B.P.A. spread on radial and transverse section wood, lignin spread on radial, tangential and transverse section wood, and Melolam 295 spread on radial and transverse section wood. For each adhesive-wood section system graphs of bond strength against log (glue-spread) were constructed; Figure 1 shows the graph obtained for B.P.A. spread on transverse section wood. The individual bond strengths can be considered to lie within a band of values, and the mid-points of the bands have been used to construct the collected plot of bond strength against glue-spread shown for all seven systems in Figure 2. With B.P.A. and lignin, which could be spread as monolayers on water, the gluespreads can be expressed as an "equivalent number of layers of adhesive" from the ratio of the glue-spread to the weight of a single layer of adhesive taken up by a wet wood slide from a Langmuir trough. Clearly this approach is approximate since it assumes that the film deposited directly from the syringe has the same structure as that deposited from the water. For B.P.A. a glue-spread of 1.0 mg/cm^2 corresponds to 1.1×10^3 monolayers, whereas for lignin a glue-spread of 1.0 mg/cm^2 corresponds to 5.1×10^2 monolayers.

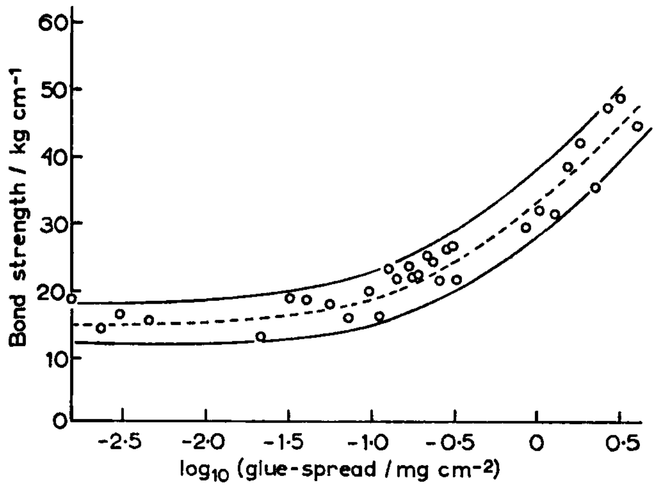

FIGURE 1 Plot of bond strength against $\log_{10}$(glue-spread) for transverse section joints glued with B.P.A.

Since wood is a porous material the penetration of some of the adhesive into the wood structure can be expected. An examination of both tested (broken) and untested joints with a scanning electron microscope suggests that adhesive penetration is minimal. For example, the structure of the unspread surface of radial section wood (Plate 1) is closely similar to that from a tested joint which had failed at the adhesive/adherend interface (Plate 2). Similar effects were observed with tangential and transverse section wood. Plates 3 to 5 show sections cut by a microtome from joints which had not been tested, and it can be seen that the wood cavities, such as cell lumen, wood rays and pores, adjacent to the glue-line are virtually "empty". The glue-spread used for the joints shown in Plates 3–5 was about 4.0 mg/cm^2, which was the maximum used in the present work. These results indicate that mechanical interlocking between glue and wood is not important in the present work, and thus support accepted opinion that interlocking is not necessary in wood adhesion.[2, 15, 16] Adhesive penetration can be deleterious, since it can lead to lower bond strengths through the formation of starved joints.[16, 17]

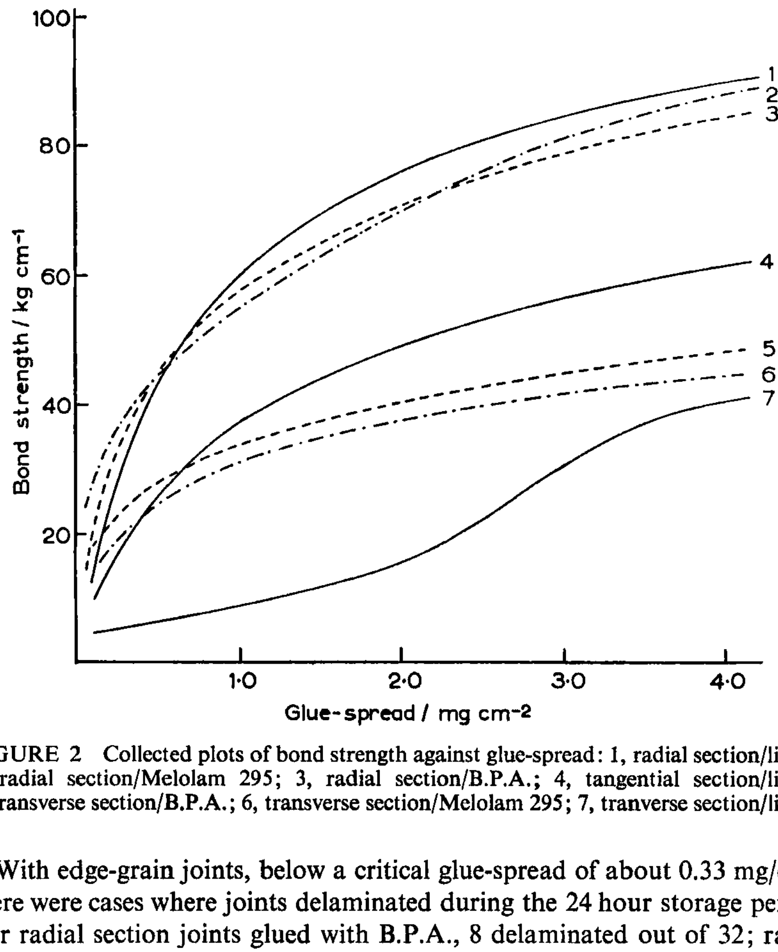

FIGURE 2 Collected plots of bond strength against glue-spread: 1, radial section/lignin; 2, radial section/Melolam 295; 3, radial section/B.P.A.; 4, tangential section/lignin; 5, transverse section/B.P.A.; 6, transverse section/Melolam 295; 7, tranverse section/lignin.

With edge-grain joints, below a critical glue-spread of about 0.33 mg/cm^2, there were cases where joints delaminated during the 24 hour storage period. For radial section joints glued with B.P.A., 8 delaminated out of 32; radial section joints with lignin, 14 delaminated out of 45; tangential section joints with lignin, 7 delaminated out of 26; radial section joints with Melolam 295, 8 delaminated out of 24. Delamination is probably due to stresses set up between the glue and the wood as the joint cools after hot pressing. With glue-spreads >0.33 mg/cm^2 the glue-line may be visualised as in Figure 3a, with the half joints being kept apart by the asperities of the second-degree texture[18] of the wood. With such glue-spreads there are flaws or voids in the glue-line at which cracks will propagate with relative ease. With glue-spreads >0.33 mg/cm^2, Figures 3b and 3c, the macroscopic valleys of the second degree texture are filled with adhesive, so that delamination through crack propagation does not occur. This need to fill the second degree texture with adhesive, and the insensitivity of delamination to the adhesive used,

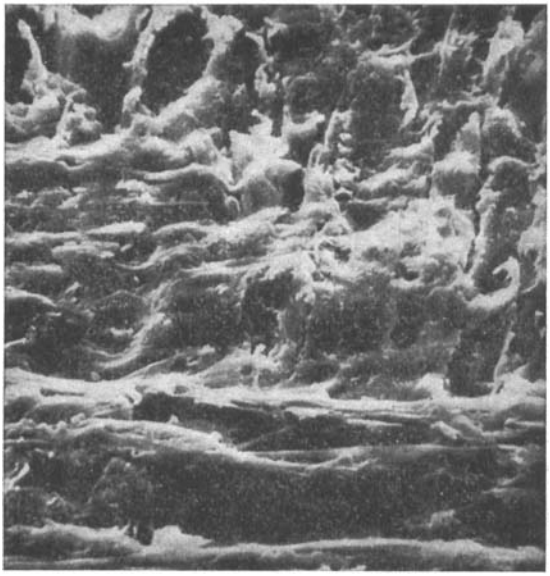

PLATE 1 Surface of radial section wood.

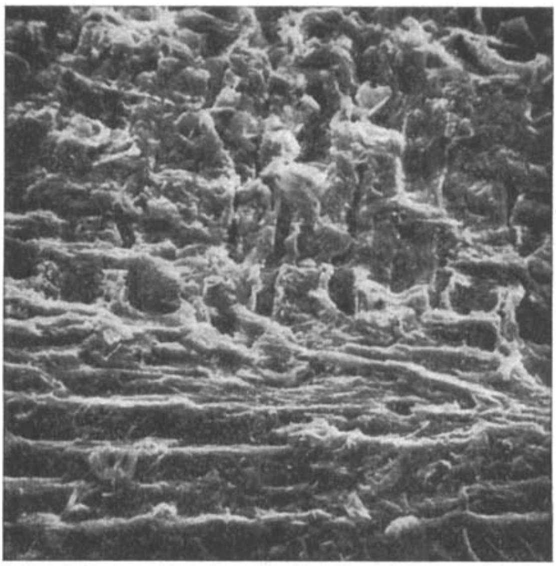

PLATE 2 Surface of radial section wood at the same magnification following adhesive/adherend failure of a joint spread with 4.0 mg/cm^2 of Melolam 295.

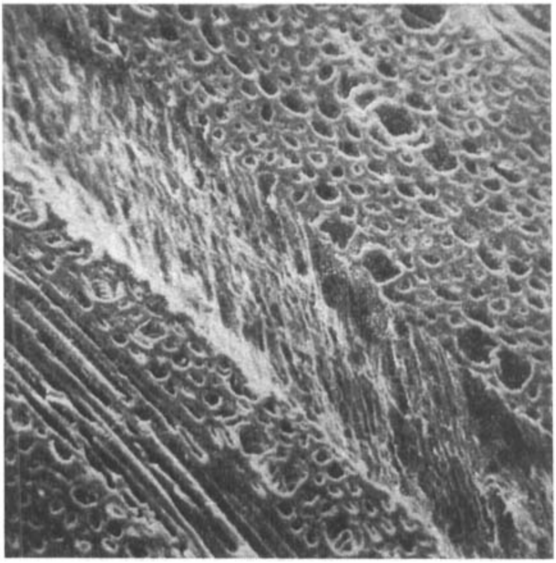

PLATE 3 Joint in radial section wood with Melolam 295, showing "empty" cell cavities and pores adjacent to the glue-line.

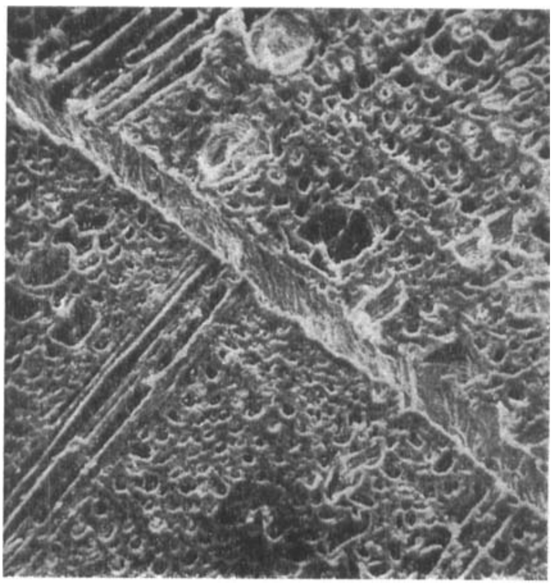

PLATE 4 Joint in tangential section wood with Melolam 295, showing "empty" pores and wood rays perpendicular and adjacent to the glue-line.

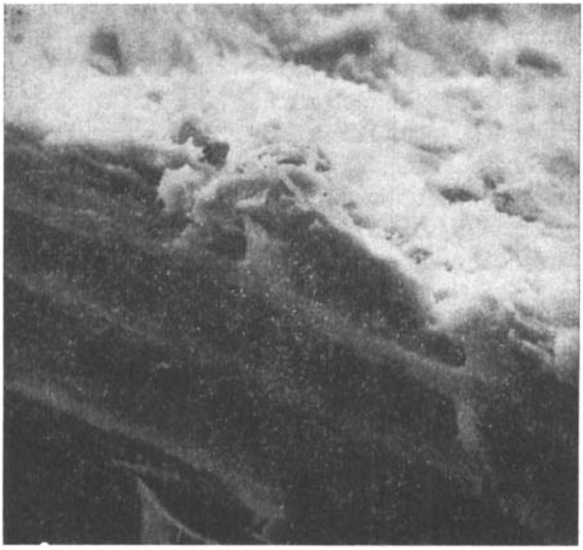

PLATE 5 Joint in radial section wood with Melolam 295, showing wood rays parallel and adjacent to the glue-line.
The magnification in Plate 5 is five times greater than that in Plates 3 and 4.

suggest that the concept of an equivalent number of adhesive layers is not applicable to a heat cured joint.

With edge-grain joints made with B.P.A. cohesive wood failure started at a glue-spread of ~ 1.0 mg/cm^2 and was complete at a glue-spread of ~ 2.7 mg/cm^2, although there were isolated cases of $<100\%$ wood failure at glue-spreads >2.7 mg/cm^2. With Melolam 295 or lignin cohesive wood failure started at a much higher glue-spread of ~ 3.2 mg/cm^2. The extent of wood failure with Melolam 295 did not reach 100% at the highest glue-spreads studied, whereas with lignin only small extents of wood failure were observed. The differences in the type of failure observed can be ascribed to differences in the molecular weight of the three adhesives. With lignin and Melolam 295 there is the possibility of lower molecular weight material of plasticiser diffusing into the wood and thus producing a "composite material". This process may increase the strength of the surface layer of the wood,[16] so that a lower percentage of wood failure occurred. With B.P.A., its high molecular weight (6.7×10^4) would make such a process unlikely. A different mode of wood failure was observed with end-grain joints; the surfaces possessed a "fuzzy" appearance and the wood slides were broken into pieces. The fuzzy appearance most probably arises from the withdrawal of fibres from the opposing wood slide. Superimposed upon the effects of the adhesive on the mode of joint failure will be the differences in the strength of the wood from one test specimen to another. Such variations are inevitable in a biological material such as wood.

Figure 2 shows that for every adhesive-wood section system studied the bond strength increased with glue-spread. With glue-spreads >0.33 mg/cm^2 we can assume that the second degree texture no longer plays a part in determining bond strength. Presumably the strength of any one joint is determined by both the strength of the adhesive/wood bond and the cohesive strength of the wood, together with the extent to which either of these factors is influenced by the glue-spread. It can be seen, however, that there are differences in the

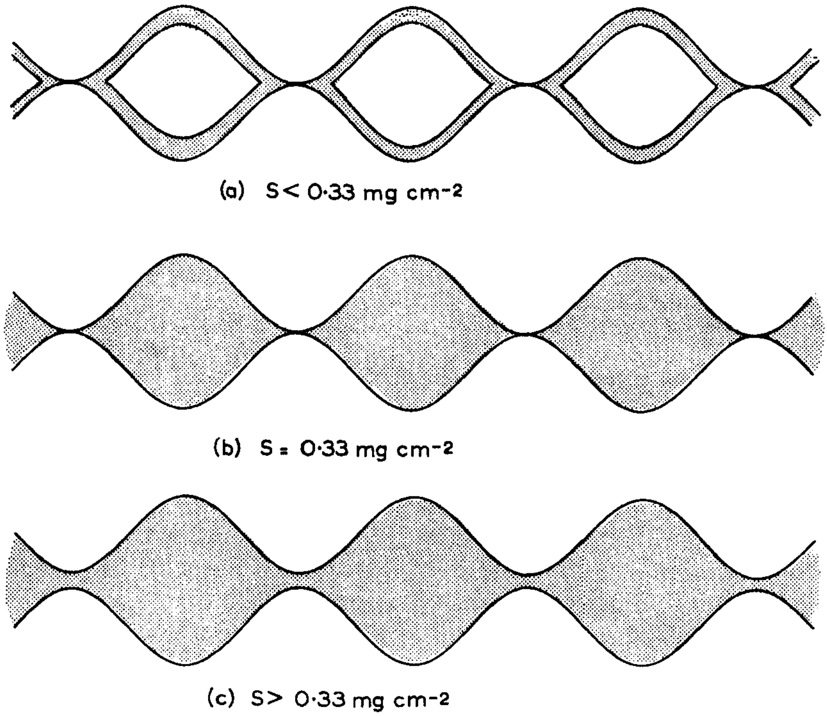

FIGURE 3 Illustrations of the glue-line at different glue-spreads: a, glue-spread $<$ 0.33 mg/cm^2; b, glue-spread $=$ 0.33 mg/cm^2; c, glue-spread $>$0.33 mg/cm^2.

bond strength arising from the type of wood section used which outweigh those arising from the type of adhesive used. For high glue-spreads the radial section gave the highest bond strength, followed in decreasing order by the tangential and transverse sections. These differences may be qualitatively explained in terms of a strength-reducing factor inherent in the wood, which is the area of cell cavities on the surface of the wood sections. The cavities will have the effect of decreasing the area of contact, thus giving rise to a lower bond strength per unit area of glue-line. Alternatively, they may act as centres for crack propagation. Taking the radial section wood as a base, the weaker joints with tangential section wood arise from the cavities represented by the cross sections of the wood rays. This is followed by transverse section wood, where the cell lumen and pores give rise to the highest area of cavities and the weakest bond strength.

With transverse section wood, joints at low and intermediate glue-spreads are weaker with lignin than with B.P.A. and Melolam 295. Experiments were made in which only 0.2–0.3 ml of solvent was spread on transverse section wood slides before hot pressing. Control joints in which no solvent was spread were also made. The bond strengths were: chloroform-methanol, no bond formed; control, 7.4 ± 1.3 kg/cm; water, 14.3 ± 0.5 kg/cm; water-ethanol, 18.2 ± 1.9 kg/cm. Since lignin is responsible for the bond in the control joint, chloroform-methanol prevents bond formation by extracting lignin from wood near to the glue-line and carrying it deeper into the two half joints. It is probable that the same process occurs, but to a lesser extent, when lignin dissolved in chloroform-methanol is used to form joints. The higher bond strengths obtained with water and water-ethanol over that of the control are in accord with the observation[14] that sorbed water acts as a low molecular weight diluent in plasticising the lignin polymer, lowering its glass transition temperature, and increasing its adhesive behaviour.

The objective of any wood adhesion project is to make the adhesive joint as strong as the wood itself. In the present work it has been shown that this can be achieved in holly with a glue-spread of B.P.A. as low as 2.7 mg/cm^2. It is fortunate that B.P.A. would not be immune to bacterial attack. Lignin, which is recoverable from spent sulphite liquors in paper manufacture, offers a good possibility of being used as a wood adhesive. It has been demonstrated that the lignin content of the wood itself can be used to form end-grain joints. If further developed this might find application in glued laminated wood where short pieces are butt-end bonded before they are laminated together.

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