Wood properties influence bond durability

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ABSTRACT

Wood bond performance is generally evaluated at the macroscopic level for strength and durability. Although this process has been very effective in making bonded wood products that perform well commercially, these tests do not always provide enough fundamental information for developing new and improved adhesives. This level of examination also leaves open the question why some adhesive-wood combinations are more effective than others. A holistic model that includes the properties of both wood and adhesive, and adhesive-wood interactions at the cellular and sub-cellular levels can explain the crucial factors for good bond durability. The performance of epoxy adhesives is used as an example of the utility of this holistic model for explaining adhesive-wood combination performance.

1. INTRODUCTION

Adhesively bonded wood products continue to encompass an increasing share of the wood products market. Given the wide variety of wood product types, the number of different production processes, the many adhesives available, and the multitude of wood species used, the difficulty in understanding the fundamentals of adhesive performance is not surprising. Because wood products are expected to last decades, if not centuries, resistance to deformation and delamination are important attributes. To resist adhesive deformation, the adhesives need to cross-link (cure) during the bonding process (Frihart 2013, Pizzi and Mittal 2003). Consequently, having the adhesives cure after application allows an adhesive that is easy to apply and flow over the rough wood surfaces, but then cure to resist flow.

The resistance to delamination is not as easily solved, especially when the bond is subjected to high and/or changing moisture levels. Why is moisture durability testing so hard on adhesive bondlines? It is well known that wood swells and shrinks as the moisture level changes, but adhesives swell and shrink less being more hydrophobic than wood and cross-linked polymers. This difference in size change with moisture leads to strains at the adhesive-wood interface, see Figure 1a (Frihart 2005). If the wood swelling is great enough and the resulting strain is not dissipated, then the stress at the interface can be sufficient to fracture the bond (Figure 2). To help alleviate this stress, adhesives could infiltrate the wood cell walls near the surface to pre-swell the wood and interact with wood cell wall polymers on a nano- or molecular level. This bulking of the cell wall free volume, along with any reaction of these adhesives, can reduce the water uptake. This process lessens the swelling, resulting in reduced interfacial strain; see Figure 1b (Frihart 2006a). Alternatively, the adhesive can have some localized ductility to reduce the strain concentration at the interface. Although these adhesives are lightly cross-linked to eliminate bulk creep, the flexible backbone allows small domains to move with the wood and relieving the strain (Figure 1c).

Do adhesives exist that can use either of these two modes for reducing the interfacial strain? Interestingly, wood adhesives can be divided into two classes based upon not only how they provide durable bonds, but also their morphology and curing mechanisms (Frihart 2009). One class, *in situ* polymerized, consists of rigid oligomers that polymerize and

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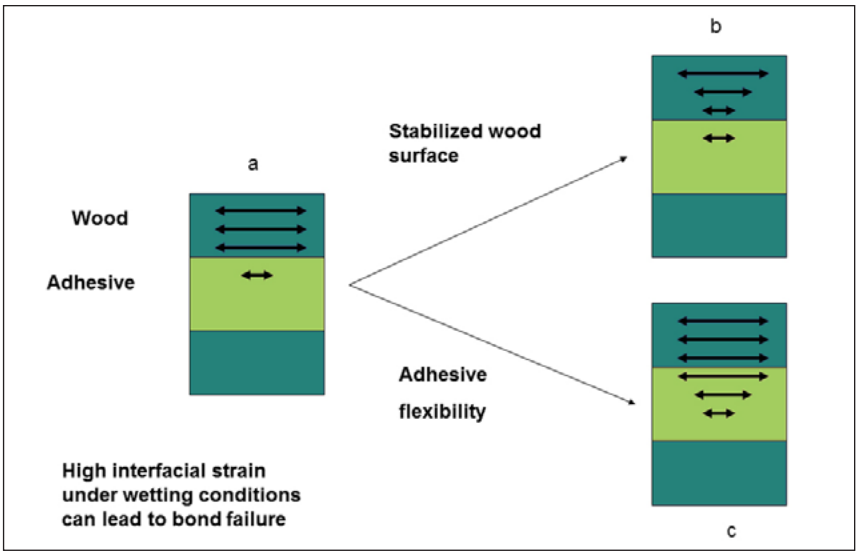

Figure 1
Models of adhesive bond strain alleviation. When a bonded wood specimen swells at higher moisture levels, strain develops at the interface; distributing the strain within the wood or adhesive reduces the probability of failure near the interface.

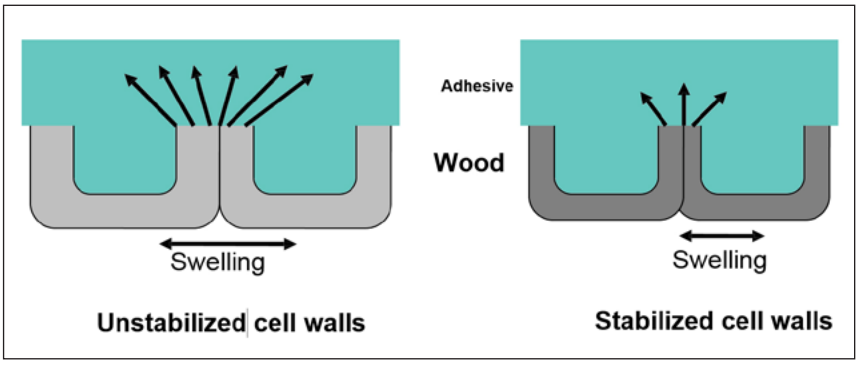

Figure 2
Swelling stress at the cell wall can generate microcracks in the adhesive that can be sufficient to cause failure in the adhesive. However, bulking of the wood can reduce the degree of swelling and the stress on the adhesive.

cross-link during the bonding process, enhancing their rigidity. The rigid nature of these polymers (including amino resins, phenolics, diisocyanates, and epoxies) would be expected to have poor bond durability as the wood swells and shrinks. However, many of these adhesives are the most durable wood adhesives. The reason is that many of these oligomers contain low molecular weight fractions that are compatible with the cell wall polymers so that they can infiltrate the cell wall and act as pre-swelling agents to minimize the swelling when later exposed to water as in model b in Figure 1 and in Figure 2. Thus, for adhesives like the phenolic and amino adhesives, the proper formulation can provide moisture durable bonds by infiltrating the cell wall (Nearn 1974, Frihart 2009). While the *in situ* polymerized adhesives can have fractions that infiltrate the cell wall and stabilize it towards swelling changes, the other class of wood adhesives, pre-polymerized (e. g. poly(vinyl acetate), polyurethane), are too large in molecular weight to infiltrate the cell wall. However, they have a very different morphology, consisting of a flexible backbone and a light degree of cross-linking. The cross-linking is generally not enough to prevent localized flexing to accommodate the swelling of the wood. Thus, the pre-polymerized case fits with model c in Figure 1.

Understanding what the adhesive needs to do to accommodate the swelling and shrinking of the wood is useful for explaining the performance of different adhesives in service. Epoxies have been used for a number of wood bonding applications from wooden airplanes to the repair of cracks and decayed portions of glued and solid timbers. The poor durability of wood bonds with epoxy adhesives is surprising, given that they are generally good adhesives and coatings in a wide variety of applications and provide durable bonds to a range of substrates, but not to wood. This paper explains why epoxy adhesives exhibit poor durability in wood bonding and under what conditions durable epoxy bonds can be made.

2. EXPERIMENTAL

2.1. WOOD SPECIES EFFECT STUDY

The five wood species used in this study were: white oak (*Quercus* spp., white group), sugar maple (*Acer saccharum),* southern yellow pine (*Pinus* spp.), Sitka spruce (*Picea sitchensis*), and aspen (*Populus tremuloides*) bonded with FPL 1A. The lumber was purchased from several local distributors; sapwood was used for all species, except the white oak. FPL 1A epoxy adhesive used in the wood bonding involved first mixing 100 parts D.E.R. 331 epoxy resin (Dow Chemical Company, Midland, MI) with 12.5 parts of benzyl alcohol (Aldrich Chemical Company,

Milwaukee, WI), and 2.50 parts of CAB-O-SIL® N70-TS hydrophobic fumed silica (Cabot Corporation, Boston, MA). After thoroughly mixing these materials, 11.10 parts of D.E.H. 24 hardener (Dow Chemical, Midland, MI) were well mixed with the epoxy resin.

The methods for wood selection, bonding, and testing for the ASTM D 2559 type tests were previously reported (Christiansen 2005). For the ASTM D 905 test, all wood strips were conditioned at 27°C and 65% relative humidity until bonded. Specimens were prepared by laminating two strips of wood, 0.6.4 cm thick, 3.2 cm wide and 22.9 cm long. FPL 1A epoxy was spread at an approximate rate of 320 to 340 g/m^2 on both surfaces with a rubber-roll hand spreader. Pressure for the epoxy was measured by squeeze out, generally about 10 psi. After removing material about 3 mm from all sides and ends, four block-shear specimens with a shear area of 2.54 cm by 2.54 cm were cut from each joint assembly to form shear blocks similar except for size differences as described in ASTM D-905-03 (ASTM 2007a) and randomly assigned to either the ambient (~10% EMC, equilibrium moisture content), wet, or wet/ambient (re-equilibration to ~ 10% EMC) shear tests. Eight specimens for each wood species were subjected to a single vacuum pressure soak (VPS) of 85 kPa vacuum for 5 minutes followed by 518 kPa pressure for one hour, and then tested for shear strength and wood failure while in the water-saturated condition. The wet/ambient specimens were subjected to the VPS procedure and then were re-dried at 27°C and 65% relative humidity until their weight reached equilibrium, typically 3 to 5 days. Ambient and wet specimens were tested in a compression-loading shearing tool as described in ASTM D 905. The maximum load at failure was recorded, and then shear strength was computed for each specimen based on the nominal shear area. Wood failure was estimated according to ASTM D 5266-99, Standard Practice for Estimating the Percentage of Wood Failure in Adhesive Bonded Joints (ASTM 2007b). The wet-tested specimens were air-dried before estimating wood failure.

2.2. ACETYLATED WOOD STUDY

Yellow-poplar (*Liriodendron tulipifera*) sapwood lumber, free of defects, was sawn into strips 6.4 or 7.9 mm thick, 3.18 cm wide and 22.9 cm long. After cutting, the strips were placed in an oven and dried at 105 °C for 24 hours. The strips were removed from the oven, cooled in a desiccator for 1 hour, and weighed.

For acetylation, the strips were placed in a glass reactor fitted with a reflux condenser. The reactor was filled with enough acetic anhydride to cover the strips even after absorption of the anhydride. The acetic anhydride and wood were heated to boiling for 4 hours and then cooled. Strips were removed, washed for 4 hours in reverse osmosis water to remove acetic acid and excess acetic anhydride, air dried overnight, and then oven dried for 24 hours at 105 °C. Weight gain due to acetylation was determined after oven drying by calculation as a percent of the original oven-dried weight. The acetylated wood strips had an average of 21.7 + 0.9 weight percent gain.

All strips, including the untreated controls, were conditioned at 27°C and 65% relative humidity until bonded. If the strips were to be planed they were 7.9 mm thick and were planed after treatment and before bonding. If the strips were not to be planed after acetylation, they were planed to be 6.4mm thick prior to treatment. Specimens were prepared by laminating two strips of wood, bonding them, and dry and wet testing them as in section 2.1, except that a commercial epoxy 305 (Lord Corporation, Erie, PA) was used.

3. RESULTS AND DISCUSSION

Despite being durable adhesives for metals and other materials, epoxies are not considered to be moisture durable for manufacturing structural wood products (AITC 1994). Formulation of epoxies has not been able to completely solve the moisture problem, although some formulations were better than others (Olson and Blomquist 1962, Lavisci et al. 2003). Surprisingly, one case showed that epoxy bonds can be very durable in both exterior exposure and severe accelerated aging (Caster 1980). A latter report indicated that for this study, the wood had been primed with a polyethylenimine, when a wood hydroxymethylated resorcinol (HMR) primer showed very durable epoxy bonds to wood (Vick et al. 1996). Their explanation was that the HMR served as a coupling agent to bond the epoxy to the wood.

The reason for the general poor moisture durability of epoxy bonded wood products had not been clear until we examined where and why failure occurred in these products. From our examination, the failure was in the epoxy layer near the wood surface (Frihart 2006b, Beecher and Frihart 2006). This is not terribly surprising since the epoxy flows into the lumens due to its low surface energy compared to the typical wood adhesive that is water-borne. Given that

Table 1: Properties of wood species used in these studies, with R (radial), T (tangential) and V (volumetric) shrink (shrinkage upon drying, SG (specific gravity), shear ‖ (shear parallel to grain, MOE (modulus of elasticity), and growth ring pattern.

Species	R shrink (%)	T shrink (%)	V shrink (%)	SG	Shear ‖ (KPa)	MOE (MPa)	Growth rings
White oak	4.4	8.8	12.7	0.68	13800	12300	Ring porous
Sugar maple	4.8	9.9	14.7	0.63	16100	12600	Diffuse porous
Aspen	3.3	7.9	11.8	0.39	7400	9900	Diffuse porous
Sitka spruce	4.3	7.5	11.5	0.36	6700	9900	Gradual transition
Southern Yellow Pine	4.8	7.4	12.3	0.51	9600	12300	Abrupt transition

the epoxy fails in the adhesive layer near the wood surface, but not at the epoxy-adhesive interface, contradicts the concept of HMR as a coupling agent.

3.1. WOOD SPECIES EFFECT

What was causing the epoxy to fail near the wood surface? One clear contributor is that wood swells (3-10% in the radial and tangential direction) more than the epoxies (~1%); thus, creating high interfacial strain on the bond near the surface (Figure 1a). Given that the cured epoxy morphology is a rigid backbone and is highly cross-linked puts epoxies into the *in situ* polymerizable category. The hypothesis for this category is that durable bonds occur if the wood swelling near the surface is lessened. One way to lessen swelling is if components in the adhesive or a primer go into the cell wall to stabilize it, but another way is to use wood species that swell less. Not all wood species swell to the same degree; more dense wood species with greater cell wall mass swell more than do less dense wood species. Thus, it was interesting to see how well epoxies bonded to a variety of wood species with different structures and densities. For this study, we used five wood species; white oak, sugar maple, aspen, Sitka spruce, and southern yellow pine, whose properties are given in Table 1. We also used the FPL-1A epoxy that gave some of the best bond strengths under wet conditions (Olson and Blomquist 1962).

Figure 3, shows that the epoxy formed a good bond (10 MPa or greater) with all the wood species when tested dry (21 °C and 50% relative humidity for a 10% equilibrium moisture content (EMC)), but this is common with most adhesives. As pointed out earlier, water soaking of the samples puts a lot of internal strain on the bondline that results in significant stress on the adhesive. This soaking results in a reduction of the strength of the bonded assembly in all cases (4-8 MPa) with the larger drop for the species with the greatest swelling. It is well known that as wood absorbs water the strength decreases, but the strength drop of the bonded assembly for the higher density species was greater than the strength drop of the lower density woods as shown in Figure 3. The lower wood failure under wet conditions provides additional information (Figure 4). The wood failure did not change much for the weaker and less dense wood species supporting the concept that water is not interfering with adhesion to the wood. Thus, good wet bonds were observed for aspen and Sitka spruce where the failure is occurring predominately in the wood. For white oak, maple and

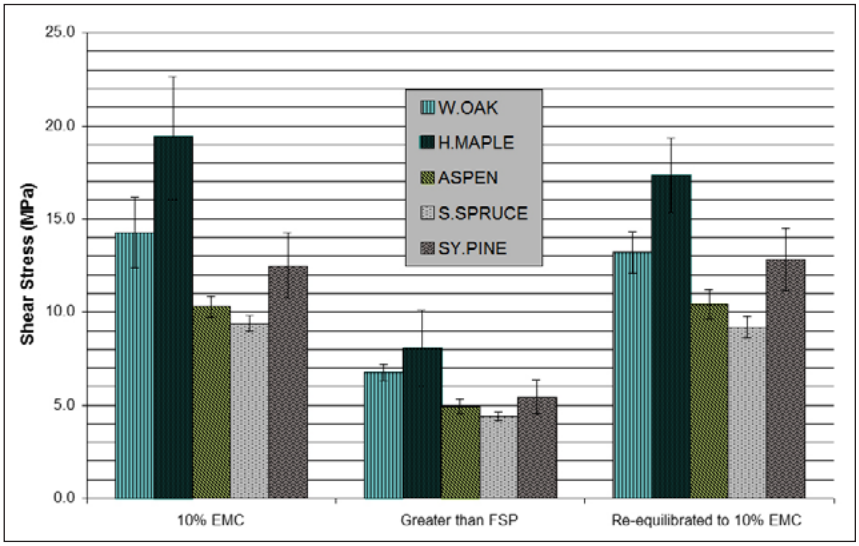

Figure 3
Strength determined using ASTM D 905 testing for different wood species with FPL 1A epoxy and tested for ambient (10% EMC), vacuum pressure soak (>FSP), and vacuum pressure soak followed by re-equilibration to 10% EMC (wet/ambient) specimens. The error bars are ± two standard deviations around the average value.

SYP, although the wood is becoming weak upon soaking, the main failure mode switches to the bondline.

When the water soaked samples are allowed to dry under ambient conditions, the strength and wood failure would be unlikely to recover to near the original levels if the water swelling cycle caused adhesion failure. On the other hand, if the lower values were mainly related to an internal stress on the bondline; then drying should return the samples to the near original strength and wood failure. This was observed in our experiments (Figures 3 and 4), and also with a polyurethane adhesive bonding European beech (Klausler et al. 2014). The data in Figure 3 supports this internal swelling strain model as being a good explanation of the poor moisture resistance of epoxy adhesives. The lack of adhesion failure is also supported by microscopy in Figure 5 that shows failure in the epoxy near the wood surface (Frihart 2006b).

Although the strength data provide useful information for dry and wet strength, a more severe test is a series of vacuum/pressure water soaks and oven drying. In Table 2, the data for the same wood species bonded with the same epoxy in the cyclic delamination test show that for the white oak, sugar maple, and southern yellow pine ASTM D-2559 bond specimens gave an unacceptable level of delamination, while the delamination was low for the aspen and Sitka spruce. Thus, the performance of adhesive can depend as much upon the wood species as on the adhesive formulation itself.

3.2. ACETYLATED WOOD

Another means to reduce the swelling strain is to modify the wood so it does not swell as much.

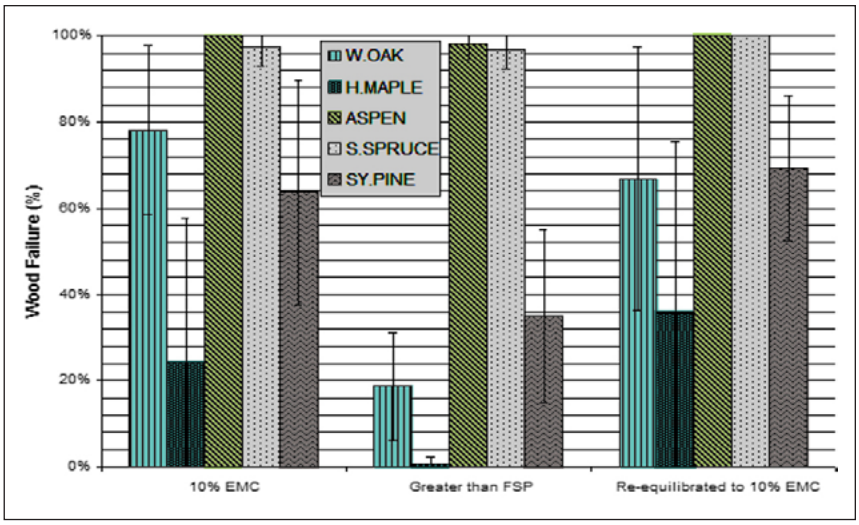

Figure 4
Percentage of wood failure for FPL 1A epoxy as determined using ASTM D 905 and ASTM D 5266 testing for different wood species and tested for ambient (10% EMC), vacuum pressure soak (>FSP), and vacuum pressure soak followed by re-equilibration to 10% EMC (wet/ambient) specimens. The error bars are ± two standard deviations around the average value.

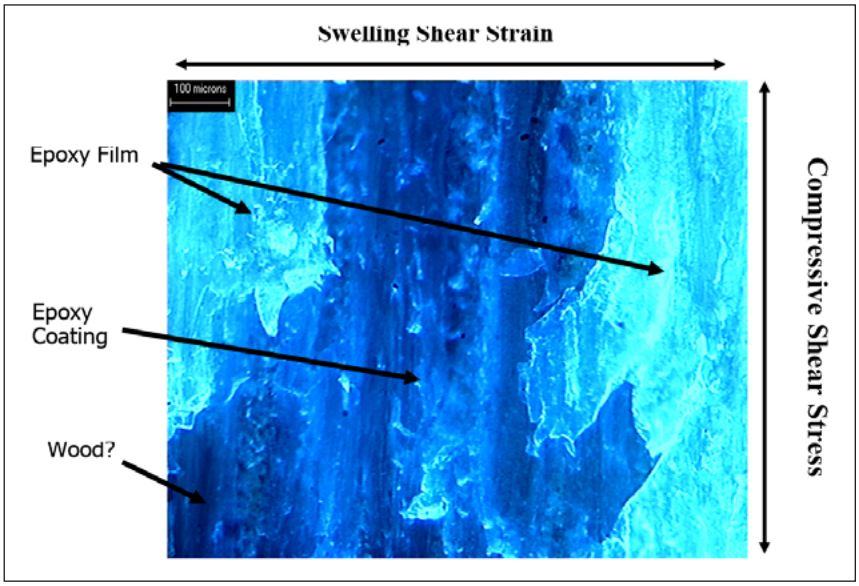

Figure 5
Failure of an epoxy bond looking at the failure surface, showing different failure locations, with the bondline. Swelling stress develops from the wood absorbing water, and compressive shear is the externally applied force.

Table 2: Percent Delamination of D2559 samples bonded with epoxy

Wood species	Delamination (%)	
	Assembly 1	Assembly 2
White oak	80	85
Sugar maple	80	85
Aspen	10	15
Sitka spruce	0	0
Southern yellow pine	70	50

Table 3: Dry and wet shear (D 905) of unacetylated wood and acetylated wood, both unplaned and planed

Wood	Dry Shear		Wet shear	
	Strength, MPa	% Wood failure	Strength, MPa	% Wood failure
Unacetylated	12.45	90	2.98	0
Acetylated, planed	11.07	30	4.47	0
Acetylated, unplaned	11.65	100	9.77	60

A common way to reduce the swelling of wood is to modify it by acetylation, which typically reduces the swelling by 40%. (Norimoto 2001) This type of modification has two modes that limit the amount of swelling. The first is that the conversion of hydroxyl groups to acetyl groups reduces the propensity of wood to absorb water; the second is that the modification bulks up the wood cell wall leaving less free void space for absorbing water. Thus, the reduced swelling of the acetylated wood is expected to impart less internal strain on the bondline, reducing bond failure even though the wood surface has fewer polar groups for interacting with the adhesive. To examine the effect of having some exposed hydroxyl groups, some of the acetylated blocks were made slightly thicker and then planed. This created an acetylated sample that has a surface with free hydroxyl groups exposed from areas that resisted acetylation. To be consistent with an earlier study on examining a variety of adhesives with acetylated wood (Vick and Rowell 1990), we used a commercial epoxy resin with yellow poplar (*Liriodendron tulipifera*).

The data in Table 3 show that the unacetylated wood provided good strength and wood failure under dry conditions, but the water soaking led to zero wood failure and low bond strength. In contrast, the unplaned acetylated samples with a fully acetylated surface not only had good shear strength and high wood failure under dry conditions, but also had the best strength and low wood failure under wet conditions. Planing of the acetylated wood caused the wood to have a drop in wet wood strength with a surface hydroxyl content between the unacetylated and the acetylated unplaned wood. The level of free hydroxyls at the surface was probed by determining the amount of trifluroacetic anhydride that reacted on the different surfaces. These values were 0.13 for poplar, 0.054 for planed acetylated poplar, and 0.011 for unplaned acetylated poplar expressed as [CF3]/[Σ carbon] (Beecher 2005)

These results with acetylated wood support our model that adhesive failure is not mainly due to a plasticization by water of the adhesive during the water soaking or a disruption of adhesion, since the bond withstands higher applied loads for the acetylated wood case. The data show that there is likely lower internal stress for the acetylated versus unacetylated wood, and the lower wood swelling of the acetylated wood leads to stronger bonds under wet conditions. However planning of the acetylated wood apparently led to increased swelling of the interface and a decrease in strength and wood failure.

CONCLUSIONS

Epoxy adhesives bring value to the wood-bonding field by having a room temperature cure and a low clamping pressure. Although they are used to repair interior glulam timbers, they are not generally recommended for glulam products due to their inability to pass accelerated aging tests. Given their durability in bonding other substrates, this lack of moisture durability for epoxy-wood bonds is surprising. Our research has shown that the failure with wet wood is in the epoxy near the wood surface. The proposal that the epoxy cannot handle the interfacial strain caused by the wood swelling is supported by the epoxy being more durable for wood species that swell less than for those that swell more. Additional support is provided by the greater moisture durability in epoxy bonding lower swelling acetylated wood than with unacetylated wood.

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