Are Epoxy-Wood Bonds Durable Enough?

Charles R. Frihart
Project Leader, Wood Adhesives Science & Technology, USDA Forest Service, Forest Products Laboratory, Madison, Wisconsin, USA

Abstract

An important aspect of any adhesive bond is that the bond maintains its integrity during its end use. Epoxies form highly durable bonds with many substrates but are usually not considered capable of forming completely durable bonds with wood by standard accelerated tests. However, epoxies are sold for wood boat construction, and some data have indicated that epoxies can have sufficient exterior durability for general use in wood products. This conflicting information raises an important question. Are the reported discrepancies caused by changes in epoxy–wood bond formulations or in the use of different durability test methods? Examination of these data has led to an interfacial strain model which provides the most compelling explanation for the durability observations of epoxy–wood bonds. The results of bond failure analysis, coupled with the ability of epoxies to bond acetylated wood and give bond durability to wood primed with hydroxymethylated resorcinol or melamine-based primers add support to this model in explaining epoxy–wood bond durability results.

Introduction

Epoxies are minor wood adhesives, but serve an important role because of their room temperature cure, gap-filling ability, and low clamping pressure. This allows them to be used in both new construction and repair of existing structures. Our interest in epoxies is their usefulness in understanding the durability of wood bonds based upon the following observations:

- Epoxies provide very durable bonds to metals, cement, and some plastics.
- Durability of epoxy–wood bonds has been dependent upon the specific epoxy used as well as test conditions.
- Epoxies come in a wide range of formulations and the properties of one can differ drastically from another.
- Epoxies are chemically different from wood so it is fairly easy to characterize their location and composition.

Despite concerns about durability, epoxies have long been used for wood bonding in certain applications, such as aircraft, boats, and repair of wood structures. Epoxies are widely used in bonding parts of motor vehicles and aircraft made of metal and plastics because of their durability. They also form durable coatings for concrete. Thus, the explanation as to why epoxies fail in many of the durability tests for wood bonds is not readily apparent.

Epoxies have the widest formulation and cureability of any adhesives (14). Epoxies can be formulated to cure rapidly (45 s) at room temperature or to be stable at room temperature, for high-temperature cure systems. The curing agents or hardeners can be amines, thiols, alcohols, anhydrides, Lewis acids, organic acids, or Lewis bases. Some formulations are one component and cure using elevated temperatures or radiation, but most are two-component systems. The only common adhesive cure mechanisms not used for commercial epoxies are water-induced, free-radical, or anaerobic cures.

Given the wide use of epoxy adhesives in other applications requiring durability, why are they limited in their use for wood bonding? For one, they are expensive compared with commonly used wood adhesives, such as urea-formaldehyde (UF) and phenol-formaldehyde (PF) resins. When compared with other high-cost adhesives, for example, poly(vinyl acetate) and polymeric diphenylmethane diisocyanate (pMDI), epoxies generally are used at greater weights per bonded surface. The epoxy-bonded wood generally suffers from a limited ability to deal with moisture changes. For structural applications, epoxies are prone to creep. Epoxies bonded at ambient conditions tend not to completely crosslink as evidenced by low glass transition temperatures (50° to 65°C). Thus, epoxies have generally been limited to wood bonding markets where the other adhesives do not perform well, such as wood repair and room temperature bonding with low bonding pressures.

Epoxy Durability Studies

Epoxies are generally classified as adhesives not suitable for exterior wood bonds (1). Epoxies have not been able to pass ASTM D 2559 (4) and water soaked ASTM D 905 (2) type tests (21). Given that the bulk of the literature shows epoxies lack good exterior durability, our emphasis has been to concentrate on those reports that come to the opposite conclusion about the durability of epoxies.

In one of the early papers on epoxies, Olson and Blomquist examined various epoxies using several different test conditions (18). They found large performance differences for 15 commercial epoxies tested using water soak or boiling water test conditions on 3-ply, yellow birch plywood. Of the 23 Forest Products Laboratory (FPL) formulations tested, the FPL 16 (bisphenol A epoxy with diethylenetriamine hardener, titanium dioxide filler, and lacquer thinner) was the best. It and two commercial formulations performed well in 120-h boil, 4-h boiling water-dry-boiling water cycle, and 48-h water soak-dry cycle tests. This formulation was modified, marketed privately as FPL 16A, and was a popular adhesive for aircraft applications (17). Despite these promising results, FPL 16A, as well as FPL 1A and commercial epoxy, yielded high delamination levels on D 2559 testing with yellow birch, yellow-poplar, Douglas-fir, and Sitka spruce (21).

Several interesting studies are the basis for accelerated durability tests of adhesive-wood bonds. One study used three adhesives – an epoxy, a phenolic, and an aminoplastic resin – in bonding plywood specimens. The samples were analyzed using the automated boil test for bond durability that is now the cycle test in ASTM D 3434 (5); this study showed the epoxy was only slightly poorer in durability than the phenolic plywood adhesive (13). In another study, 11 adhesives were tested using both exterior exposure (up to 16 years) and the automated boil (up to 800 cycles) for Douglas-fir plywood. The results showed that one of the most durable adhesives was the epoxy, which even outperformed phenol-resorcinol-formaldehyde (PRF) and PF adhesives (6). Although not explicitly mentioned in the study, it has been reported that the specimens tested were actually pretreated with polyethylenimine primer (23).

How can these results be rationalized? Certainly from the work of Olson and Blomquist (18), we know that the epoxy formulation has an effect on bond durability. However, this effect is not sufficient for explaining all of the data, especially in comparing their good plywood results with the poor performance in laminate studies that used FPL 16A and yellow birch (21). An important factor for plywood durability is that the cross-ply can limit the swelling of the wood. Laminated beams do not possess this advantage and swelling is much less restricted. Additionally, the plywood veneer often cracks (possibly from lathe checks) to relieve swelling stress under water soaking. The laminates in glulam are less likely to crack in a normal direction to the bondline. If swelling stresses are less concentrated on the bondline, the epoxy may have sufficient strength to hold together.

Clearly the failure or success of epoxy bonds cannot be fully understood simply from a review of the earlier literature. However, some recent studies are helping us better understand the general lack of durability of epoxy–wood bonds. These studies are discussed in the next section, as they have been crucial to the development of the interfacial strain model, which proposes that failure occurs when the strain from wood swelling exceeds the ability of the interphase region to dissipate this force through the wood or adhesive. For clarity, the discussion of these studies will be arranged according to research areas rather than the chronology of the research.

Recent Studies

Most of the studies of epoxy bonds have reported strength data, and in some cases percent wood failure, but have not concentrated upon determining the failure location within the bondline. Failure is generally classified as percent wood versus bondline failure (3). On the other hand, bond failure has been classified into several zones by Marra (15) and this scheme has been further evaluated (10). The failure zones of Marra are bulk wood, wood interphase, wood–adhesive interface, adhesive interphase, and bulk adhesive. The interface is the sharp transition from wood to adhesive, while the interphase zones are those adjacent to the interface, whose properties differ from the bulk adhesive or wood (10).

For epoxies, lack of bond strength occurs mainly upon water exposure. In examining bondline failure for epoxy adhesives from both ASTM D 2559 and D 905 (wet), failure was often in the epoxy interphase layer (8). This failure in the epoxy interphase region was highly irregular because of the cellular structure of the wood surface. The failure surface has a highly anisotropic nature like the wood itself as shown in **Figure 1**. Closer examination of the failure surface using fluorescent microscopy showed the roughness of the fracture surface and a thin epoxy layer covering most of "wood" surface, as illustrated in **Figure 2**. In addition to the fluorescent microscopy, infrared, and x-ray electron spectroscopy, light and scanning electron microscopy were used to show that even though the failed bondline appeared optically like a wood surface, it was mainly covered by epoxy (8). If we consider the difference in the expansion coefficient between the cured epoxy and the wood during the wetting of the sample, we

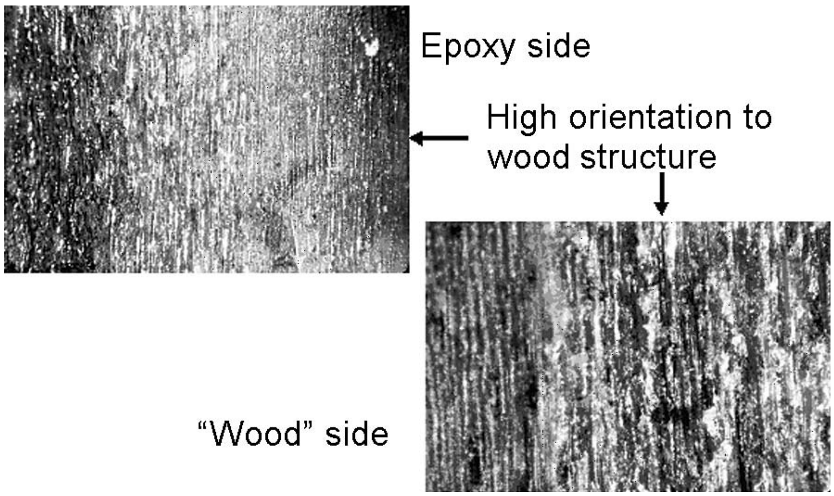

Figure 1. ~ *Optical microscopy of the failure surface from D 905 testing of a commercial epoxy on wood to illustrate the high orientation on the side mainly covered in epoxy and the side that appears to be bare wood but is not.*

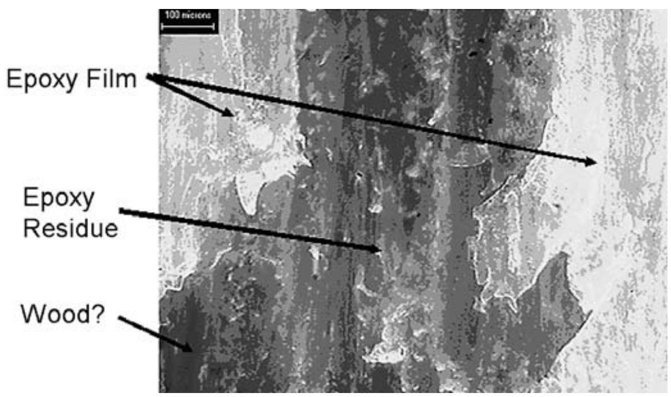

Figure 2. ~ *Use of fluorescent microscopy showing a specimen similar to that in Figure 1, where the fluorescent epoxy shows up as the lighter color and the non-fluorescent (dark) areas are mainly wood.*

can explain the failure of the bond. As the wood becomes wet, its natural tendency is to swell. The epoxy bondline absorbs less water and hence does not swell to the same degree. The stresses in this disparate swelling, therefore, build up and ultimately concentrate on the rigid bondline. This strain can exceed the strength of the epoxy with the forces being the most intense near the epoxy–wood interface. This force gives a fracture surface a wood-like texture, although the data show it is generally covered with epoxy. Further support for this model is provided by the studies discussed below.

One way of solving the poor durability of epoxy wood bonds is to first prime the wood with hydroxymethyl resorcinol (HMR) (22). As shown by **Figure 3**, this primer was found to dramatically improve the epoxy bonds to Sitka spruce, Douglas-fir, yellow birch, and yellow-poplar (21). In most cases, HMR priming of the wood allowed ASTM D 2559 laminates bonded with epoxy to pass the soak-and-dry cycle test with minimal delamination com-

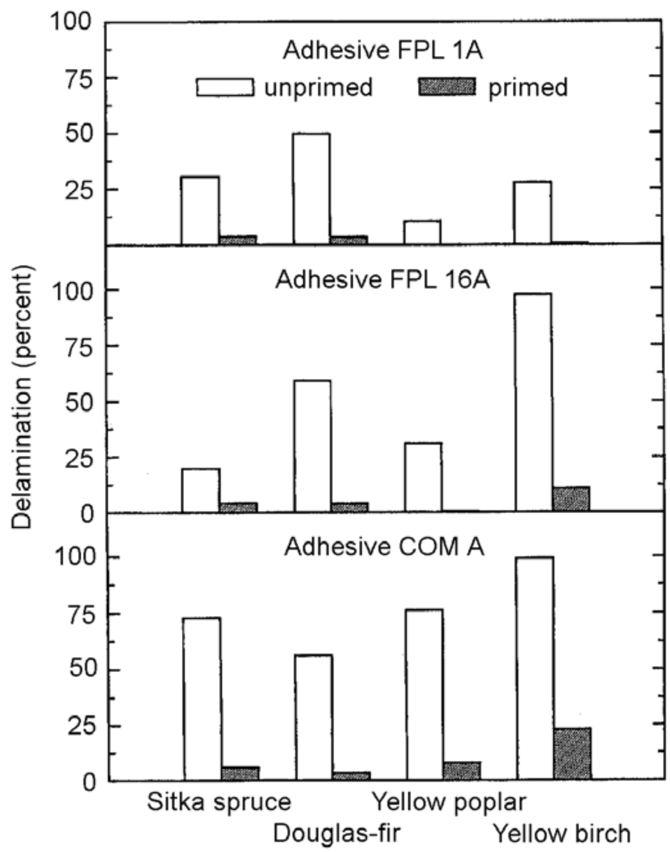

Figure 3. ~ *Reduced delamination using the ASTM D 2559 delamination cycles for different wood species treated with HMR and bonded with epoxies (21).*

pared with the unprimed samples. The original explanation was that the HMR served as a chemical coupling agent between the wood and the epoxy, but stabilization of the wood surface by the HMR is now the more generally accepted model (11). HMR-treated hard maple veneer has been shown to have 65 to 75 percent lower weight gain and about 80 percent reduction in swelling of wood during the water soak than an untreated veneer (20). Although these data do not prove that the HMR primer provides a more durable epoxy bond because of decreased strain at the interface, it does give strong support to the concept.

Further support of the interfacial stress model can be found in the studies using melamine-containing primers. Hexamethylolmelamine methyl ether (MME) with yellow-poplar has been shown to be a good primer for epoxy bonding. The wood failure of epoxy bonds in samples tested using ASTM D 905 increased from about 10 to 100 percent as shown in **Table 1** (7). MME has also been shown to reduce the swelling of wood (16) and increase the hardness of wood (16) and wood cell walls (12). Furthermore, a low molecular weight melamine-urea-formaldehyde (MUF) primer was also shown to have a similar effect, improving the percent wood failure under wet shear conditions (7).

Bonding to acetylated wood has also been shown to improve the strength of epoxy–wood bonds. A general model of wood adhesion states that hydrogen bonding between the adhesive and the wood is an important aspect of the adhesion process. This model would suggest that acetylation should lower the bond strength by replacing the strong hydrogen-bonding hydroxyl groups with weaker hydrogen-bonding acetate groups. In support of this, percent wood failure drops with some adhesives in comparing the unmodified wood to highly acetylated wood. However, Frihart et al. found in compressive shear D 905 tests that epoxies gave higher percentage wood failure with the acetylated wood (60%) than with the unmodified wood (0%) (9). These data conflict with what one would expect according to standard adhesion theory. One possible explanation is that the bond to the acetylated wood has less internal stress than the unmodified wood because of the lower swelling of the acetylated wood.

Table 1. ~ *Forest Products Laboratory (FPL) 1A epoxy bonded samples of yellow-poplar tested using compressive shear blocks for wood primed with either a low molecular weight melamine-urea-formaldehyde (MUF) resin catalyzed with p-phenol sulfonic acid in water and a 1:1 molar ratio of acid to morpholine or hexamethylolmelamine methyl ether (MME catalyzed with p-toluene sulfonic acid (7).*

		Tested dry		Tested wet	
Priming chemical	Level	Wood failure	Strength	Wood failure	Strength
		(%)	(MPa)	(%)	(MPa)
None	Control	85	14.21	3	5.07
MUF primer	5%	99	14.68	15	5.54
MUF primer	4%	92	13.33	6	5.48
MUF primer	3%	100	15.38	93	5.78
MUF primer	2%	98	14.99	100	5.37
MME primer	2%	100	17.20	99	7.20

It seems that the greater swelling of wood compared with that of the adhesive during water soaks should create high internal strain at the interface. Demonstrating this internal strain is hard to do; however, one experiment which supports the idea has been done using the ASTM D 905 test. Yelle and Frihart bonded white oak, hard maple, southern yellow pine, Sitka spruce, and yellow-poplar with the standard Forest Products Laboratory (FPL) 1A adhesive (21). Not only were these blocks tested in the standard dry and water-soaked compressive shear, but some of the water-soaked blocks were allowed to dry back to the original moisture levels at ambient conditions and then tested in compressive shear. The data in **Figures 4 and 5** show that the water soaking lowered both the measured shear strength and percentage wood failure, but upon re-drying most of these properties were recovered. The lower percentage wood failure under wet conditions

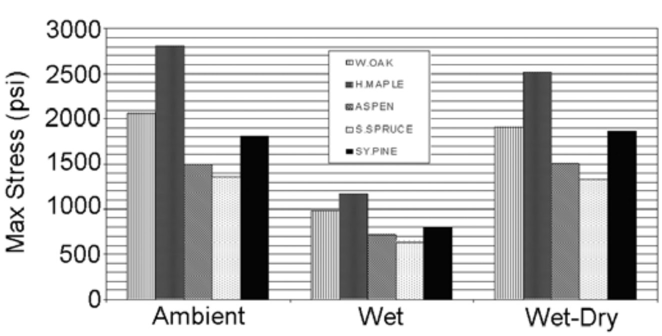

Figure 4. ~ *Strength determined using ASTM D 905 testing for different wood species and tested for ambient, vacuum pressure soak (wet), and vacuum pressure soak followed by ambient drying (wet-dry) samples. Gradual redrying of D 905 specimen allows recovery of strength.*

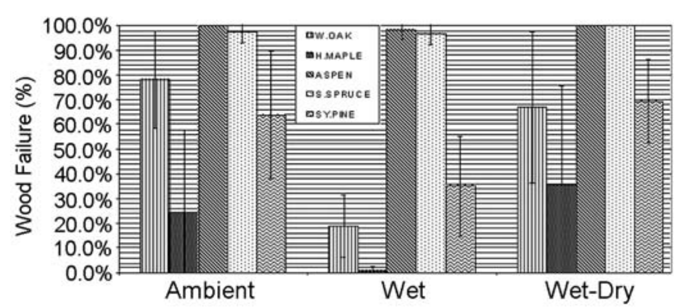

Figure 5. ~ *Percentage wood failure as determined using ASTM D 905 and ASTM D 5266 testing for different wood species and tested for ambient, vacuum pressure soak (wet), and vacuum pressure soak followed by ambient drying (wet-dry) samples.*

indicated that the bond properties dropped more than wood strength. This is likely caused by the internal bondline strain from the difference in swelling of the wood and the adhesive. The recovery of properties upon re-drying indicates that the effect of the water was not a permanent change in the bond, but a temporary effect of the water on the bond.

While none of these experiments alone shows that the difference in dimensional change between the adhesive and the wood causes the lower bond durability, each of these experiments point to this differential expansion as being a critical issue. Thus, studying an adhesive that is sensitive to test conditions can provide insight into what is critical for the formation of durable bonds.

What is Necessary for Durability?

The two main factors which contribute to durable wood bonds are the lack of creep and the ability to withstand the strain resulting from moisture changes in the wood. While not true for all applications, wood bonds generally need to bear some type of load. Crosslinking of the adhesive polymer chains is the most common way for adhesives to maintain their original shape. This crosslinking is not without its problems. The main one is that the adhesive's rigidity can often prevent the adhesive from adjusting to the dimensional changes in wood as it swells and shrinks.

Chemical bonds will fracture under sufficient concentrated force. Although precise measurements are difficult to make, the force exerted by the swelling of wood has been determined to be in the hundreds to thousands of pounds per square inch depending upon the measurement technique (19).

To avoid stress concentration at the interface, the forces need to distribute through the adhesive, the wood, or both. It has been proposed that the formaldehyde adhesives penetrate, modify, and stabilize the interfacial wood cell walls and distribute the expansion/contraction differences more evenly into the wood cellular structure (10). Crosslinked poly(vinyl acetate) and emulsion polymer isocyanate are generally too high in molecular weight to enter into cell walls but have enough flexibility to distribute the stress through the adhesive. The epoxy does not appear to stabilize wood surfaces and is too crosslinked to distribute the stress through the adhesive.

Future Studies

The analysis of the literature from a number of studies has led to the proposal of the interfacial strain as being an important aspect of bond durability. When the strain can be reduced by modification of the wood surface via the use of resorcinol- or melamine-based primers or acetylation of the wood, the epoxy bonds are more durable. It is important that further research be aimed at determining the correctness of the interfacial strain model versus other models for bond durability. The following paragraphs provide some suggestions for ways of examining wood durability models.

Undoubtedly, wood changes dimensionally with changes in moisture level. However, more research needs to be done to examine how adhesives and primers alter the ability of wood to change dimensionally as the moisture level varies. In addition, we need to understand how these dimensional changes contribute to the internal forces upon the bond.

If stabilization of wood surfaces leads to more durable bonds, what processes do this most efficiently? HMR priming is an easy process to perform and is reasonable in cost because the resorcinol concentration is low. Do easier, more economical ways exist to stabilize wood surfaces toward dimensional changes? To advance to the next stage, it is important to better understand which chemicals enter cell walls and stabilize them. The available literature supports the cell wall stabilization ability for PF adhesives, HMR primer, and melamine primers, but the literature is not available to show the cell wall stabilization of other adhesives, such as UF, epoxies, and pMDIs.

The ability of this interfacial strain model to explain many reported studies of epoxy-bonded wood has helped in understanding the seemingly conflicting data in the literature. However, techniques other than standard bonding experiments will be needed to validate this model. There is a need for more swelling data on adhesive–wood combinations, better microscopic techniques, and cell wall nanoindentation to help advance our understanding of adhesive–wood interactions.

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Forest Products Society
2801 Marshall Court
Madison, WI 53705-2295
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