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Adhesive Bonding and Performance Testing of Bonded Wood Products

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ABSTRACT: Despite the importance of durable wood bonds, the factors that lead to durability are not well understood, and the internal forces exerted upon the bondline are often overlooked. Durability requires that the bonded assembly resist dimensional changes of wood with fluctuation of wood moisture levels. Both bonding and bond breaking steps need to be understood at cellular and nanoscale, in addition to the larger spatial scales normally examined. With both internal and external forces being significant, interphase and bulk adhesive properties need to be better understood. Systematic studies of the bonding process, the forces upon the bondline, and the locus of failure using different types of adhesives and wood species should improve our ability to design wood adhesives. Modifications of wood surfaces, along with spectroscopic and microscopic analyses, are important tools to understand bond formation and failure.

KEYWORDS: wood, bond formation, bond failure, cellular, microscopy

Introduction

Wood adhesives date back several millennia, and research on wood-adhesive interactions has been ongoing for over 75 years [1]. The past century has seen tremendous advances in adhesive chemistry, comprehension of the adhesion process, and knowledge on aspects that lead to failure in durability testing. For many applications, the critical aspects of bond formation that lead to durability have been well defined. However, the critical chemical and physical properties that lead to durable bonds have not been as well defined for wood adhesives as they have for metal and plastic adhesives. This discussion is not intended to ignore the excellent work that has been done in this field but rather to define where more work needs to be done.

Why are we still unable to define the chemistry and physical properties that are necessary to lead to a successful adhesive bond for a specific application? In reality, wood has more complex chemical, structural, and mechanical properties than most other substrates. This paper is aimed at addressing some of these issues.

The chemistry of wood adhesives has been studied extensively, mainly related to the initial reaction and polymerization stages, and is known well enough to predict the results of alteration in the chemistry [2]. The preparation of wood surfaces also has been studied, and optimum conditions have been determined [3]. Numerous studies have been carried out on the durability of wood bonds using both natural and accelerated aging [4]. Among the less well understood areas are adhesive interactions with wood surfaces, wood-adhesive interphase physical and mechanical properties, and failure zone for many wood bonds. Although some excellent studies have been done in these areas, knowledge is still insufficient to predict the performance of a new adhesive or different wood species, resulting in mainly a trial and error process. A better

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understanding in these areas can aid in solving current adhesive problems, developing new adhesives, and providing new uses for wood adhesives. In general, bonding of wood is not difficult for specimens that are not under high continuous load or at high or varying moisture levels. Some factors that lead to durability of wood bonds have been discussed [3–5], but the understanding diminishes rapidly as the spatial scale being examined decreases from millimeter to micrometer (cellular) to nanometer (cellulose, hemicellulose, lignin domains) [6].

Wood failure is often considered to be as important as the strength of the bond. Deep wood failure is easy to observe, but determining where and why failure takes place in the bondline has been difficult. This paper presents ideas on how a better understanding of the failure of wood bonds can be obtained.

Experimental

Wood for these tests was obtained from local suppliers, with the actual test specimens selected according to the protocol in ASTM D 905 [7]. Wood species used were aspen, hard maple, Sitka spruce, southern yellow pine, and white oak. The wood was selected and prepared according to this method, bonded using FPL-1A [8] at a spread rate of 0.34 kg/m^2 and clamped for a day at room temperature at a pressure sufficient to cause a light squeeze out.

Specimens for microscopy were obtained from the bonded specimens and from the samples after the D 905 tests. For transverse sections, samples were water-soaked for 2–24 h prior to microtoming. The sections were analyzed using a scanning electron microscope (JEOL 840 after gold plating the samples) or a Leitz Orthoplan epi-fluorescence microscope with a 150-W mercury lamp light source, an A2 UV filter cube, and a Nikon DS-5M digital camera, or a Carl Zeiss Axioskop epi-fluorescence microscope with a 100-W mercury lamp, a UV filter set, and CCD camera. Fluorescence microscopy was used to examine the failure surface.

Bonding

High bond strength and durability depend upon developing excellent adhesive-wood interaction and good dissipation of internal and external forces under end use conditions. Wood adhesion models have generally been based upon general adhesion models, which concentrate on surface interactions between the adhesive and the adherend. These general models work well for most adherends but need to be modified when wood is the substrate. Factors causing these modifications include adhesive penetration into the wood, high wood surface roughness, the multi-polymer composition of wood, and wood variability. These factors do not displace the importance of primary or secondary bonds between the wood and the adhesive used in normal adhesion theory but can be additional mechanisms that can either increase or decrease the durability of the interphase region. For understanding wood bond strength, Horioka used the analogy of links in a chain [5]; each domain is a separate link, and the weakest link is the site of failure. To use this analogy, one needs to understand what these links look like in a real bond. In Fig. 1, fluorescence microscopy is used to distinguish the adhesive from the wood. The striking feature in this photograph is how large the wood interphase region is compared with the interface, adhesive interphase, and bulk adhesive regions. Pictured is a relatively thick adhesive layer; often there may be no significant bulk adhesive layer. Although interface properties are important, this figure shows that adhesive penetration into the wood could play a dominant role.

Flow of the adhesive to fill the surface micro-roughness is important for all bonding, but adhesive penetration into the substrate is not a significant issue in most bonding applications.

However, good penetration into the wood is a very important aspect of wood bonding. Standards such as ASTM D 2559 require bond formation within the minimum and maximum of the recommended open and closed assembly times [9]. Sufficient penetration into the wood is considered important for good bond formation, but overpenetration produces a starved bondline that is the weak link. Overpenetration does not occur with non-porous substrates; thus, different factors need to be considered in formulating and using wood adhesives. A lower viscosity adhesive is normally better for the wetting and adhesion, but for wood the adhesive can be so thin as to overpenetrate into the wood.

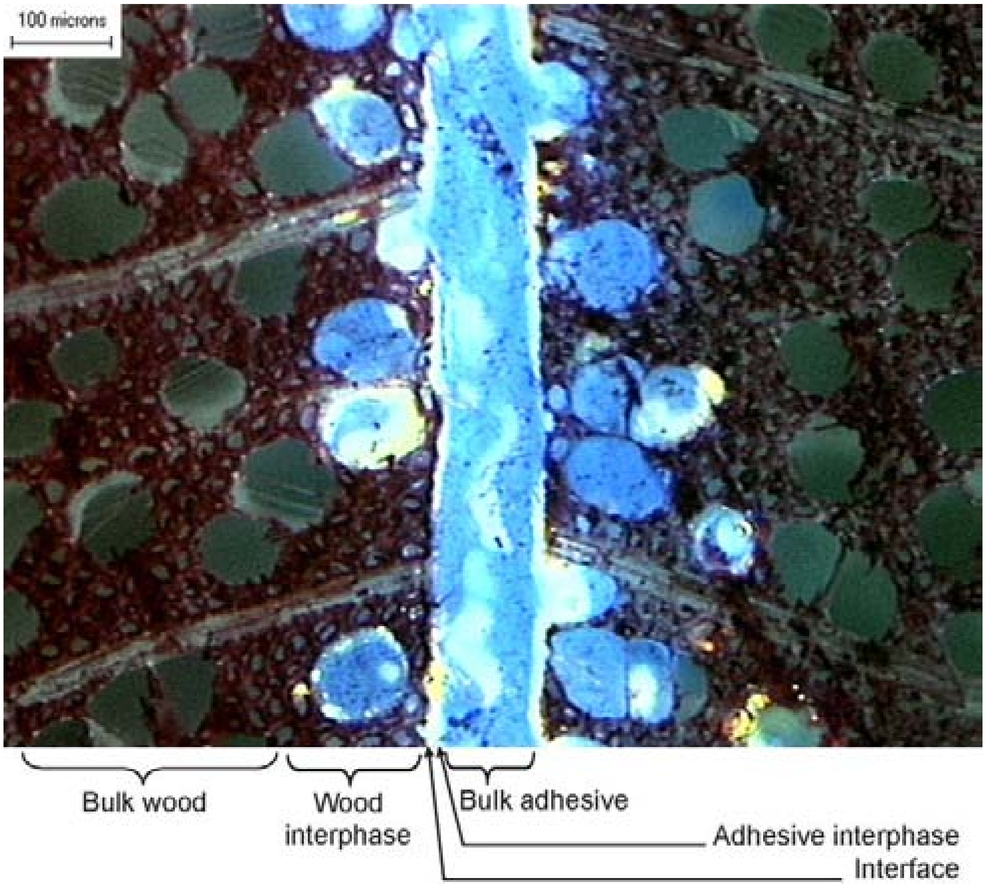

FIG. 1—*Wood bondline of an epoxy adhesive using fluorescence microscopy to show regions of the bonded assembly.*

Although penetration is a very important aspect in wood bonding, the relative importance between penetration into lumens and into cell walls is not normally discussed. For bonding, penetration into a lumen depends on the adhesive's contact angle on the wood surface and the bulk adhesive viscosity, whereas penetration into the cell walls depends upon molecular size of the adhesive components and may depend upon the water or solvent swelling of the wall structure. For performance testing, filling of the lumen is a mechanical interlock that provides additional mechanical strength, while penetration into the cell walls can change their mechanical strength and swelling ability [6,10]. The reduced swelling could have significant effect in reducing the stress concentration at the interface. In addition, penetration of adhesive into the wall changes a sharp wood-adhesive interaction into a more diffuse boundary layer. In Fig. 2, adhesive penetration into the micro-channels in the wood [11] could serve as a nano-mechanical interlock (interdigitation). Another model involves shallow adhesive penetration and crosslinking within the surface cell wall layer to form an adlayer. Deeper penetration and more crosslinking within the wall causes the formation of an interpenetrating polymer network [12], which of all these mechanisms, would most stabilize the wall towards dimensional changes. If the adhesive penetrates into the cell wall to form a bridge, then the role of primary and secondary chemical bonds at the adhesive-wood interface might be less important. Although numerous methods have shown that some adhesives penetrate the cell wall and change its physical properties [6], reports of data describing the effect on adhesive strength are very limited. It would be useful to determine if adhesives giving poor durability do not stabilize the cell wall, whereas those that have durability do provide stability to the cell wall.

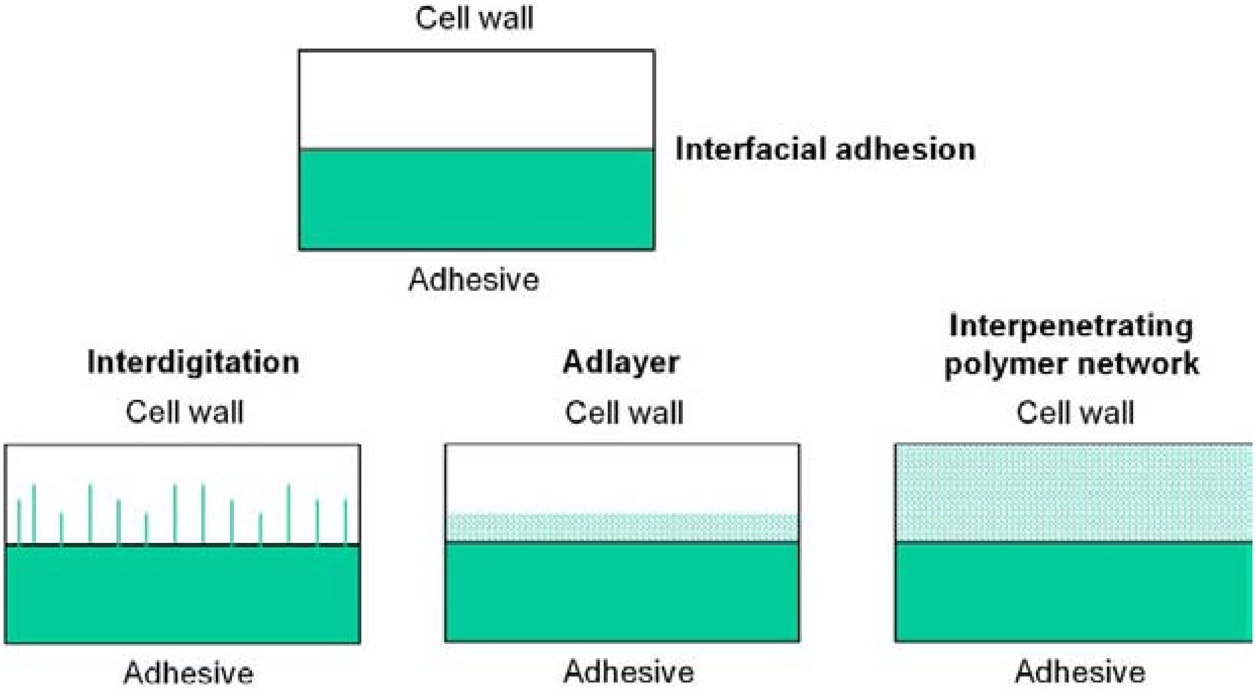

FIG. 2—*Models to illustrate the difference between an interfacial bond and those involving adhesive penetration into the wood cell wall, including interdigitation, adlayer, and a fully interpenetrating polymer network.*

Another difficulty in understanding wood bonding is that although much discussion focuses on primary and secondary bonds between the adhesive and the wood, the chemical composition of the surface layer is not clearly understood. Although cellulose is the main wood component, fracture is probably more likely in the hemicellulose and lignin layers because of their weaker mechanical strength. Prior work has indicated that hemicellulose is the main compound for hydrogen bonding on wood surfaces because of its greater accessibility [13]. On the other hand, lumen walls can be high in lignin content from the warty layer [14]. The planed wood surfaces in Figs. 3 and 4 do not show much evidence of cellulose fibrils on the surface but are more consistent with a material, like lignin, that can flow and create a smoother surface. Factors favoring lignin-rich over cellulose-rich surfaces include the following: it has been identified as the main component of the warty layers that exist on many lumen walls, it is the most likely to flow upon the friction and heat of planing, and it provides the lowest energy surface. Hemicellulose may also be present, but the cellulose is likely to be the least accessible.

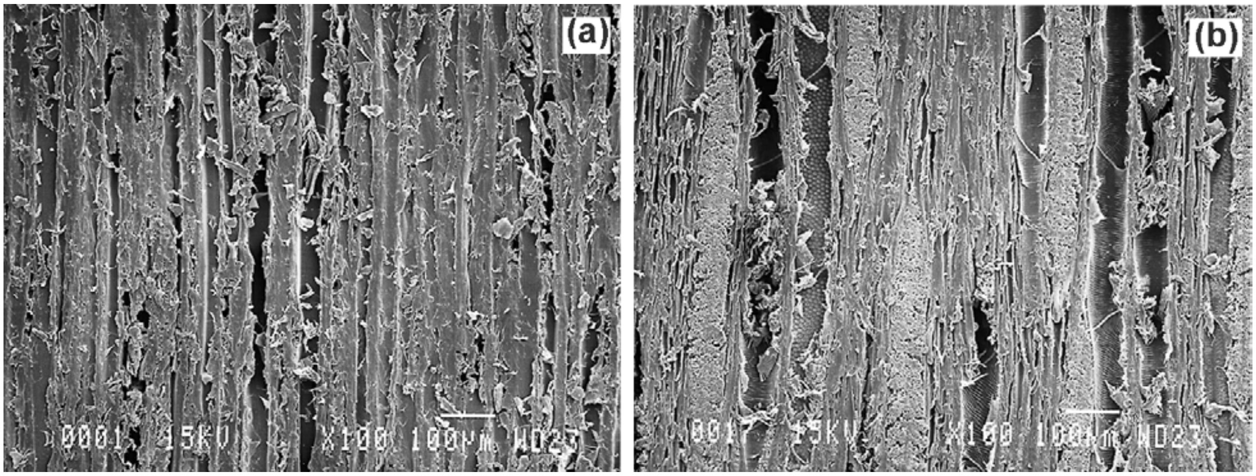

FIG. 3—*Scanning electron micrographs of (*a*) the softwood southern yellow pine and (*b*) the hardwood hard maple to show the fragmented surface produced by planning.*

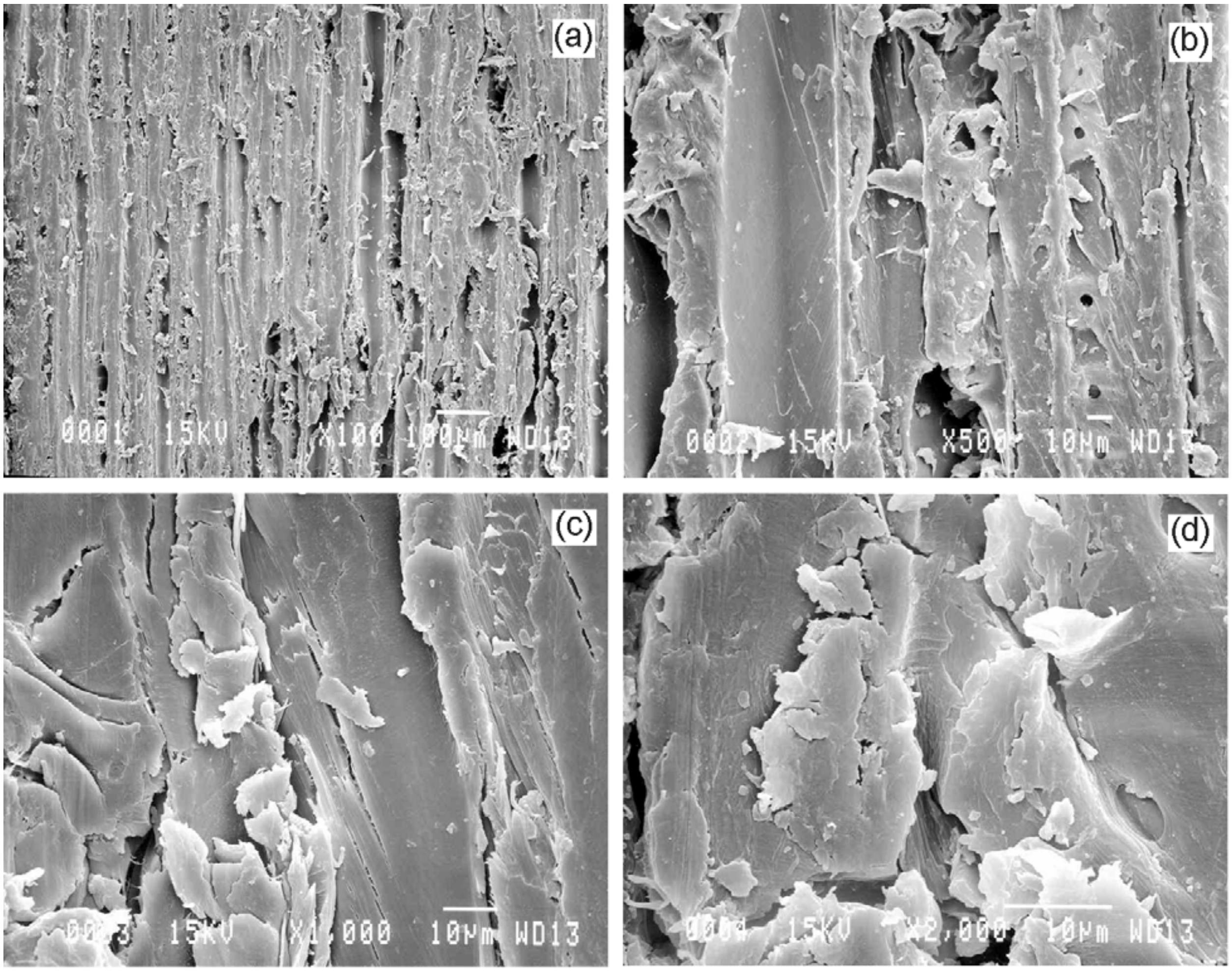

FIG. 4—*Scanning electron micrographs of yellow poplar to show that the fragmented surface produced by planing is evidenced by examining under higher magnification.*

Surface roughness is an important factor for all bonding applications because it often limits surface wetting. The surface roughness of wood, with its cellular structure, is orders of magnitude greater than that of surfaces present in most other adhesive applications. However, the surface is not the orderly structure normally pictured; rather it is much more fractured and irregular, as the examples of hard maple (hardwood) and southern yellow pine (softwood) in Fig. 3 illustrate. Even though sharp planer blades were used to prepare these surfaces, the surfaces are still very fractured and ragged. Obviously, fragments and covered ray ends on the wood surface can limit adhesive penetration into the wood and bonding to sound surfaces. The higher magnification of planed yellow poplar in Fig. 4 emphasizes the extensive shredding of surface cell walls. Although several articles have addressed the penetration issue [15,16,17], the effect of adhesive type and wood species on penetration is not well understood.

Understanding adhesive-wood cell interactions is more difficult because of the tremendous variability in wood cell types. With tracheid, parenchyma, and fiber cells, vessels, resin canals, and ray cells that vary in composition and structure in the earlywood, latewood, sapwood, and heartwood domains, there is a tremendous variety of bonding surfaces, each of which may interact differently with the adhesives. The most dramatic difference is often between wood species because of the large difference in cellular architecture. Bonding of different species often requires changes in adhesive formulation to control penetration into the wood. Although some work has been done on determining penetration into cell lumens and walls [6], this information is usually not related to the performance of the bonded assembly.

Many classes of adhesives are used in wood bonding because of different production and end-use conditions. Most adhesives can give acceptable wood bonds if the use conditions are not too strenuous or at high moisture levels. The interaction of a hot-melt adhesive with wood should be quite different from that of a water-borne adhesive, not only because of viscosity differences but also because of the lack of cell wall swelling by hot-melt adhesive. In addition, some adhesives penetrate and change the mechanical properties of cell walls [10,18], but it not known if all adhesives that penetrate cell walls change their mechanical properties.

Although many techniques have been used to examine how adhesives interact with wood [6], the observations generally have not been related to bond performance. Does penetration into lumens result in better strength, or is penetration into the cell walls more important, especially for durable exterior bonds?

Performance Testing

Adhesives are used to hold substrates together under the desired end use conditions. This means that a bond needs sufficient strength and durability to hold the substrates together under a defined set of conditions. Generally, strength and accelerated tests are used to establish the suitability of an adhesive for a specific application. The approval of an adhesive, especially for those applications that are more demanding upon the adhesive, are often quite specific for the bonding of a single wood species under specific conditions, as in ASTM D 2559 [9]. The durability of adhesives that pass these accelerated tests has been borne out by their performance in actual end use over many years. The question arises whether these accelerated aging tests are too conservative—some adhesives may not pass these tests but may actually be durable enough in the end use. The concern is that the tests involve rapid wetting and drying, not allowing stress to be dissipated through the stress relaxation of the wood. The validity of accelerated tests is always a difficult question, but for an accelerated test to be an accurate predictor, the mode of failure in both the accelerated test and end use must be the same. This implies that we need to understand the forces exerted upon the bondline for both the accelerated tests and the end use conditions.

The question remains as to why some wood adhesive bonds are durable under exterior conditions and many are not. Most of the research has been on the adhesive chemistry, adhesive formulation, and testing durability; only limited resources have been applied toward understanding what contributes to durability. It is generally understood that for structural adhesives, the adhesive needs to have sufficient crosslinking to support creep resistance. However, why do only a few adhesives provide low delamination in the ASTM D 2559 test? The poor durability of urea-formaldehyde has been explained on the basis of depolymerization of the adhesive, but why does the addition of melamine to the urea-formaldehyde promote durability? Why is a phenol-formaldehyde adhesive more durable than an epoxy adhesive in wood bonding, given the durability of epoxies in metal bonding and coatings, cement coatings, and other applications?

For wood bonds, when failure is identified, it is usually divided only between wood and bondline failure. Bondline failure is often considered strictly a lack of adhesion because of the difficulty in determining the main failure location on the complex wood surfaces. Horioka has classified five locations for failure in going from the bulk wood to the bulk adhesive [5]. For epoxies, failure usually occurs in the bulk wood under dry conditions and often in the adhesive interphase region under wet conditions [19]. Why should there be significant failure in the epoxy interphase region under wet conditions? One explanation is that the epoxies are unable to withstand the strain during the expansion of the wood as it absorbs the water under soaking conditions, as in Fig. 5. Microscopic examination supports this concept in that the fracture lines are highly oriented in the longitudinal direction as expected from a swelling force (Fig. 6). Also supporting this concept is the increased wood failure in ASTM D 905 compressive shear tests when using epoxy-bonded acetylated wood with its low volume swelling compared with bonded unacetylated wood with its high swelling [20]. In another set of experiments, the effect of increased stress on the bondline from water soaking has been exhibited for FPL-1A epoxy bonding of several different wood species evaluated using ASTM D 905 tests. As expected, the percentage wood failure dramatically decreased in going from the dry tests to the vacuum-pressure water soaks in the shear test. Surprisingly, the percentage wood failure returned to its original values when the wet samples were allowed to dry in an 80°C, 65 % humidity room (Fig. 7). This indicates that increased bondline failure results from a combination of internal stress from the swelling of the wood and the applied load, but removal of the internal stresses upon drying causes recovery of much of the bondline strength. This cell wall stabilization model has also been used to explain the ability of hydroxymethylated resorcinol primer to reduce the delamination of epoxy adhesives in the D 2559 test [21].

Understanding where failure occurs has often been difficult with wood adhesives because of problems with visualization of colorless adhesives, such as epoxies. Fluorescence microscopy can be one way to learn where and why failure is occurring. This can be done by looking down onto the failure surface (Fig. 8) or in the cross-section (Fig. 9).

How to determine sufficient durability of wood bonds is still of great concern. ASTM D 2559 has been considered the ultimate test, but there has been little discussion of forces on the bondline during this test and how representative they are of those experienced in the actual application. The test involves cycles of rapid water soaking by vacuum-pressure soaks followed by rapid drying in a hot oven. Not surprisingly, this test causes considerable distortion and fracturing of the wood itself because the dimensional changes are so rapid that the wood does not have a chance to stress relax. The bondline needs to deal not only with shear, tension, and compressive forces in the radial direction but also with the distortion of the wood and normal tension forces. Can these results be correlated to ASTM D 905 tests that involve shear tension forces from the wood swelling in the radial direction and compressive shear forces in the longitudinal direction as has been claimed [22]? How do these tests compare to laminated beam applications where bondlines are under tension at the bottom of the beam and compression at the top as the wood is more gradually swelling and shrinking? This is not to imply that ASTM D 2559 does not have utility, but we do not have the scientific understanding to know if it is excessively conservative (useful adhesives being unable to pass this test) or too liberal (adhesives not being under external forces during the swelling and shrinking).

Despite the success of many adhesives in a variety of applications, other challenges still exist with wood adhesives. How can we get equal performance out of a lower cost adhesive? As the wood supply changes from old growth wood to more juvenile wood, how do we develop the proper adhesives, and how do we address performance standards when the wood itself is weaker?

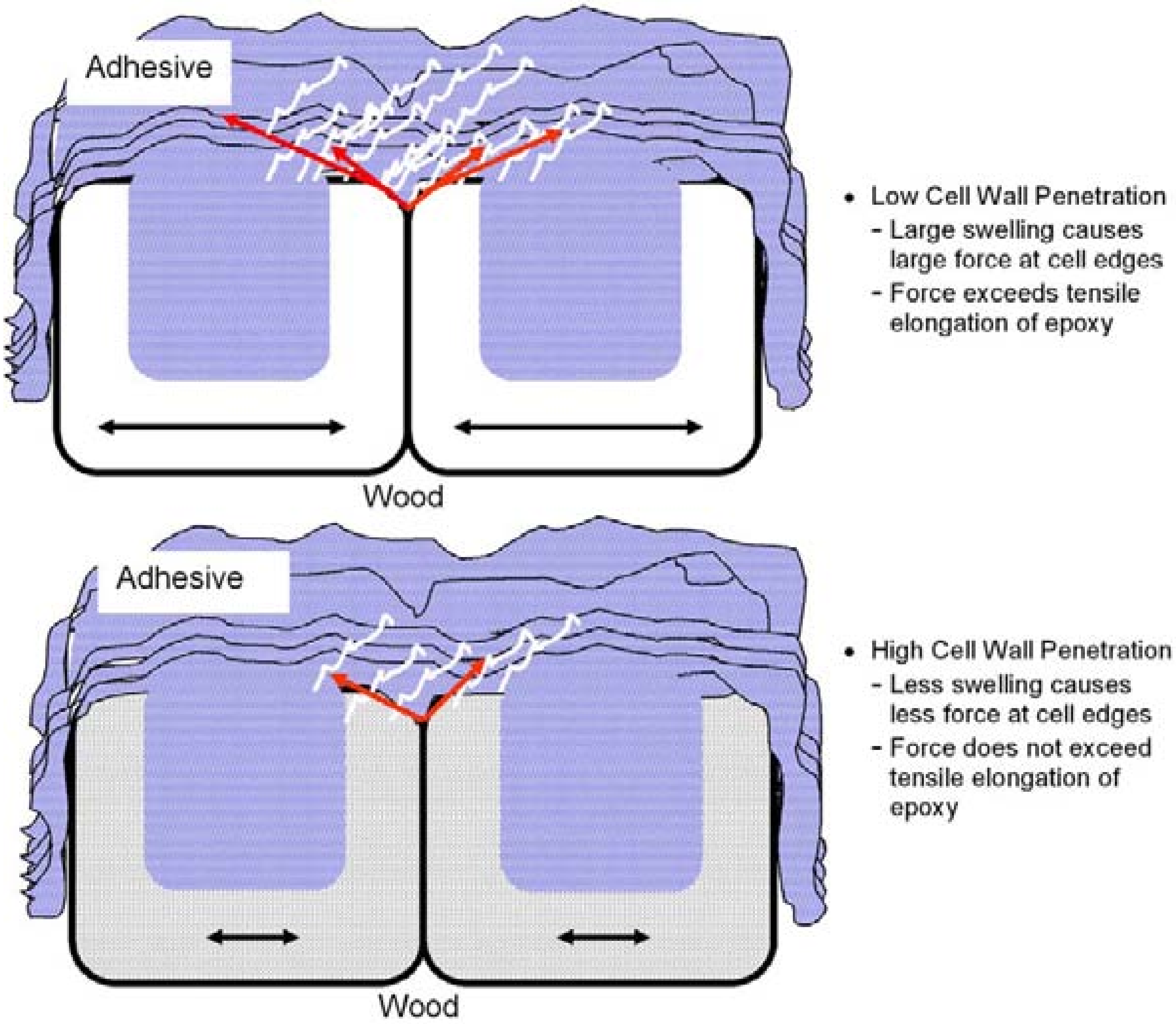

FIG. 5—*Drawing to illustrate the difference in force upon the adhesive interphase region between cells with high and low swelling.*

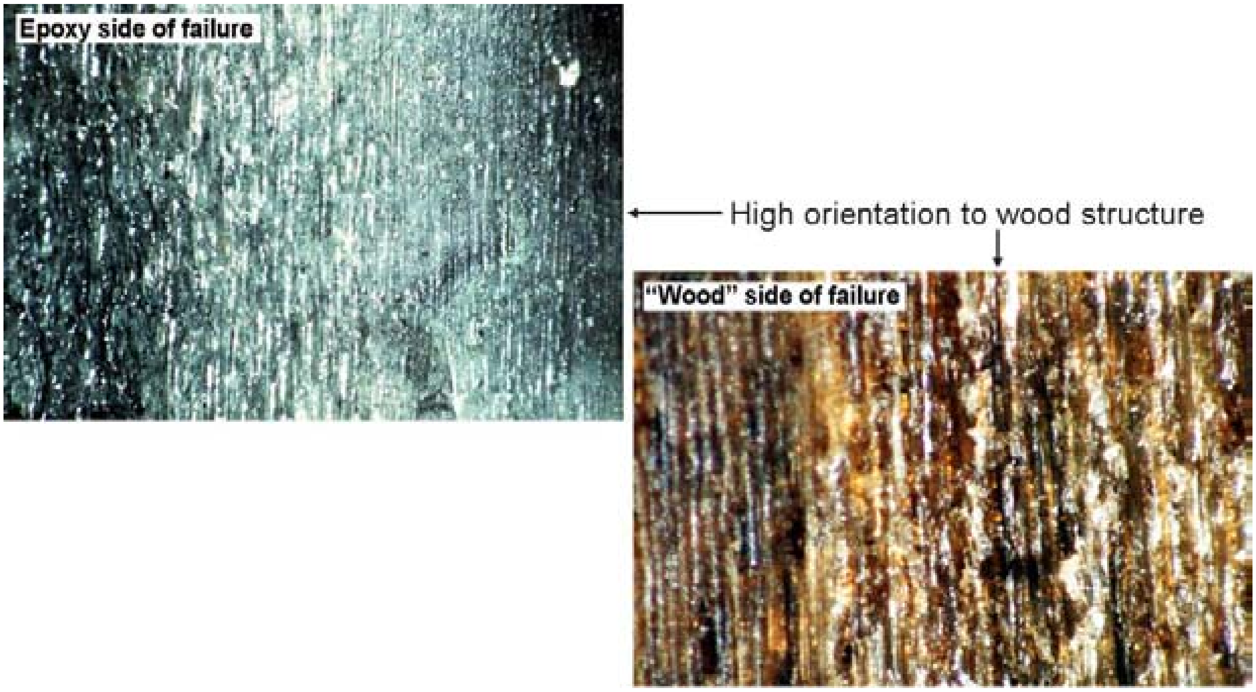

FIG. 6—*Fracture surface produced under ASTM D 2559 to show the high orientation parallel to the wood orientation even though fracture is mainly in the epoxy interphase region.*

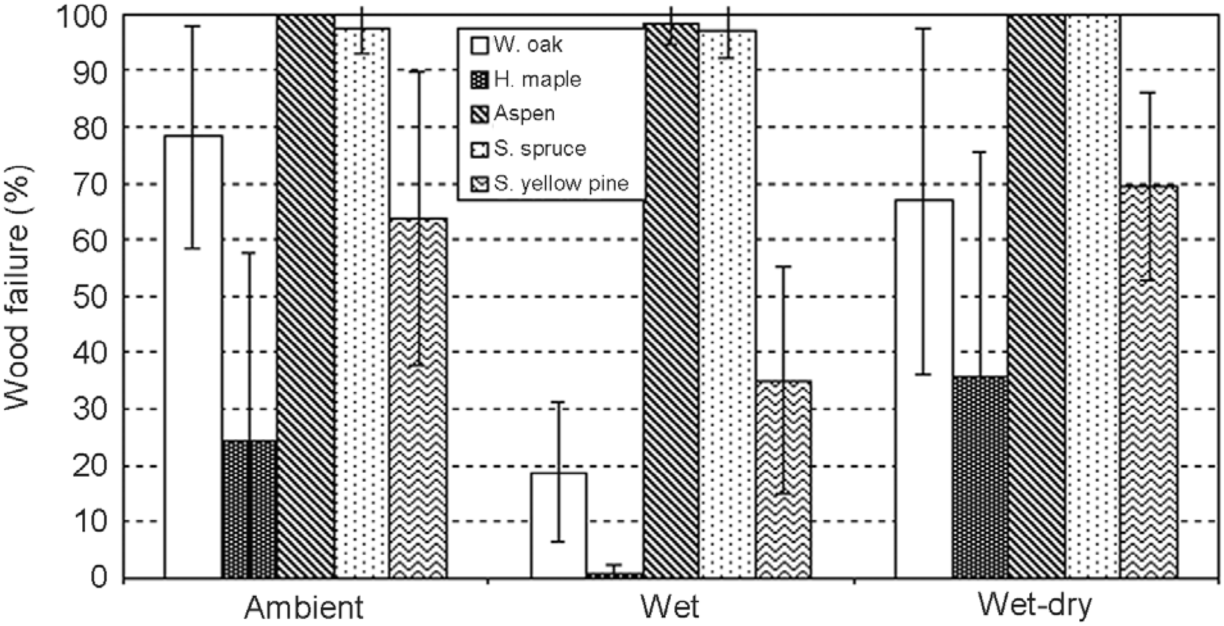

FIG. 7—*Percentage wood failure of five wood species bonded with FPL 1-A epoxy using ASTM D 905 to show loss of bondline strength from vacuum-pressure water soak and recovery upon redrying.*

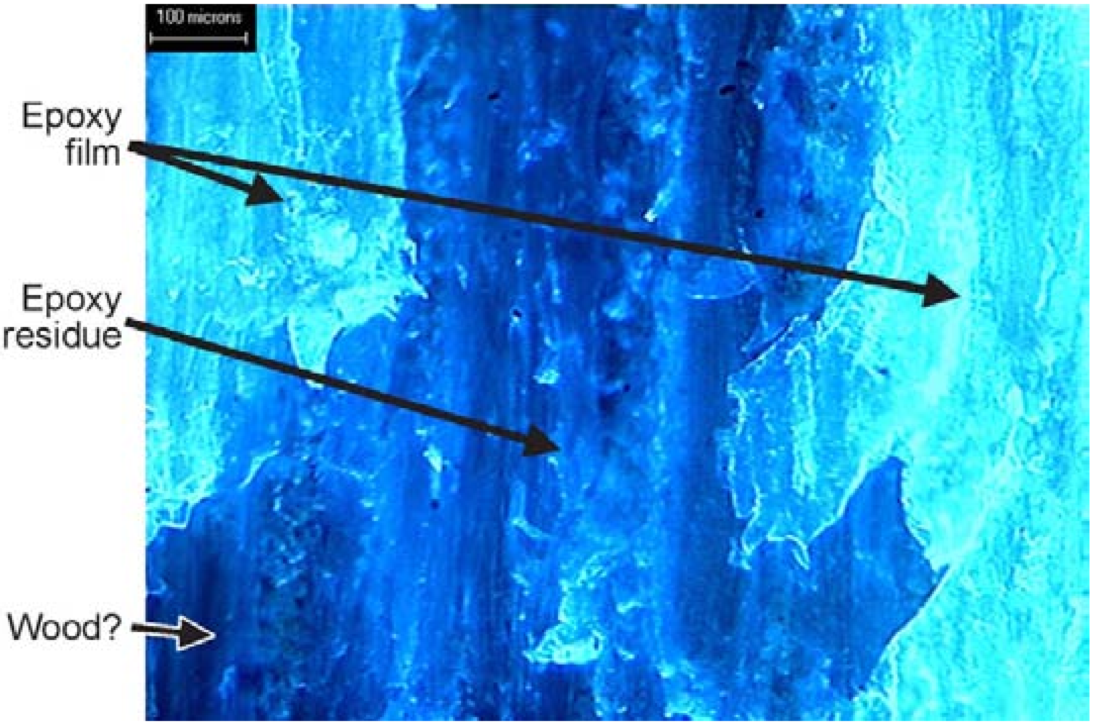

FIG. 8—*Use of fluorescence microscopy to show the failure location of epoxy-bonded yellow poplar after vacuum-pressure water soak.*

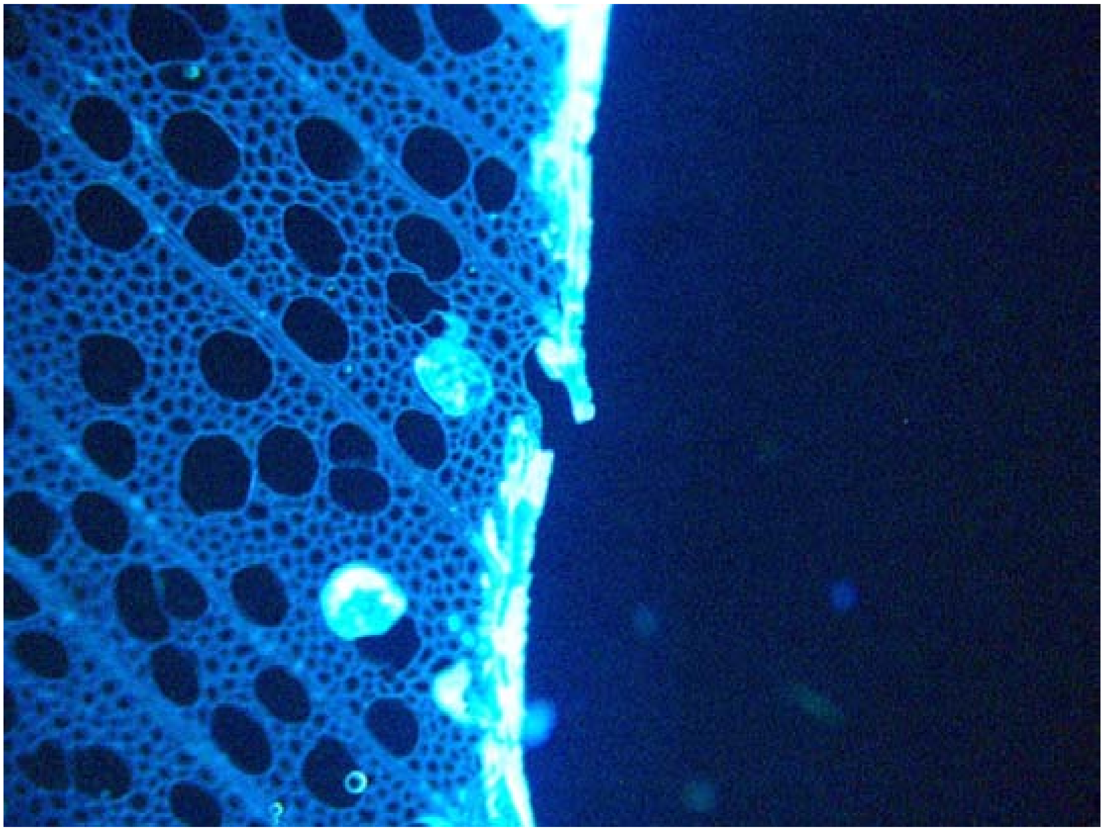

FIG. 9—*Use of fluorescence microscopy on a cross-section to show failure of yellow poplar bonded with epoxy adhesive after vacuum-pressure water soak.*

Conclusions

Adhesives provide good wood bonds for a wide variety of applications. Knowledge of adhesive chemistry, adhesive formulation, and adhesive-wood interactions has increased dramatically. Understanding the location and cause of bondline failure has lagged behind. To understand bonding and performance testing processes, more emphasis needs to be placed upon

- understanding the location of bondline failures using the Horioka model,
- determining the internal and external forces causing bondline failures,
- relating strength factors to the bonding process, and
- knowing the morphology and chemistry of the wood bonding surface better.

By applying existing and newly developed techniques to the study of specific adhesives, a much better knowledge of factors leading to durable bonds can be obtained. It is important to use analysis techniques in concert and to apply them to samples evaluated by the standard adhesive performance evaluation methods.

By using these processes, we have made progress toward understanding why normally durable adhesives, such as epoxies, are not as durable in bonds with wood. The stabilization of the surface cell wall toward expansion and contraction seems to play an important role in minimizing the stress concentration in the interphase region. Although this mechanism is not definitively proven, it is consistent with the data on unmodified wood, acetylated wood, and wood primed with hydroxymethylated resorcinol.

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