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Quantitative wood–adhesive penetration with X-ray computed tomography

Jesse L. Paris [a,*], Frederick A. Kamke [b,**]

[a] Willamette Valley Company, 585 McKinley Street, Eugene, OR 97402, USA
[b] Department of Wood Science and Engineering, Wood-Based Composites Center, Oregon State University, 119 Richardson Hall, Corvallis, OR 97331, USA

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ABSTRACT

Micro X-ray computed tomography (XCT) was used to analyze the 3D adhesive penetration behavior of different wood–adhesive bondlines. Three adhesives, a phenol formaldehyde (PF), a polymeric diphenylmethane diisocyanate (pMDI), and a hybrid polyvinyl acetate (PVA), all tagged with iodine for enhanced X-ray attenuation, were used to prepare single-bondline laminates in two softwoods, Douglas-fir and loblolly pine, and one hardwood, a hybrid polar. Adhesive penetration depth was measured with two separate calculations, and results were compared with 2D fluorescent micrographs. A total of 54 XCT scans were collected, representing six replicates of each treatment type; each replicate, however, consisted of approximately 1500 individual, cross-section slices stacked along the specimen length. As these adhesives were highly modified, the presented results do not indicate typical behavior for their broader adhesive classes. Still, clear penetration differences were observed between each adhesive type, and between wood species bonded with both the PF and pMDI adhesives. Furthermore, penetration results depended on the calculation method used. Two adhesive types with noticeably different resin distributions in the cured bondline, showed relatively similar penetration depths when calculated with a traditional effective penetration equation. However, when the same data was calculated with a weighted penetration calculation, which accounts for both adhesive area and depth, the results appeared to better represent the different distributions depicted in the photomicrographs and tomograms. Additionally, individual replicate comparisons showed variation due to specimen anatomy, not easily observed or interpreted from 2D images. Finally, 3D views of segmented 3D adhesive phases offered unique, in-situ views of the cured adhesive structures. In particular, voids formed by CO_2 bubbles generated during pMDI cure were clearly visible in penetrated columns of the solidified adhesive.

1. Introduction

Wood-based composites represent a broad class of materials which are critically important to residential construction, timber engineering and furniture manufacturing industries. The performance of all adhesively-bonded materials, including wood products, depends on the ability for the adhesive to transfer mechanical stresses across the joint interface. As wood is a natural, porous material, the nature of this interface is highly variable, and liquid adhesives can flow and penetrate the cellular substrate during bonding. While the term “interface” is designated to a virtual two dimensional surface between the adhesive and the adherend, “interphase” relates to the volume adjacent to the interface, which includes the region of adhesive penetration and transition between pure and mixed material properties [1]. Adhesive penetration occurs at multiple scales in this interphase region. Bulk, or micro-scale, penetration describes flow into and between cell lumens and voids. However, some adhesives containing low-molecular weight components may penetrate the wood cell walls, at the molecular level, or nanometer scale. Bulk penetration transfers bondline stresses into sound wood to avoid stress concentrations around damaged cells caused during surface preparation prior to bonding. High stress zones might also occur at abrupt boundaries between the adhesive and adherend(s) when they have different elastic moduli, but bulk penetration can provide a smoother material-property gradient in the interphase region [2]. Furthermore, bulk flow increases the surface area over which the intermolecular forces and chemical associations governing adhesion effectively operate, and once cured, solidified adhesives provide mechanical interlocking resistance to hold the substrates together [2]. Molecular scale penetration can increase cell wall modulus and dimensional stability, which can ultimately affect bondline moisture durability [3]. Over penetration, on the

* Corresponding author. Tel.: +1 804 338 4383.
** Corresponding author. Tel.: +1 541 737 8422; fax: +1 541 737 3385.
E-mail addresses: jesse.paris@wilvaco.com (J.L. Paris), fred.kamke@oregonstate.edu (F.A. Kamke).

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other hand, can result in a starved bondline, where too little adhesive remains in the bondline for efficient load transfer [1]. Furthermore, composite manufactures wish to avoid excessive penetration, which is wasteful of expensive resin materials.

While the influence of adhesive penetration on wood–composite joint performance has been extensively studied [1,2,4], there is still no clear consensus as to what represents an optimal penetration depth. This is, in part, due to the high variability between different adhesive types and formulations, wood anatomy and chemistry, and manufacturing parameters, all of which influence penetration behavior [1,2]. Yet, another primary reason for the lack of stronger depth-to-performance correlations is that adhesive penetration is traditionally observed and quantified with various 2D microscopy techniques, which have two inherent flaws. First, bondline photomicrographs show where an adhesive has penetrated, but not the 3D path it followed to get there. Information is often lost above and below the image-plane that describes the true connectivity of the cured adhesive network. Second, microscopy specimen preparation often requires soaking and/or sectioning, which are inherently destructive to the wood–adhesive bondlines [1,4,5]. Researchers often rely on matched specimens, or ones prepared under similar conditions, to correlate mechanical performance and penetration results, but natural anatomical variations can be fairly drastic even for matched specimens.

Micro X-ray computed tomography (XCT) offers a unique solution to both challenges. Several researchers have demonstrated the power of non-destructive, micro XCT for studying 3D wood anatomy [6–8]. XCT analyses of wood–adhesive bondlines, on the other hand, have been more challenging due to the lack of X-ray absorption contrast between wood cell walls and adhesive polymers [9]. Some studies have tried doping adhesives with heavy metal particles or ions to improve the adhesive X-ray attenuation relative to wood [10,11]; however, quantitative results were often complicated by independent tag mobility and agglomeration, which lead to heterogeneous adhesive X-ray properties.

Recently, the authors presented methods for uniformly tagging three different adhesives, a phenol formaldehyde (PF), a hybrid polyvinyl acetate (PVAc) mixed with PF, and a polymeric diphenylmethane diisocyanate (pMDI), with iodine, such that they could be quantitatively segmented with absorption-contrast, micro XCT [12–14]. These adhesives, hereafter referred to as IPF, hybrid PVAc, and IpMDI, respectively, were used to bond laminates with two different softwood and one diffuse porous hardwood species [13,14]. The broader goal of this work was to use digital, segmented wood and adhesive volumes as inputs in a 3D, numerical model to assess the relationship between adhesive penetration and bondline mechanical performance [12]. The same, undamaged, XCT specimens were subsequently mechanically tested with various micro-bond tests, in parallel to the model simulations [15,16]. Comparisons between the simulated and actual results were then used to calibrate and validate the model based on a reverse problem solving methodology, including the true specimen anatomy and penetration profile [12,15]. Testing the influence of adhesive penetration on bond performance by manipulating penetration is impossible. Any change to the resin formulation will influence cured polymer behavior and thus invalidate any mechanical test on the bond. The approach of this study, and the subject of a future report, is to use computer numerical modeling. The model is based on the physical system defined by the tomography data, where each phase identified by segmentation is assigned a set of material properties. The results of the model are then compared to mechanical test results on the same specimen that was reproduced in the tomography data [12].

In total, 54 specimens, representing six replicates per adhesive/adherend treatment, were scanned with micro XCT. In addition to providing a robust sampling for the model development, this work also represents one of the largest, if not the largest, digital collections of micro-scale, 3D wood–adhesive penetration data, to date. Segmented wood cell wall and adhesive material phases in these specimen volumes offer novel 3D views of the full interconnectivity and adhesive distribution in the wood bondline interphase [14]. Furthermore, the 3D perspectives provide unique, qualitative information about the in-situ structure and morphology of the cured adhesives. Additionally, cell wall penetration was observed in several of the IPF and IpMDI bondlines [14]. While material segmentation in a cell wall was below the resolution limits in this study, averaged adhesive/wood X-ray gray values were observed in cell walls adjacent to lumens filled with these adhesives.

The primary intent of this paper is to introduce the effectiveness of the micro XCT technique for quantitative measurement of 3D wood adhesive penetration. Penetration depth was calculated along the length of each specimen bondline and compared with 2D photomicrographs of specimens excised from the same bonded laminates. Two separate penetration depth calculations are used. The first, effective penetration (EP), has been previously used in several wood–adhesive studies [5,17]. The second, weighted penetration (WP), is proposed here, and is similar to the second moment of area calculation often used in mechanical engineering disciplines [18]. The WP calculation accounts for both the area and perpendicular distance an adhesive object has penetrated, and is proposed to offer a better representation of the adhesive penetration and distribution than the EP calculation. With these penetration results, effects of adhesive type, wood species, and variability in wood anatomy between and within replicates are compared.

2. Materials and methods

XCT adhesive bondlines were scanned and reconstructed at the Advanced Photon Source, at the Argonne National Laboratory in Argonne, Illinois. Tomogram resolution was 1.45 μm per voxel side length. The presented adhesive penetration results and subsequent discussion refer to sub-volume regions of interest, which were digitally excised from the central region of each XCT specimen. Sub-volume dimensions were approximately 500–750 voxels (725–1080 μm, radial or perpendicular to the bondline) × 750 voxels (1087 μm tangential or parallel to the bondline) × 1500 slices (2175 μm longitudinal or along the bondline length).

Specific methodology regarding adhesive formulation, bonding, XCT scanning procedures, 3D tomogram reconstruction, and material segmentation in XCT sub-volumes is described in detail elsewhere [12–14]. Still, pertinent descriptions of each system are again provided here. The wood species bonded were Douglas-fir (*Pseudotsuga menziesii,* DF), loblolly pine (*Pinus taeda,* SYP) and a hybrid poplar (*Populus deltoidesxPopulus trichocarpa,* HP). Prior to bonding the wood adherends were all equilibrated in a 20 °C, 65% relative humidity conditioning room (moisture content between 10% and 12%) [19]. The IPF was prepared according to a typical 44% solids, resol PF recipe, but meta-iodophenol was used in substitute of phenol [13]. The hybrid PVAc system was a 50:50 blend of an emulsion PVAc (54 wt% solids) with the IPF system above. The two neat adhesives were first diluted with water for increased blend homogeneity, and the resulting hybrid system had 24.1 wt% solids [14]. The IpMDI adhesive was prepared by reacting a commercial pMDI resin (Huntsman, Rubinate 1840) with 2,4,6 triiodophenol dissolved in tetrahydrofuran (THF). The resulting adhesive had 78.3 wt% solids and an isocyanate content of 20%, reduced from 31% in the neat pMDI resin [14]. Cured samples of the IPF, hybrid PVAc, and IpMDI systems contained 39.5, 23.9 and 24.0 wt% iodine, respectively.