

Chemical Imaging of the Polylactic Acid–Wood Adhesion Interface of Bonded Veneer Products

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Supplementary Materials

Materials

The poly(lactic acid) PLA polymer grades used were semi-crystalline (3052D) and amorphous (4060D) obtained from Natureworks LLC (USA). The wood veneers were European white birch (*B. pendula*, 1.60 mm thickness) and spruce (*Picea abies*, 1.60 mm thickness). The 5-ply PLA laminates were prepared using a laboratory press using three different temperatures of 140, 160, and 180 °C and pressing to stops using a maximum pressure of 45 kN. Further details on sample preparation and testing are available in Grigsby, et al. (2020) Bonding Wood Veneer with Bio-Based Poly(Lactic Acid) Thermoplastic Polyesters: Potential Applications for Consolidated Wood Veneer and Overlay Products, *Fibers* 2020, 8(8), 50

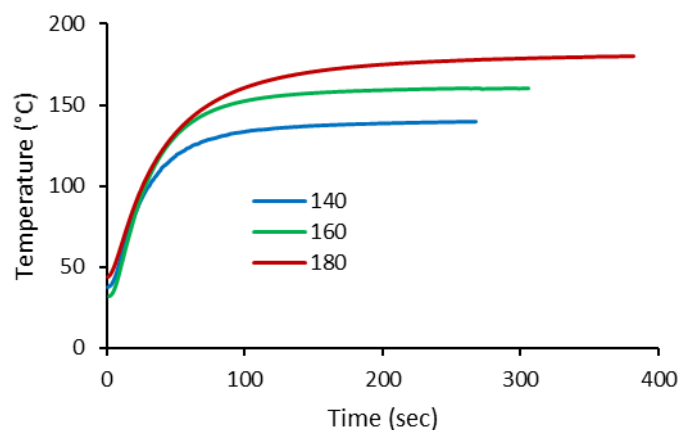


Figure S1. Internal panel core temperature profiles and indicative pressing times for consolidated panels birch-PLA 5-ply panels. adapted from *Fibers* 2020, 8(8), 50.

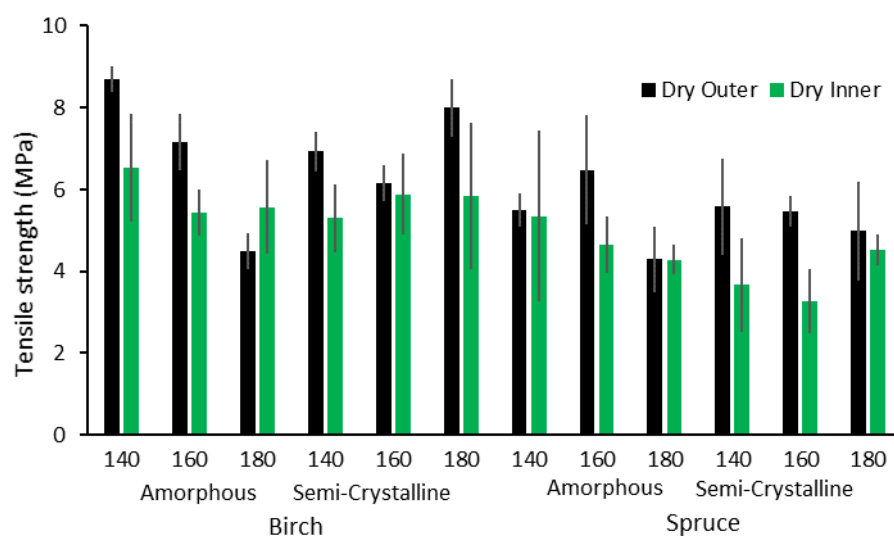


Figure S2. Tensile strength values of 5-ply birch and spruce laminates prepared with amorphous and semi-crystalline PLA. Testing used ASTM D906 - 98(2017) criteria and at least 5 replicates specimens per bondline. Data adapted from Grigsby et al., *Fibers* 2020, 8(8), 50.

Table S1. Representative light microscopy images of 5-ply laminates formed with birch and spruce veneer and PLA foils at differing temperatures.

Wood Species and PLA type	Pressing Temperature (°C)					
	140		160		180	
Birch and Semi-Crystalline PLA						
Birch and Amorphous PLA						
Spruce and Semi-Crystalline PLA						
Spruce and Amorphous PLA						

Note: scale bar is 3 mm and common to all images. At least 5 images were acquired per 5-ply panel sample.

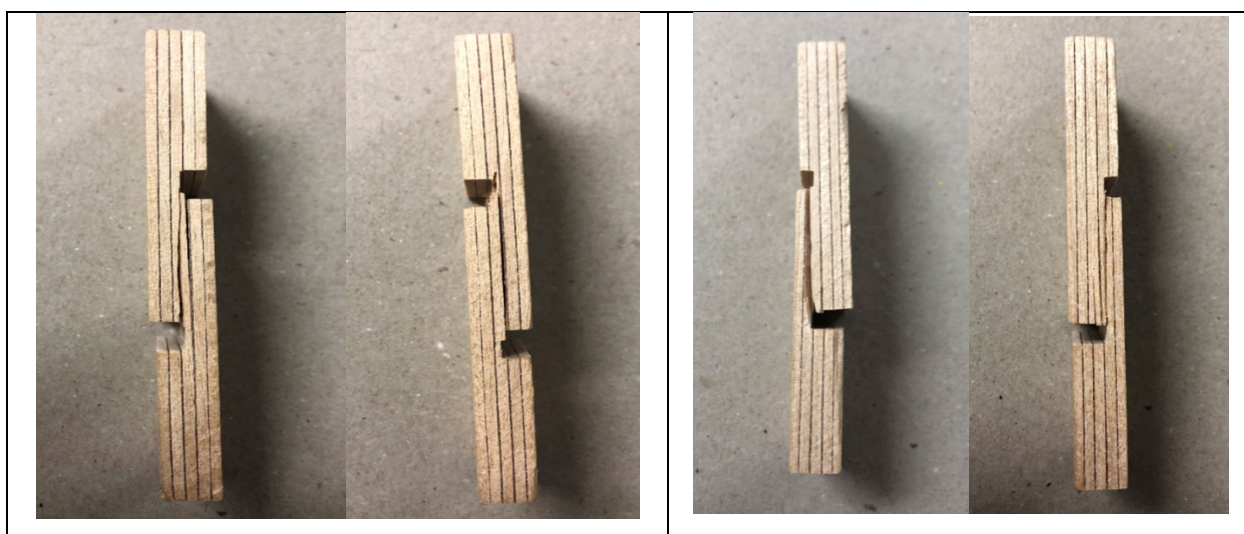


Figure S3. Examples of failed tensile testing specimens profiling modes of wood failure. Shown are the tested inner, core (left) and outer (right) bondlines of a birch-semi-crystalline PLA (140 °C) specimen.

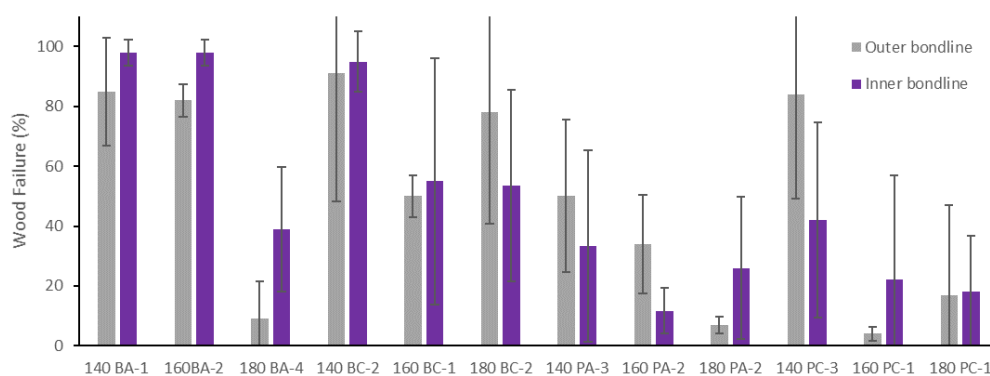


Figure S4. Average %wood failures of samples produced with differing PLA types and pressing temperatures. Assessments compare outer, surface and inner, core bondlines and using both visual and microscopy examination of failed specimens. Note: wood failure assessments are for at least 5 test specimens per bondline (as per ASTM D906 - 98 testing).

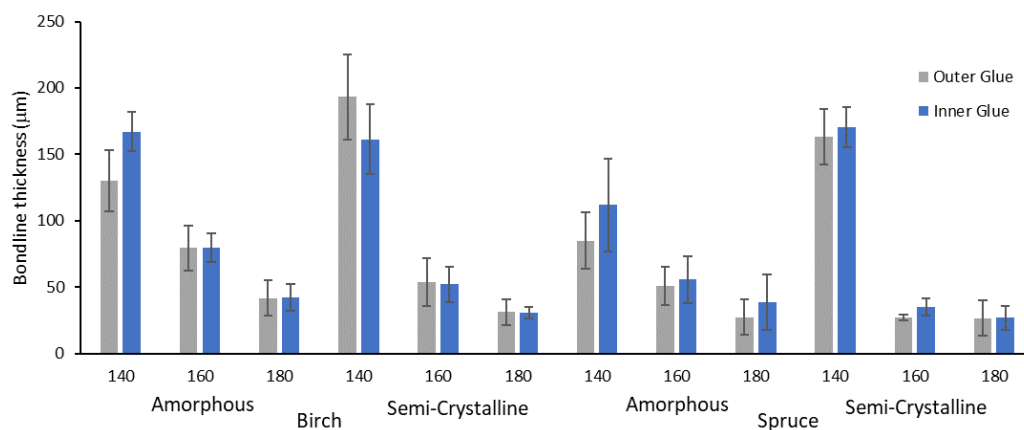


Figure S5. Results of low resolution light microscopy assessments of bondline thickness values for birch and spruce samples produced with different PLA types and pressing temperatures.

Table S2. Comparisons of correlation values between average bondline thickness values and either average sample tensile strength or %wood failure values.

Bondlines, PLA type	Tensile Strength (R^2)		Wood Failure (R^2)	
	Birch & Spruce	Birch-only	Birch & Spruce	Birch-only
Outer, All	0.13	0.10	0.57	0.59
Outer, Amorphous	0.74	0.93	0.79	0.70
Outer, Semi-crystalline	0.01	0.06	0.54	0.42
Inner, All	0.02	0.03	0.34	0.69
Inner, Amorphous	0.62	0.79	0.41	0.53
Inner, Semi-crystalline	0.03	0.94	0.35	0.97

Spatial Chemical Imaging using FTIR

Sample specimens were obtained from sections of 5-ply laminated panels. Specimens were cut into small sections (20×20 mm). Prior to microtoming, specimen surfaces were wetted with ethanol and the softened area then smoothed by microtoming. After microtome preparation, samples were then dried under vacuum for 24 hours prior to FTIR analysis. Samples were mounted on a FTIR microscope (Bruker Hyperion 3000) in preparation of analysis. Each specimen used two areas (*ca.* 2×2 mm) for analysis which included either an inner or outer bondline. User defined, single point measurements were undertaken using MCT detector in ATR mode across each bondline as shown in Figure S6. The resolution was 4 cm^{-1} and used 32 scans.

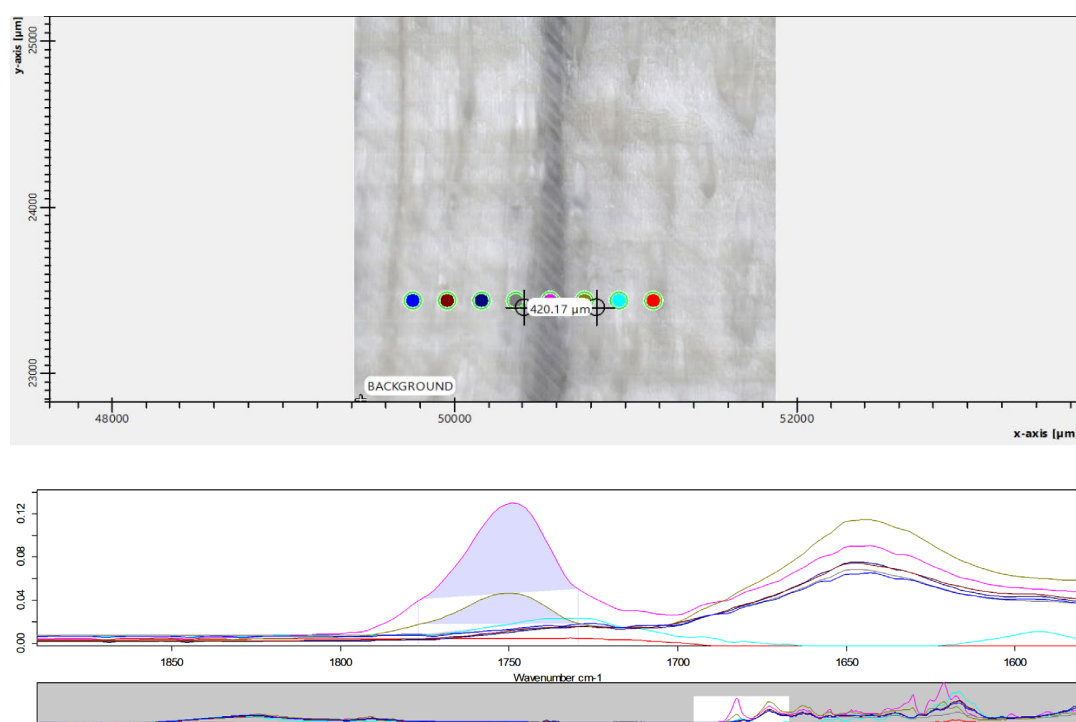
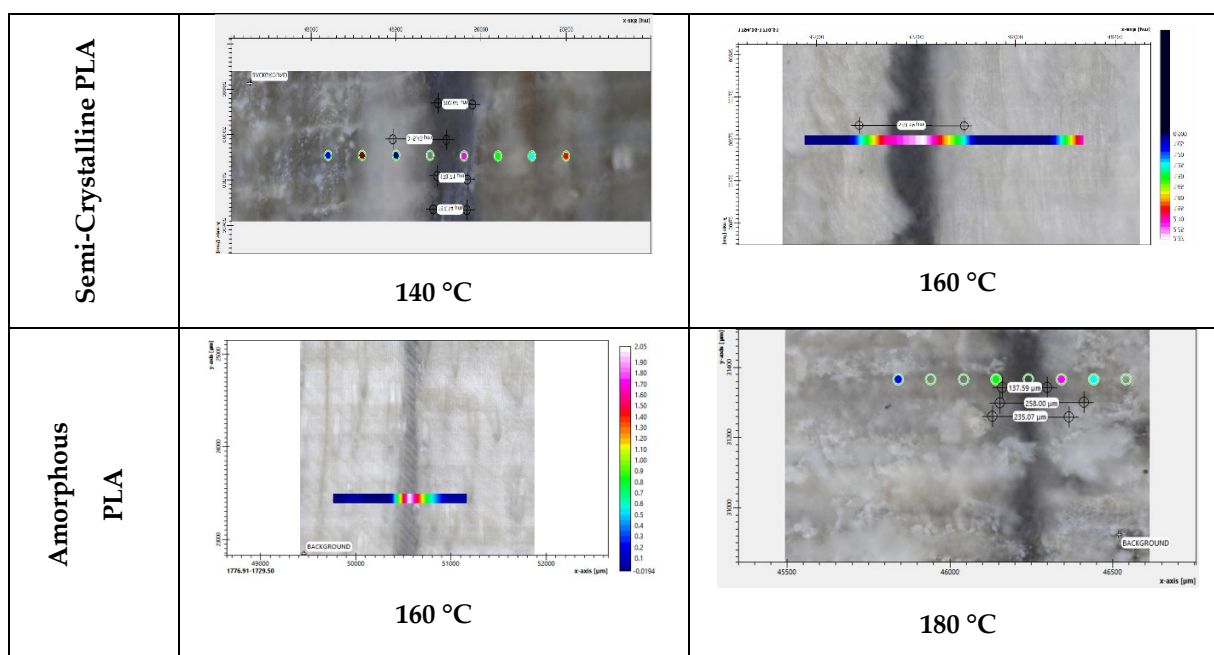


Figure S6. Original, full FTIR microscope image (software generated) of the birch amorphous PLA bondline formed at 160°C prior to image cropping. Each FTIR spectrum (bottom, full spectrum 400 to 4000 cm^{-1} and expanded 1550 to 1800 cm^{-1}) was obtained at defined points (see coloured dots, top) across this image. The PLA carbonyl peak (*ca.* 1750 cm^{-1} , represented by different colours) is profiled across this bondline cross-section.

Table S3. Selected examples of chemical spatial imaging projections of 5-ply laminate inner bondlines formed with birch veneer and PLA foils at differing temperatures.



Using OPUS 8 software (Bruker), PLA carbonyl peak (1750 cm⁻¹) contour plots were developed from FTIR point spectra and projected to imaged cross-sections. User defined measurements of bondline thicknesses and the extent of PLA migration were obtained from PLA contour plots of the PLA carbonyl peak projections. Equation 1 was used to calculate average penetration values from these bondline thickness and migration measurements:

$$\text{average penetration} = (\text{migration} - \text{thickness})/2 \quad \text{Equation (S1)}$$

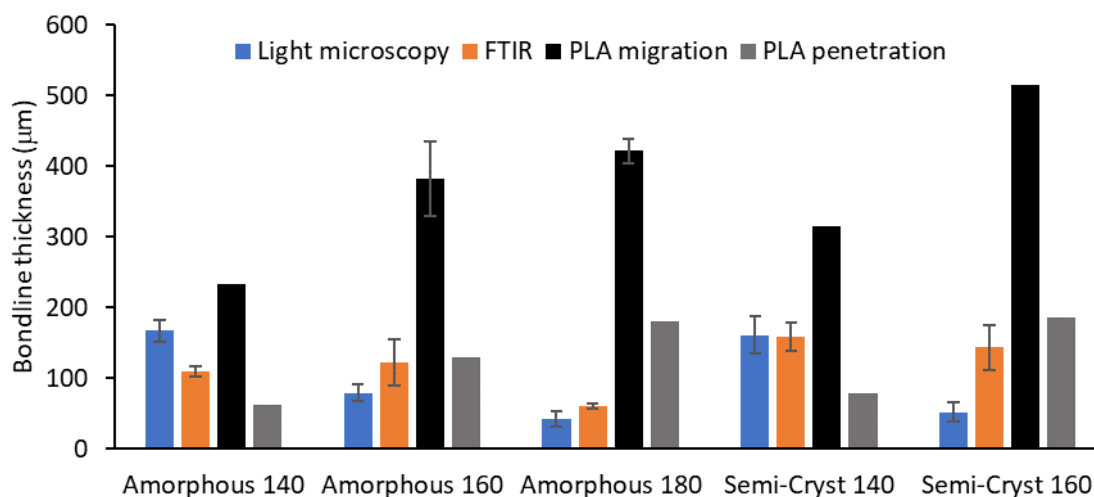


Figure S7. Comparisons of PLA bondline thickness, PLA interphase migration and calculated PLA penetration values from spatial chemical imaging inner bondlines of birch-PLA panels.

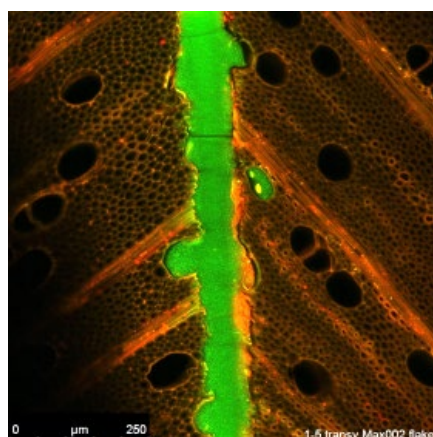


Figure S8. A representative fluorescence confocal microscopy image of a PLA bondline formed with maple veneer and PLA foil in which a fluorescence contrast agent had been grafted to the PLA polymer. Related images can be found in Grigsby et al., *Fibers* 2019, 7(2), 15.