EXAMINATION OF ADHESIVE PENETRATION IN MODIFIED WOOD USING FLUORESCENCE MICROSCOPY

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Abstract

Adhesive bonding takes place when an adhesive undergoes the conversion from liquid to solid. The liquid properties are needed for the adhesive to fully wet the bonding substance, and the solid properties are needed for the strength required for the union of the final product. The mobility of an adhesive depends heavily on its own physical and chemical properties and those of the wood surface upon which it is being applied. To improve the interaction, wood is often resurfaced prior to bonding to provide a smooth surface with minimal extractives and other debris. Scientists question whether this step is necessary for wood modified by acetylation, which creates a more hydrophobic material.

The amount of adhesive penetration into a wood substrate has a direct correlation to the bond quality. Insufficient penetration causes minimal surface contact for chemical bonding or "mechanical interlocking." Over-penetration of adhesive will create "starved‘‘ or dry bond lines. Fluorescence microscopy is an excellent method of examining adhesive penetration into lumens. It is possible for most adhesives to fluoresce either in their natural form (primary or auto-fluorescence) or when treated with chemicals capable of fluorescing (secondary fluorescence), such as added dyes or pigments. Fluorescence microscopy in our study revealed that the degree of penetration was good for both acetylated and unacetylated wood despite the difference in bond durability. In addition. good lumen penetration did not correlate to poor strength previously observed, therefore leading to the conclusion that lumen penetration does not always relate with bond strength or percent wood failure.

Introduction

Penetration of adhesive deep into the capillary structure of wood is possible when good wetting conditions are achieved [1]. Creation of a bond between an adhesive and the wood substrate requires resin to sufficiently penetrate wood components and links to develop between the resin and the exposed wood surface (Figure 1) [2].

The mechanism of adhesion between the resin and wood components, which is still currently under debate, is generally thought to involve mechanical interlocking, covalent bonding, and secondary interactions, such as Van der Waals forces and hydrogen bonding [1]. Good penetration of the adhesive is promoted by excellent wood-to-adhesive–surface interaction and excellent adhesive mobility. In experimental efforts to improve the wood-adhesive interaction and provide a smooth surface with minimal extractives and machining debris, wood is often resurfaced prior to bonding.

Other variables also influence adhesive penetration, such as the material's characteristics and processing factors and methods of heating the adhesive bond [3]. Grain direction, permeability, porosity, roughness, surface energy, temperature, pressure, and time are among the other wood and processing factors that could influence the adhesive penetration [1, 4]. Variation in any of these factors can alter the quality and durability of the bond.

Sernek et al. [3] define adhesive penetration as the spatial distance from the interface of the adjoining substrate. The depth of the adhesive penetration determines the size of the interphase region, which Brady and Kamke [5] define as the volume containing both wood cells and adhesive. Care must be taken,

however, during adhesive wetting of the wood. According to Johnson and Kamke [6], over-penetration of adhesive will create “starved” or dry bond lines. Conversely, insufficient penetration causes minimal surface contact for chemical bonding or “mechanical interlocking” and will leave a thick film of adhesive on the surface. An ideal amount of adhesive penetration would repair machining damage to the wood surface and permit better stress transfer between laminates. Sernek et al. previously observed that adhesive penetration is optimum at a favored equilibrium moisture content [3]. Thus, to help identify sufficient penetration, it is critical to use a technique that will allow quantitative measuring.

Fluorescence microscopy provides an excellent method of studying a material that can be made to fluoresce, either in its natural form (primary or auto-fluorescence) or when treated with chemicals capable of fluorescing (secondary fluorescence). The basic principle is to permit excitation light to irradiate the specimen and then to separate the much weaker re-radiating fluorescent light from the brighter excitation light [7]. The objective of this experiment is to use fluorescence microscopy to determine the location of the adhesive in lumens, cell walls, rays, etc. and quantitatively measure the strength of acetylated and non-acetylated wood bonded with an epoxy adhesive.

Experimental

Yellow-poplar sapwood lumber was sawn into strips ¼ in. by 1 ¼ in. by 9 in. After cutting, the strips were placed in an oven and dried at 105°C for 24 h. The weight of each oven-dried strip was measured immediately after removal from the oven.

These strips were acetylated according to the following procedures. Separated by stickers, strips were placed into a stainless steel vacuum/pressure reactor. The reactor was filled with acetic anhydride to cover the strips initially and after they had absorbed much of the chemical. The acetic anhydride and wood was heated for 4 h and then cooled. Ships were removed, washed for 4 h in reversed osmosis water, air dried overnight, and then oven-dried for 24 h at 105°C. Weight gain caused by acetylation was determined by calculating a percentage of the original oven-dried weight of wood. All strips, including the untreated controls, were conditioned at 80°F and 65% relative humidity until bonded.

The acetylated wood and unmodified wood were bonded using four different commercial adhesives: resorcinol-formaldehyde (RF), melamine-formaldehyde (MF), polyamide-hardened epoxy, and an emulsion polymer isocyanate (EPI). The resins were similar to the adhesives used in a previous study [8]. The unmodified wood was planed according to standard practices; the acetylated wood was both planed and un-planed.

After bonding, block shear specimens were cut from the joint assemblies and randomly assigned for wet or dry shear tests according to ASTM D 905 [9]. The acetylation process and block shear results are explained in further detail in another study [10].

For image analysis, block shear samples were viewed as bonded assemblies and as failed shear samples that were cut to thin sections (ca. 25–30 µm) using a sliding microtome. The thin sections were dried in an oven at a temperature of 65°C for 30 min. After drying, some sections were stained in a 0.5% Saffranin O solution for no longer than 5 min. Sections were then mounted onto a microscope slide using glycerin. The samples were viewed under fluorescence using an epi-fluorescence (Zeiss Axioskop™) and a 100 W Mercury (HBO) lamp. The optical filter set used consisted of a 365-nm excitation filter, 395-nm dichromatic mirror, and a 420 emission filter. Digital images were taken with a Dage™ MTI CCD-72 video camera.

Results and Discussion

From the previous study of Frihart et al. [10], all four adhesives yielded good strength under dry conditions.

However, under wet conditions, the epoxy and MF adhesives gave poor wood failure for the control and acetylated wood while the EPI adhesive gave poor wood failure for the acetylated wood (Table I).

		Wood Failure (%)	
Wood Sample	Adhesive	Wet	Dry
Control	Epoxy	5.6	80.0
	MF	36.4	43.9
	EPI	91.7	91.7
	RF	98.6	97 2
Planed Acetylated	Epoxy	0	45.8
	MF	0	5.8
	EPI	0	98.9
	RF	94.7	98.0
Un-planed Acetylated	Epoxy	60.3	98.6
	MF	0	0
	EPI	0	69.7
	RF	91.4	96.4

Table 1: Wood failure for four different adhesives bonded on acetylated and un-acetylated wood under wet and dry conditions [10].

The epoxy adhesive had strong fluorescence that aided in examining penetration into both wood types Figures 2 and 3 illustrate that planed control and un-planed acetylated surfaces both showed good penetration into the vessel and some fiber-cell lumens. Figure 2 shows a planed control sample bonded with epoxy in which the wood is clearly distinguishable from the adhesive, and many of the lumens near the surface are filled with resin. The good penetration of the epoxy into both types of wood does not explain the difference between the percentages of wet and dry wood failure obtained for the epoxy-bonded assemblies.

The wood shown in Figure 4 was bonded with EPI and also shows penetration into the lumens. Compared to Figure 2, however, penetration does not go as deep into the lumens, and the bondline is much thinner. Both pictures show an adhesive bond with mechanical interlocking, hut the epoxy appears to have deeper penetration into the cell lumens, which would be expected to produce a stronger bond. On the contrary, a previous study showed [IO] that EPI samples yield good results for shear strength and wood failure under wet and dry conditions for the control wood. The epoxy samples give desired strength and wood failure under dry conditions but weaken significantly under wet conditions. Therefore. the correlation of penetration depth to strength is inconclusive.

The MF bondlines seen in Figures 5 and 6 were difficult to detect for this set of samples. The only method of detection relied on finding the adherend interface, which could be distinguished by different direction of wood rays. The abutment of the two wood pieces without any bondline indicated over-penetration of the adhesive. Staining the wood did not emphasize the adhesive; therefore, it was difficult to assess the degree of penetration or the causes of poor wood failure. The conclusion for over-penetration is also supported by the scanning electron microscopy images of the interfacial region. In contrast to MF, the RF bondline was very easy to recognize by the thick, dark film RF created on the surface. We were unable to explain, however, that the planed wood surface seen in Figure 7 shows an even bondline with penetration no more than two cells deep compared to the unplanned sample with penetration up to three cells deep (Figure 8).

Figure 9 is a fluorescent image of the planed control sample bonded with epoxy that was sheared under wet conditions in compression according to ASTM D 905. The adhesive seen in the bondline contains 1% of a fluorescent pigment that did not require this sample to be stained. Most interesting is the small piece of failed adhesive that extends out from the wood surface. At higher magnification, seen in Figure 10, it is much easier to see this cleavage as well as the small amount of fluorescence visible in the general wood cellular structure. This cell wall fluorescence probably arises from the lignin that is present in the middle lamella.

Conclusions

Fluorescence microscopy has once again proven to be a powerful tool in investigating the interaction between wood and a wood adhesive bond at the micro-level. In our work, we used fluorescence microscopy as a method of analyzing wood adhesive bonds and bond failure. A clear distinction between wood and adhesive can be seen when an adhesive bond has been made to fluoresce. Because several variables changed during our study, it was difficult to obtain definitive results on wood failure, hut insight can be gained on future work by examining the adhesive systems intently. Epoxy bonds are thick and fill cell lumens very well but do not yield consistent strength like EPI bonds, which are thin and do not fill lumens well. The MF bonds seem to over-penetrate at the bondline and do not fluoresce very well. The RF bonds appear to penetrate well and will fluoresce enough to distinguish the wood from the adhesive.

Future work will attempt to determine where adhesive flows and attempt to relate penetration measurements [3] to bond strength. This will possibly aid in determination of optimum bond surface

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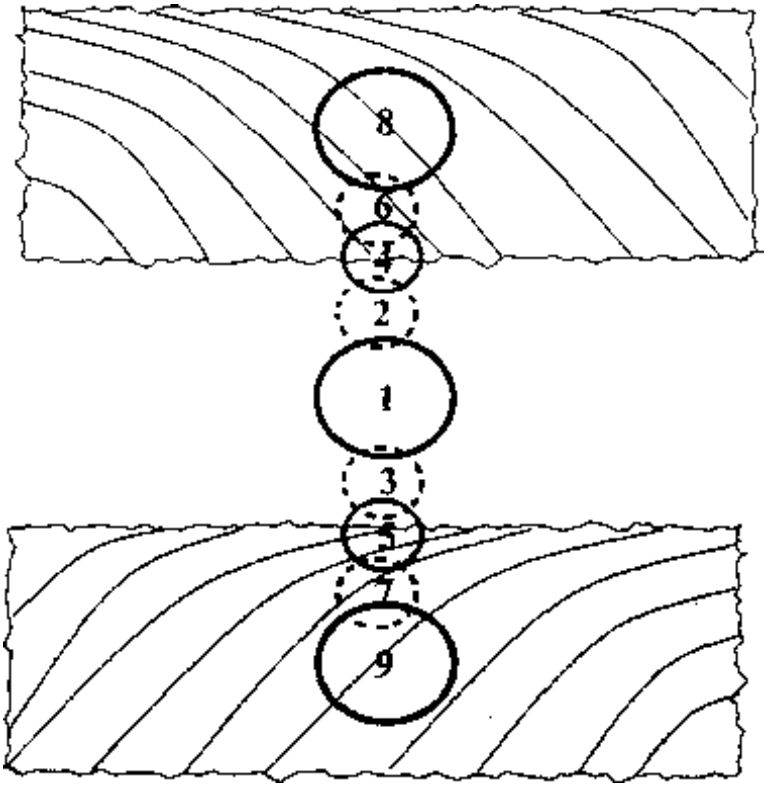

Figure 1: Links in an adhesive bond. Dashed links are most vulnerable to malformation.

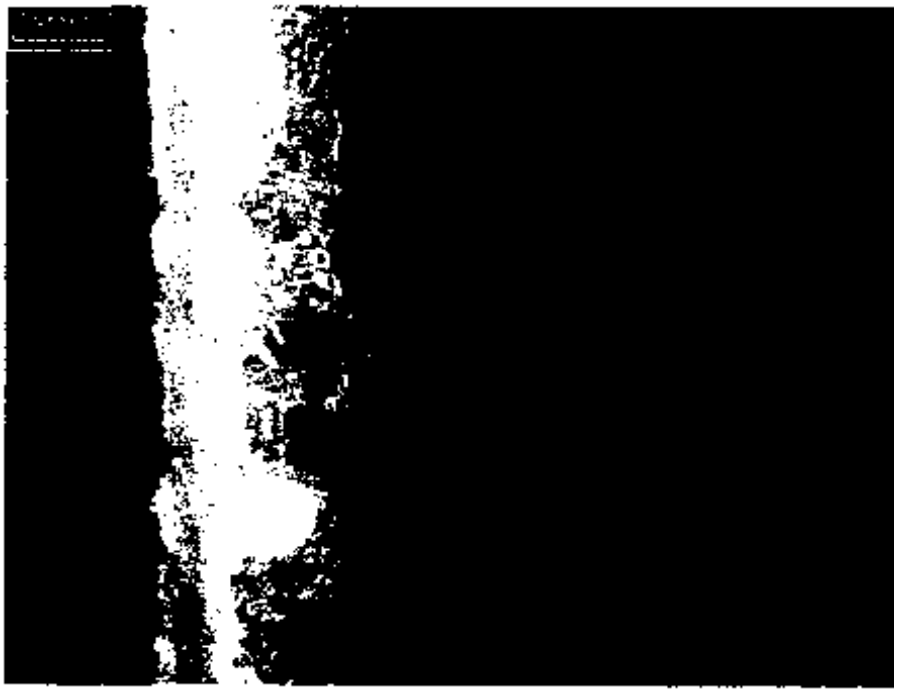

Figure 2: A planed control sample of yellow-poplar bonded with epoxy, viewed under a UV filter.

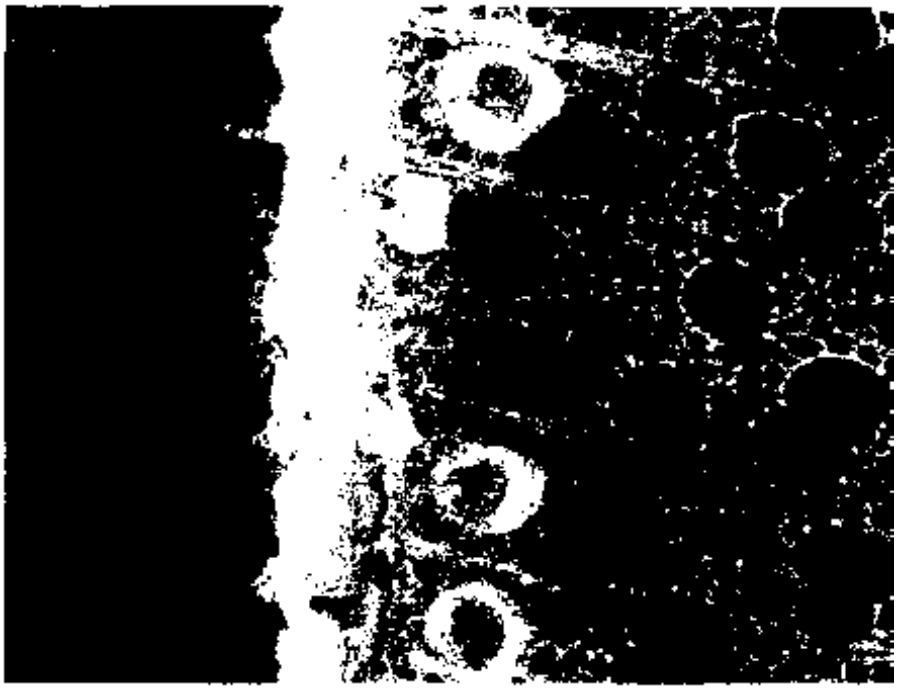

Figure 3: An un-planed acetylated sample of yellow-poplar bonded with epoxy, viewed under a UV filter.

Figure 4: A planed acetylated sample of yellow-poplar bonded with Emulsion Polymerized Isocyanate and stained with 0.5% Saffranin O, viewed under a blue filter.

Figure 5: A planed control sample of yellow-poplar bonded with Melamine-Formaldehyde and stained with 0.5% Saffranin O, viewed under a UV filter.

Figure 6: An un-planed acetylated sample of yellow-poplar bonded with Melamine-Formaldehyde and stained with 0.5% Saffranin O, viewed under a UV filter.

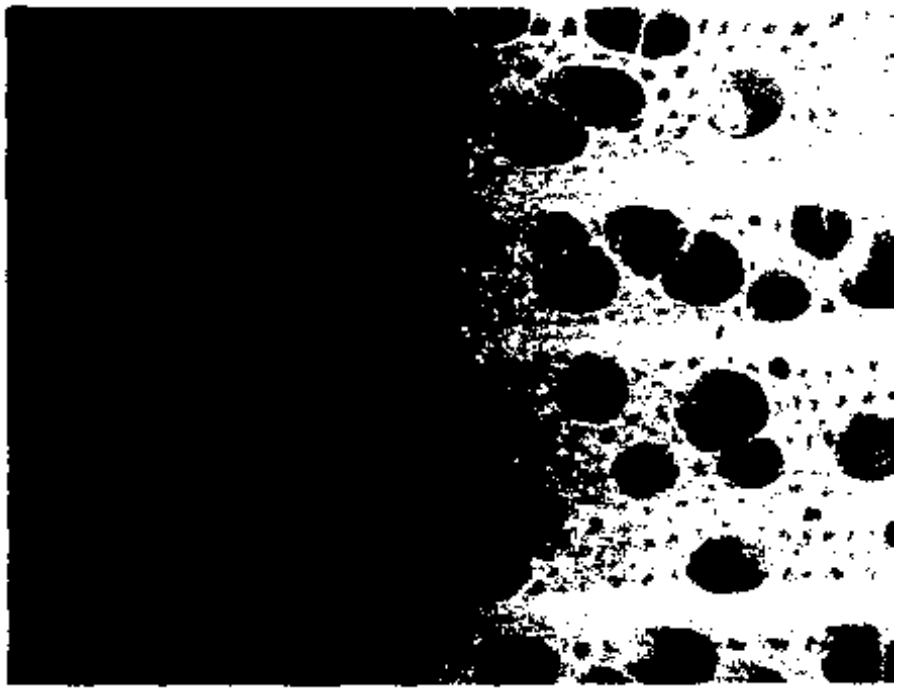

Figure 7: A planed acetylated sample of yellow-poplar bonded with Resorcinol-Formaldehyde, viewed under a UV filter.

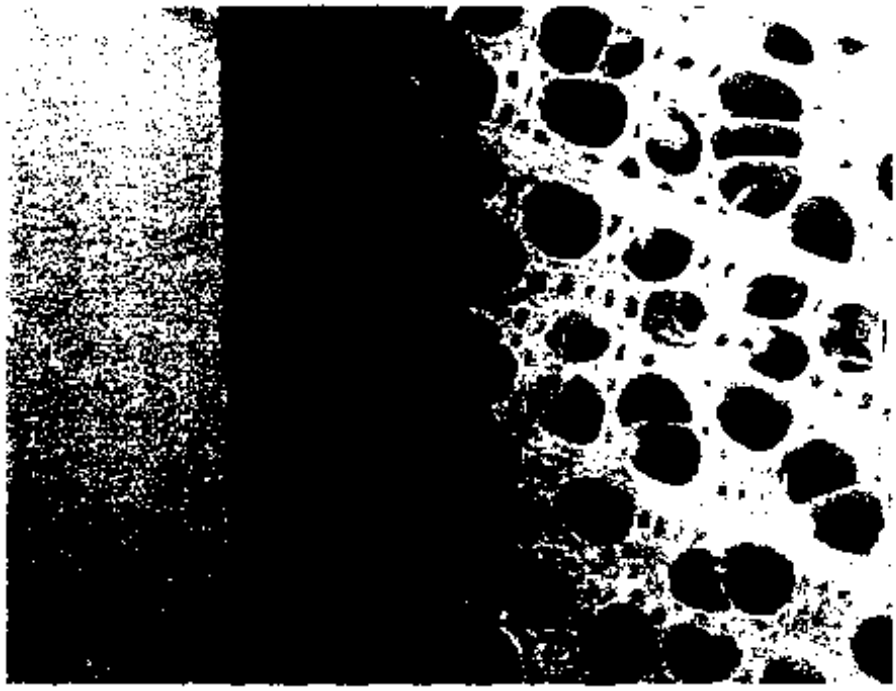

Figure 8: An un-planed acetylated sample of yellow-poplar bonded with Resorcinol-Formaldehyde, viewed under a UV filter.

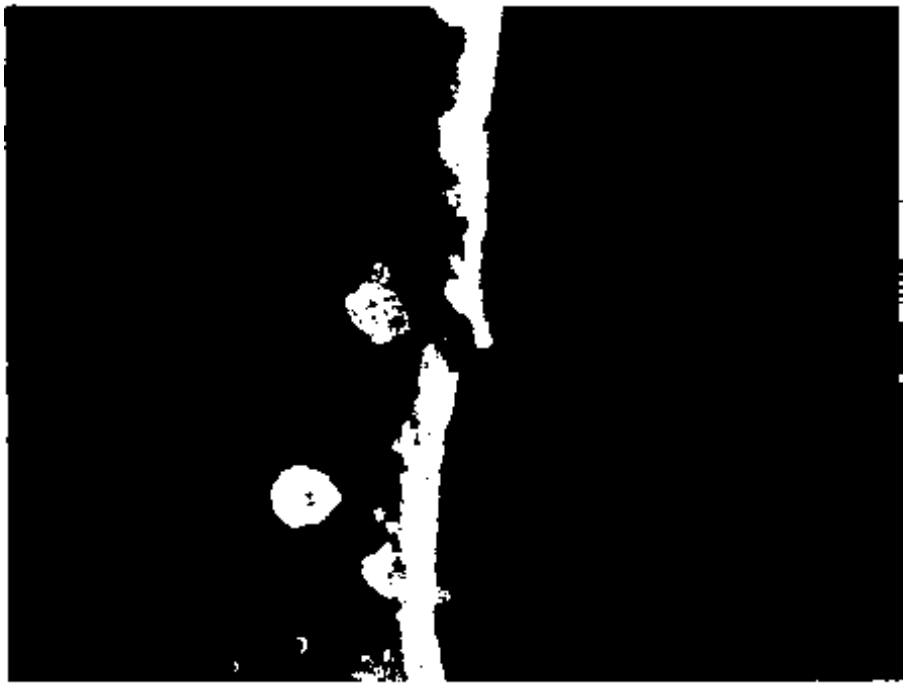

Figure 9: A cross-sectional view for failure surface of yellow-poplar bonded with Epoxy and 1% fluorescent pigmentm viewed under a UV filter. Image at 8 × of eye.

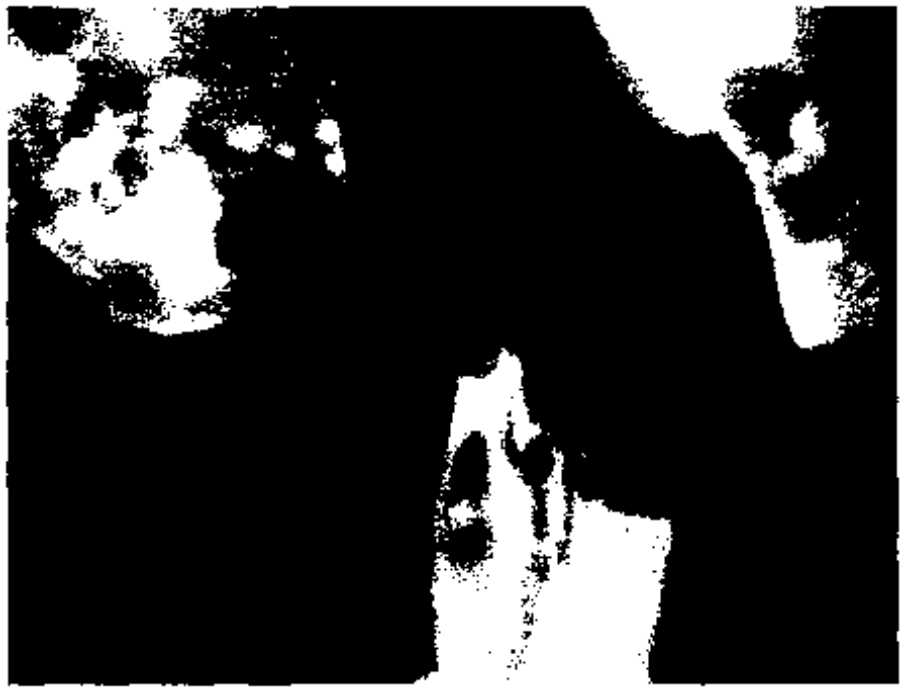

Figure 10: Cross-sectional view for failure surface of yellow-poplar bonded with Epoxy and 1% fluorescent pigment, viewed under UV filter. Image is Figure 9 at 50 × of eye.

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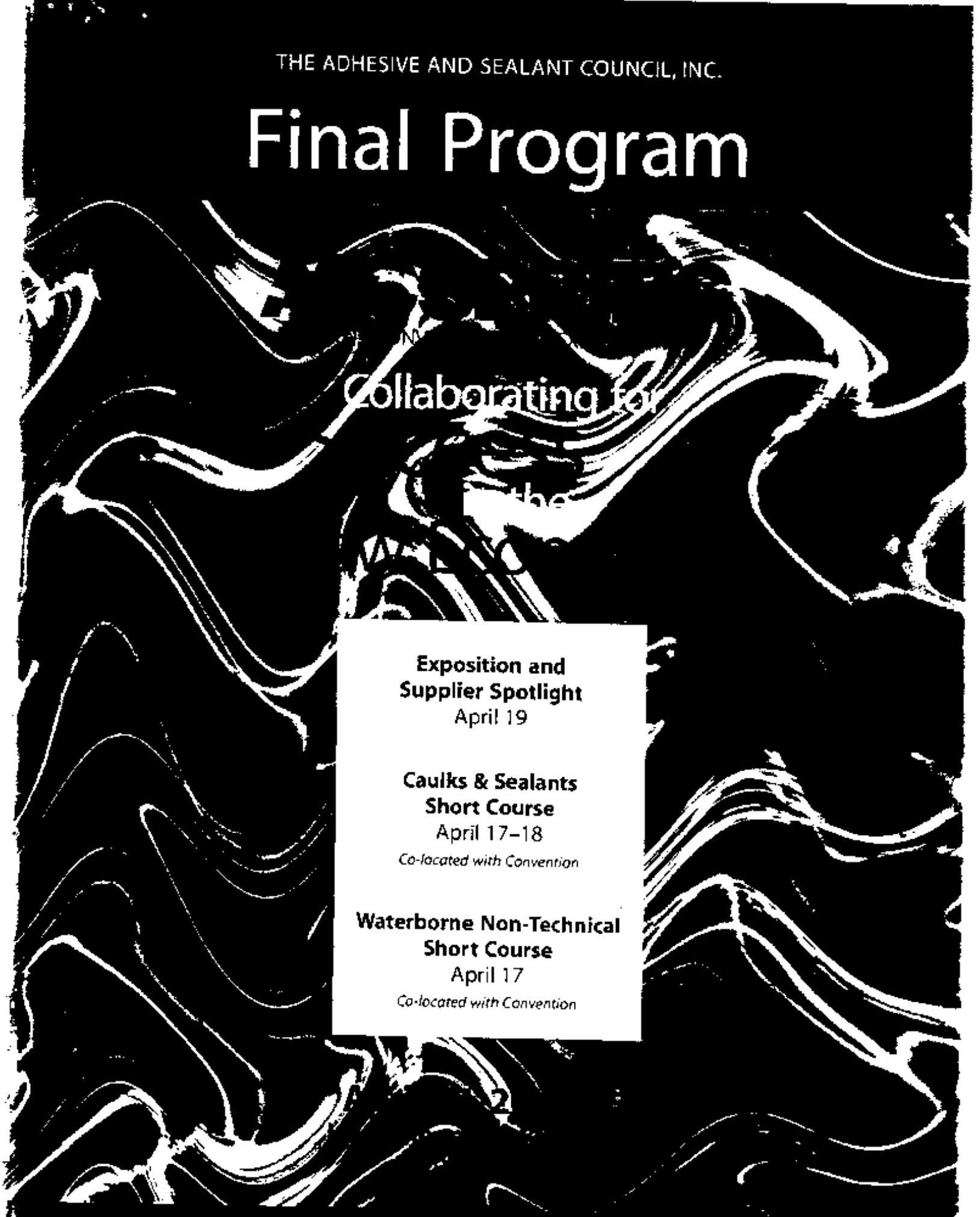

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