Correlation of Adhesive Performance between Automated Bond Evaluation System Tests and Plywood Tests: A Case Study of Lignin-Phenol-Formaldehyde Adhesives*

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Abstract

The automated bond evaluation system (ABES), which recently became ASTM D7998-15 standard test method, is an effective tool for screen testing of different water-based wood adhesive formulations. This method enables rapid evaluations of mechanical responsiveness of different adhesive formulations to various press temperatures and/or press times, providing an efficient and realistic comparison of bondability and reactivity among the adhesive formulations.

Based on extensive testing work, this article provides experimental findings and evidence for the use of this method to evaluate bonding performance of lignin as a major ingredient in the phenolic adhesive system. The relationship between bond strength development and press temperature can be established for a particular adhesive formulation using this method, which can then help the formulation and optimization of a wood adhesive containing lignin. Softwood plywood experiments demonstrated that there is a strong correlation between ABES test results and adhesive performance in the panel products.

Lignin is the most abundant aromatic biomaterial on Earth and is abundantly available as a by-product from the pulp and paper industry and the newly developing biofuel industry, which produces ethanol from biomass. Lignin has attracted a lot of global research interest for applications such as adhesives (Klašnja and Kopitović 1992, Kouisni et al. 2011, Feng et al. 2016), foam (Li and Ragauskas 2012, Pan and Saddler 2013), and carbon fiber (Xue et al. 2014, Mainka et al. 2015).

Wood adhesives are a key component of wood composites and engineered wood products, but they are mostly based on fossil resources. Lignin has been shown to be a viable partial substitution of phenol-formaldehyde (PF) adhesive resin (Kouisni et al. 2011, Feng et al. 2016), which is one of the most common adhesives for manufacturing wood composite and engineered wood products. However, the most challenging issue for the applications of lignin in the phenolic type of wood adhesives is its lower reactivity, which hinders productivity at a relatively high level of substitution of PF resins. In addition, there exist various types of lignin, e.g., kraft softwood lignin, kraft hardwood lignin, organosolv lignin, and hydrolysis lignin (H-lignin) from kraft process, thermomechanical pulping (TMP)–bioprocess, and supercritical hydrolysis. Each type of lignin might have different chemical properties, such as molecular weight and solubility, and exhibit different reactivity to formaldehyde and PF resin. Therefore, it is important to find an effective tool and method to evaluate the reactivity and bondability of lignin-substituted PF resin and screen the lignin type with best performance for engineered wood products manufacturing application.

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With the automated bond evaluation system (ABES), small adhesive bonds are precisely formed under rapidly increased temperature conditions and tested in lap shear mode immediately after selected formation times at stable temperatures. Repetition of this procedure for a range of temperatures enables adhesive bond strength development and strength maxima to be evaluated as a function of temperature. This method enables the mechanical responsiveness of different adhesive formulations to temperature to be evaluated and thus provides a realistic comparison of bondability and reactivity among the adhesive formulations. This method was developed into ASTM standard test method D7998-15 (ASTM International 2015). Figures 1 and 2 illustrate how ABES works.

The objective of this article is to give a few examples showing the advantage of using ABES in evaluating and screening different types of lignins used to partially substitute PF in oriented strand board (OSB) and plywood manufacturing.

Materials and Methods

Plywood PF and OSB PF resins are commercial products that were obtained from Canadian wood adhesive providers. Lignin samples were obtained from cooperating industrial partners and were produced using different processing techniques and under different production conditions.

Preparation of PF-lignin adhesives

The PF-lignin adhesives were prepared by mixing PF resin with alkaline lignin solutions on the basis of a designated ratio of solution to solids. All PF-lignin glue mixes were mixed well manually and kept at ambient temperature for 1 hour for ‘‘digesting’’ with occasional stirring at an interval of 10 min before subjecting the mixture to ABES tests.

Automated bond evaluation system

After conditioning at room temperature and 50 percent relative humidity, sliced maple veneers were cut into strands of 117 by 20 by 0.6 mm and used for the ABES tests. A glue mix was applied to a piece of wood strand in order to form a bonding area of 20 by 5 mm. Immediately after each bond was cured at the designated temperature for the designated press time, the glued wood sample was destructively tested in lap shear mode. The tensile load was monitored digitally during bond pulling and shear stress-to-failure (area-corrected peak load) was calculated. Five replicates were performed for each press condition. The bonding tests were performed at press temperatures ranging from 110°C to 150°C at 2 MPa pressure.

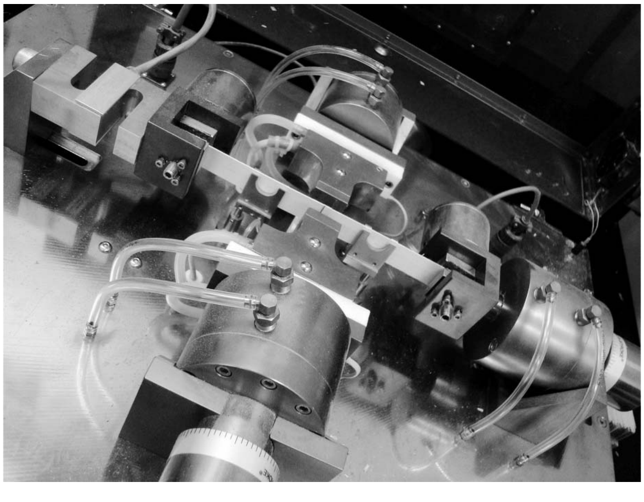

Figure 1.—Typical test sample mounted in an apparatus for rapid bond heating and pressing followed by bond pulling.

Plywood production and test

Three-ply plywood panels were manufactured with dry spruce veneers (moisture content, ~3%) for different PF-lignin adhesives and a typical commercial plywood PF glue mix containing 30 percent fillers. The PF-lignin adhesive or PF-fillers glue mix was manually applied on veneers with a paint roller. A small press was used for the panel pressing. Key parameters for the plywood production are listed in Table 1. The resulting plywood panels were tested according to the CSA O151-09 (Canadian Standards Association 2004) and ASTM D5266 (ASTM International 2013) standards.

Results and Discussion

Evaluation of reactivity and bondability of plywood PF substituted by FPInnovations' KL at different ratios

A type of kraft lignin (KL) with reduced sulfur odor was produced using a recently developed FPInnovations' patented process called the LignoForce System (Kouisni et al. 2012, 2016). This type of lignin has demonstrated low lignin odor, good solubility in water and diluted alkaline solution, and considerable reactivity with PF resins (Kouisni et al. 2011, Feng et al. 2016) and was successfully used in partially substituting PF resins in OSB and plywood manufacturing in pilot plant scale and industrial mill trials. Extensive work is continuously conducted in order to further increase the PF substitution ratio at the scale of industrial production.

In order to evaluate the relative reactivity and bondability of plywood PF substituted by FPInnovations' KL at different ratios, ABES was used to determine the bonding strengths of the formulations of commercial plywood PF with KL at 25 to 50 percent substitution ratios at 110°C to 150°C pressing temperatures (Fig. 3). Figure 3 shows that the reactivity of PF-KL formulations was lower than the commercial plywood PF control resin, exhibiting a decreasing trend with the increase of kraft lignin replacement ratio. When comparing the plywood PF control resin at 110°C

Table 1.—Plywood production conditions.

Parameter	Value
Spruce veneer thickness (mm)	3
Spruce veneer moisture content (%)	~3
Panel size (cm)	38.1 × 38.1
Panel type	Three-ply
Glue spread rate (g/m^2)	
Single glueline for PF-lignin adhesives	112.3
Single glueline for PF–fillers glue mix	146.5
Assembly time (min)	20
Press pressure (MPa)	1.38
Press temperature (°C)	150
Press times (s)	180, 210, 240
Replicates per condition	3

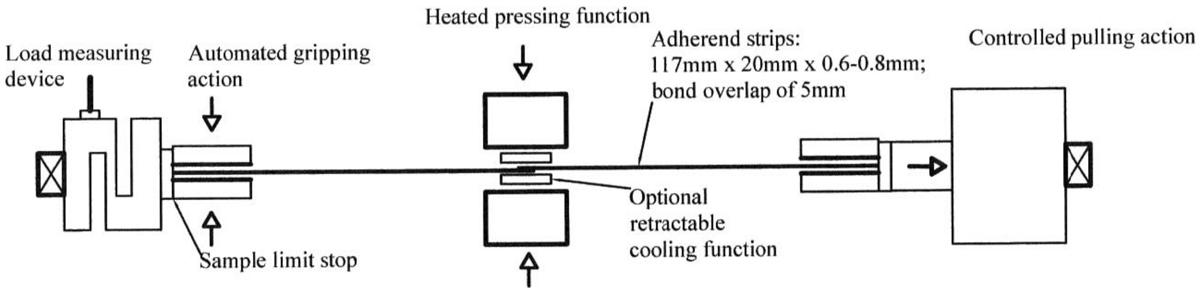

Figure 2.—Schematic of the bonding and testing concept. Reproduced with permission from Dr. Philip E. Humphrey, Adhesive Evaluation Systems Inc.

pressing temperature, the bonding strengths of PF-KL formulations decreased gradually with lignin additions from 25 to 50 percent. At 130°C pressing temperature, the bonding strengths of PF-KL formulations at 25 to 30 percent substitution ratios were comparable to the PF control resin, whereas the bonding strengths of PF-KL formulations at 37 to 50 percent substitutions were lower than the PF control. At the elevated pressing temperature of 150°C with up to 45 percent PF substitution, all PF-KL formulations had comparable bonding strengths to PF control, but the PF-KL formulation with 50 percent PF substitution exhibited a lower strength than the PF control.

The ABES test results indicate that the reactivity of plywood PF-KL formulations decreased gradually with a KL replacement ratio from 25 to 50 percent. Under the conditions of 150°C pressing temperature and 90 seconds pressing time, comparable bondability to that of control PF resin could be achieved for the PF-KL formulations with up to 45 percent replacement ratios.

Three-ply spruce plywood panels were produced at 150°C for different pressing times ranging from 210 to 270 seconds using the six PF-KL formulations in which 25 to 50 percent PF was substituted with lignin. A typical commercial plywood PF glue mix containing 30 percent fillers was used as the control. The plywood panels were tested according to the CSA O151-09 (CSA 2004) and ASTM D5266 (ASTM International 2013) standards.

Figures 4a and 4b show the percent wood failure test results after the plywood specimens were treated with water either under the vacuum–pressure (VP) condition or under the boil–dry–boil (BDB) condition. The VP test reflects water resistance of the glue lines, whereas the BDB treatment of shear specimen mimics accelerated aging, and the test indicates the durability of the adhesive bonding. According to CSA O151-09 (CSA 2004), the evaluation of softwood plywood is focused on percent wood failure. The softwood plywood industry requirement for percent wood failure is 80 percent minimum for both the VP test and the BDB test. Because higher wood failure means lower glue

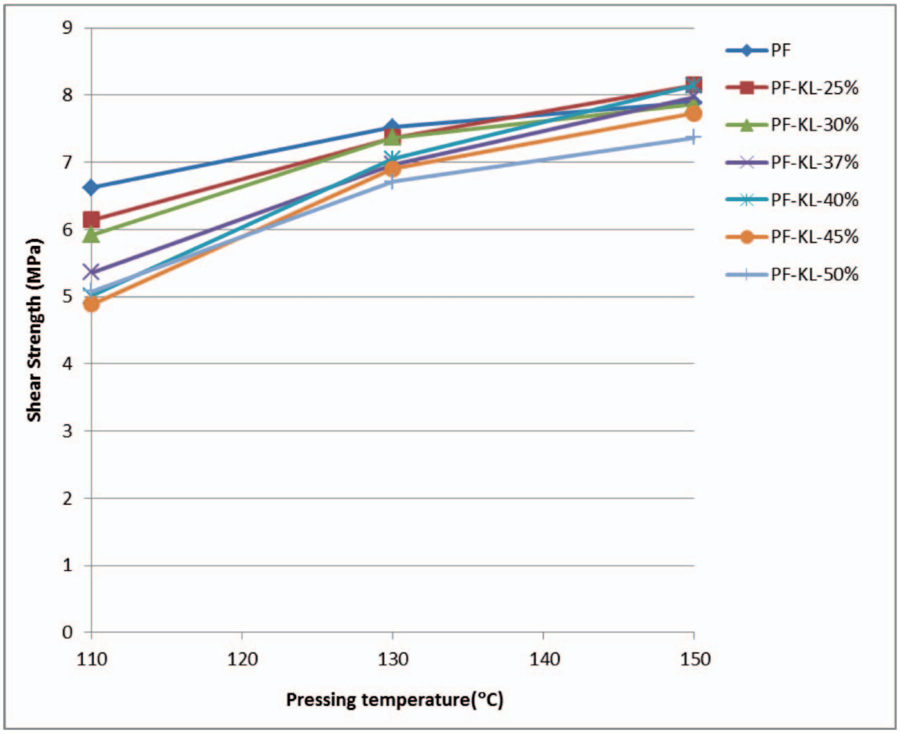

Figure 3.—Automated bond evaluation system bonding strengths of the formulations of plywood phenol-formaldehyde (PF) with FPInnovations' kraft lignin (KL) at 25 to 50 percent substitution ratios. (Color version is available online.)

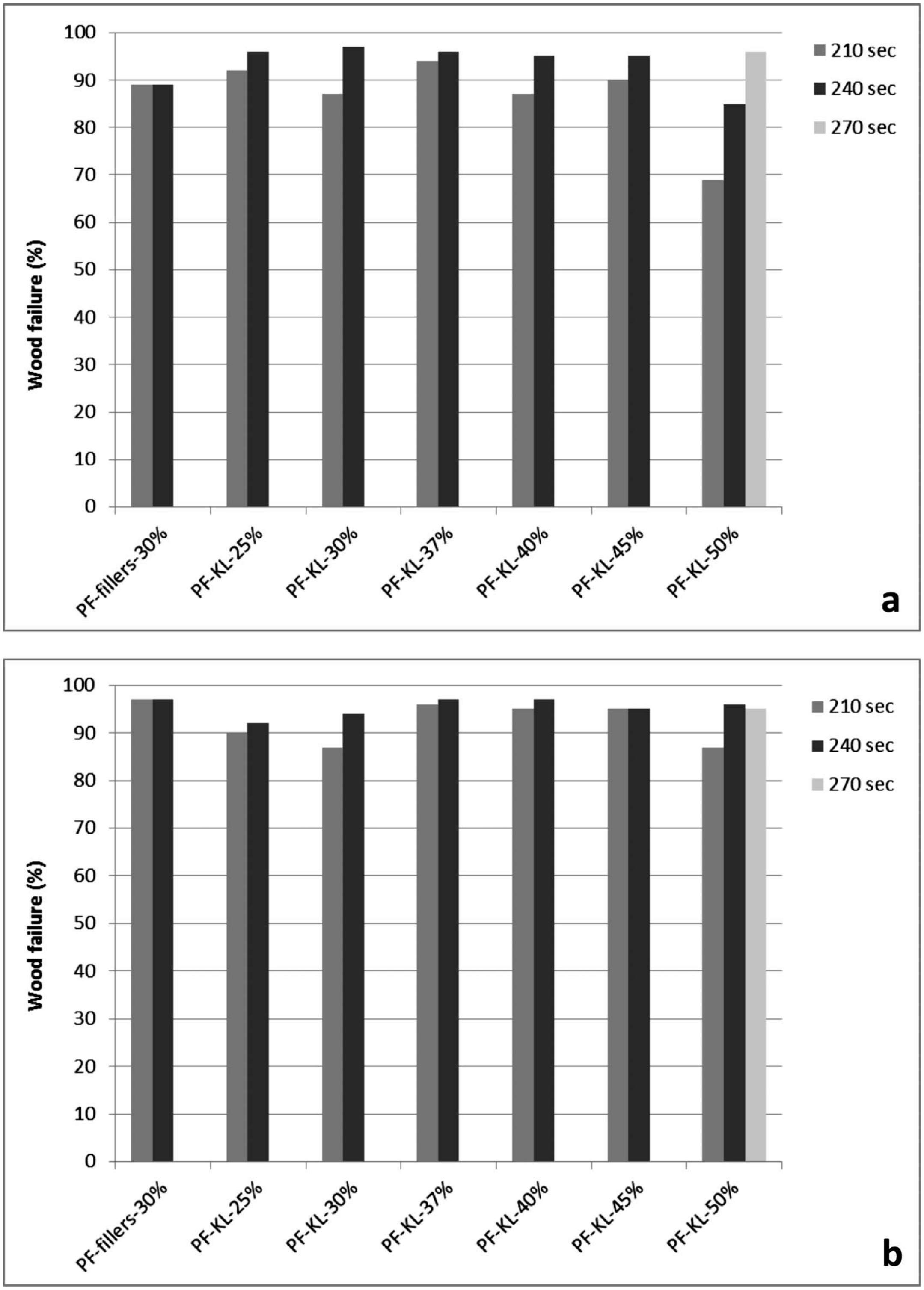

Figure 4.—Percent wood failure of plywood panels produced from the formulations of plywood phenol-formaldehyde (PF)–kraft lignin (KL) at 25 to 50 percent PF substitution ratios and from a typical plywood PF glue mix containing 30 percent fillers under vacuum-pressure treatment (Fig. 4a) or boil–dry–boil treatment (Fig. 4b).

failure, the higher the wood failures are, the better the test outcomes are.

As can be seen from Figure 4a, after VP treatment with up to 45 percent replacement, at 210 seconds pressing time, all PF-KL formulations had a comparable percent wood failure to that of PF–fillers–30% glue mix; at 240 seconds pressing time, the PF-KL formulations had a higher percent wood failure than PF–fillers–30% glue mix. At 210 seconds pressing time, the PF-KL formulation with 50 percent replacement did not reach the 80 percent wood failure requirement, but a prolonged pressing time of 240 to 270 seconds also resulted in satisfactory percent wood failure.

These results are very consistent with ABES test results shown in Figure 3.

After BDB treatment with up to 50 percent replacement, all PF-KL formulations had a slightly lower or comparable percent wood failure at 210 to 240 seconds or 270 seconds pressing times compared with PF–fillers–30% glue mix at 210 to 240 seconds pressing times. All percent wood failure values exceeded the 80 percent requirement for the softwood plywood industry. It was noted that after DBD treatment, the percent wood failure values for all PF-KL formulations were similar, and the percent wood failures were higher in the DBD test than in the VP test. This could be explained by further curing of the glue lines during the DBD treatment that provided extra heat energy.

Screening lignin types for the high performance in substituting plywood PF and OSB PF resins

In order to evaluate the relative bond performance of different lignin products in substituting commercial PF resins, the formulations of commercial plywood PF resin with 10 different types of lignin samples (named Lignin I, Lignin II, Lignin III, . . . , Lignin X) produced from different processing techniques and production conditions, and the formulations of commercial OSB PF with seven of these lignin samples, were tested using ABES. The PF substitution ratio by lignins was 30 percent.

The ABES bonding strengths of 10 plywood PF-lignin formulations at different pressing temperatures from 110°C to 150°C are illustrated in Figure 5. The test results

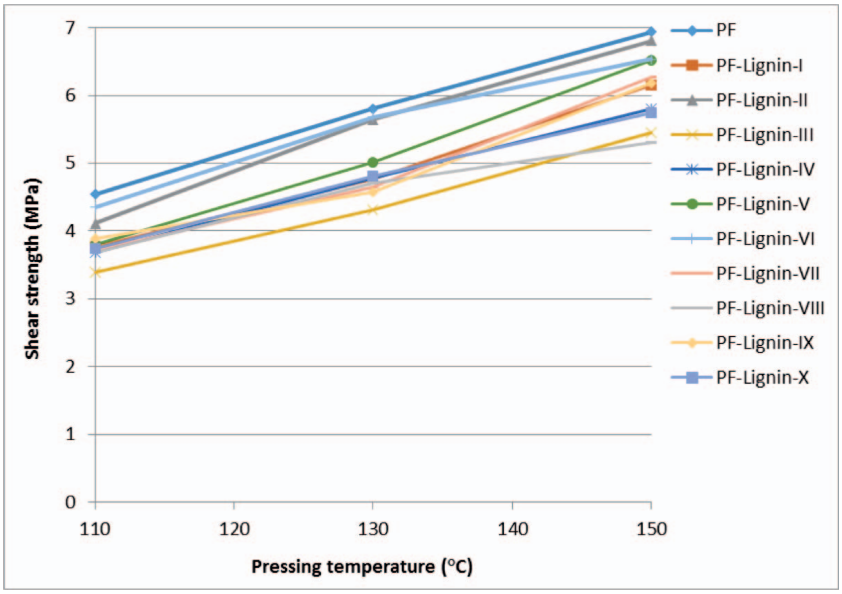

Figure 5.—Automated bond evaluation system bonding strength of the formulations of plywood phenol-formaldehyde (PF) with 10 different lignins. (Color version is available online.)

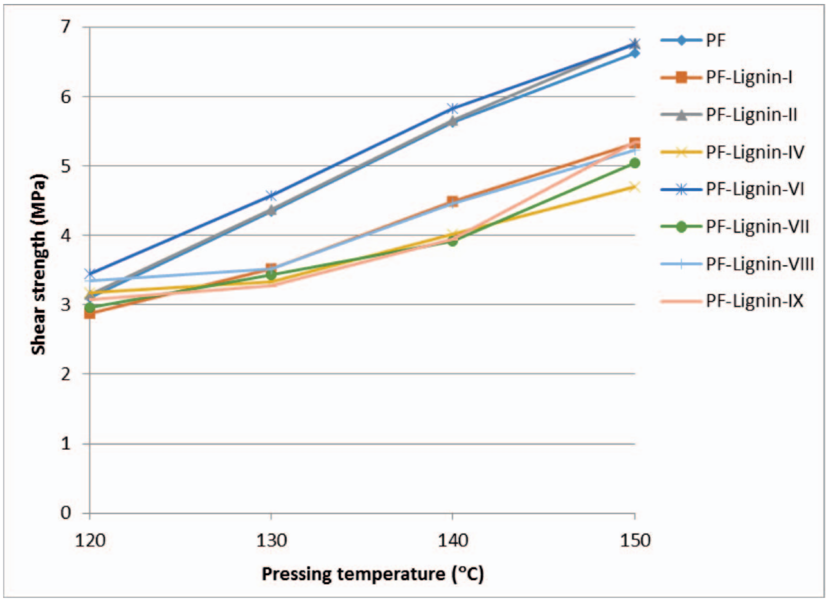

Figure 6.—Automated bond evaluation system bonding strength of the formulations of oriented strand board phenol-formaldehyde (PF) with seven different lignins. (Color version is available online.)

indicated that the bond performance of all PF-lignin blends with 30 percent replacement was lower or slightly lower than that of the PF control resin, but Lignin II and Lignin VI showed the best performance among the 10 samples studied, and Lignin III showed the lowest performance.

The relative bond performances of seven OSB PF-lignin formulations at different pressing temperature from 120°C to 150°C evaluated by ABES are illustrated in Figure 6. As in the experiments with plywood PF-lignin formulations, Lignin II and Lignin VI again showed the best performance. Moreover, in contrast to the plywood formulations, the bond performance of their blends (with 30% replacement) was very close to the OSB PF control resin at all test temperatures (120 to 150°C). The other lignins showed much lower performance and could not provide acceptable bondability even at more severe conditions (150°C).

Because Lignin II and Lignin VI exhibited the best performance in both plywood and OSB PF formulations, they were selected for further studies, and the major focus was on Lignin II because it was most representative of current products. The sample of Lignin I was used for comparison representing those lignins with low performance according to ABES test results shown in Figures 5 and 6. Through OSB and plywood panel production experiments, it was found that the performance of OSB panels (regarding internal bond, modulus of elasticity, modulus of rupture, etc.) and plywood panels (regarding percent wood failure) using Lignin II was higher or much higher than Lignin I at 20 to 30 percent PF substitution ratios under the same pressing conditions (the data are not listed here because of commercial confidentiality), which is very consistent with ABES test results.

Conclusions

It is convenient to determine the bonding strength of lignin-PF adhesives at different pressing temperatures for target pressing times using ABES. More importantly, ABES makes it possible to test and compare the reactivity and bondability of different lignin-PF formulations in parallel and to screen the formulations rapidly. Experimentally, a good correlation was observed between the ABES test results and the performance of composite panel products pressed using the tested lignin-PF adhesives.

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