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Use of organosolv lignin in phenol-formaldehyde resins for particleboard production

II. Particleboard production and properties

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Abstract

The objective of this work was to demonstrate the utility of lignin-based resins designed for application as an adhesive in the production of particleboard. Bond qualities of lignin-phenol-formaldehyde resins, phenolated-lignin-formaldehyde resins and commercial phenol-formaldehyde (PF-com) resin were assessed by using an automatic bonding evaluation system, prior to production of particleboards. In order to evaluate the quality of lignin-based resins, particleboards were produced and physical and mechanical properties were investigated. These physical properties included internal bond, modules of rupture and modulus of elasticity. Thickness swell and water absorption properties of particleboards bonded with lignin-based resins were also determined. The lignin-based resins have been reported previously in Part I of this study. The results showed that particleboards bonded with phenolated-lignin formaldehyde resins (up to 30% lignin content) exhibited similar physical and mechanical properties when compared to particleboards bonded with PF-com. The work has indicated that phenolated-lignin formaldehyde resins (up to 30% substitution level) can be used successfully as a wood adhesive for constructing particleboard. The performance of these panels is comparable to those of boards made using PF-com resin.

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1. Introduction

Wood particleboard is manufactured from lignocellulosic wood material, in the form of discrete particles, combined with synthetic resin and bonded together under heat and pressure in a hot press [1]. In the process, the majority of the interparticle bonds are created by the added binder. In particleboard, 95–98% of all particleboard is flat pressed and the particles lie with their longitudinal axis randomly oriented but essentially parallel to the board surface. As the quality of particleboard continues to rise to meet particular requirements, greater control on chip geometry is required, together with a higher degree of sophistication in the lay up and subsequent pressing of the board. Thus, it is normal to find that the rather coarse chips in the board are produced from waste timber, while the thin flat particles in the surface layer originate from round wood. The type of adhesive used, the density of the board, level of strength and the degree of surface smoothness vary with the use to which the board is going to be put [2].

Bonding of thermosetting adhesives plays an important role within wood-based composites. During hot pressing, there is a continuously changing state of factors such as temperature, moisture content and vapour pressure within the mattress, which affects the process of adhesion and the development of bond strength. This affects the properties of the final product, the required hot-pressing time and energy consumption [3].

In commercial practice, the amount of PF resin used is generally 5–15% w/w. Even at these low concentrations, resin cost is the overriding factor affecting final product cost [1,4] and there have been many attempts to replace phenol with lignin in PF resins suitable for particleboards in order to reduce resin cost. Roffael and Dix [5] looked at employing sulphite spent liquor in the

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particleboard industry with a major focus being on resin properties. Sulphite spent liquors have been employed along with diisocynates and tannin resins as extenders with binders, but these require long pressing times and high temperatures. The employment of lignin sulfonate in the adhesion process made it possible to leave out 50% of the expensive PF resin, and substitute the cheaper sodium sulphite liquor, without lowering the physical and chemical properties of the boards [5]. The utilisation of kraft lignin from *Pinus radiata* black liquor, as a copolymer in LPF resin binders were studied. Different modified lignins were tested in the preparation of LPF resin binders. The mechanical properties and water resistance was evaluated through testing particleboard panels manufactured with the resins obtained. The best resin had properties comparable with typical commercial PF resins [6]. Lignosulfonates blended either with a PF resin in a separate step during the synthesis, or mixed with the resin, accomplish satisfactory cross-linking during the hot pressing of the final product. Panels like plywood, particleboard and fibreboard were successfully produced using the LPF resin, although the resin required longer press times or higher press temperatures compared with commercial PF resins [7,8].

The objective of this study was to investigate the extent to which organosolv lignin could replace phenol in PF resins designed for application as an adhesive in particleboard production. The production and evaluation of particleboards bonded with lignin-based formaldehyde resins were made. The mechanical properties and dimensional stability of particleboards manufactured with the lignin-based resins were measured and comparisons were drawn between the different resin types. The lignin was substituted for up to 30% of the phenol in the phenolated-lignin resins.

2. Materials and methods

2.1. Bond strength of resins

The bonding ability of the resins was examined in a lap joint test using an automatic bonding evaluation system (ABES) tensile tester. This equipment is designed to enable the bond strength development characteristics of a diverse range of adhesives to be explored, while varying the parameters. Thin (heat diffusive) adherent birch veneer (wood strips) of thickness of 0.8 mm, length 104 mm and width 20 mm were used. Resin was applied to an area of 4 mm × 20 mm on one side of one end section of the two pieces (Fig. 1). Resin coated, overlapped end sections of a series of specimens (pair of strips) were pressed together in a hydraulic press at a pressure of 5 kg cm^{-2} using a range of curing periods (30, 60, 90, 120, 180, 240 s) at 180°C. At the end of the selected curing period, and while still hot, each bond was rapidly (within seconds) cooled and tested to failure in tension. For each bond forming condition (temperature and pressing time), five replicates were conducted.

2.2. Preparation of particleboards

Industrial woodchip furnish, comprised of a variety of species, principally softwood, was used for producing test panels. The resins used for manufacture of boards were PF-com and lignin-based resins. No resin hardener or wax was added. Resins were applied at a rate of 10% (resin solids based on the oven dry weight of wood particles). Particles were dried to approximately 3% moisture content prior to application of resin. Board target thickness was either 6 or 12 mm; target density was 650 kg m^{-3} in all cases. Test samples were prepared and conditioned at 65±5% RH and 20±2°C until weight stability was reached. After conditioning, sample weight and dimensions were determined. All tests were carried out to the appropriate European Standards. The tests performed on the specimens were internal bond strength perpendicular to the plane of the board (IB) [9], static bending [modulus of rupture (MOR) and modulus of elasticity (MOE)] [10], thickness swelling and water absorption [11] and boil test [12].

3. Results and discussion

3.1. Bond strength development analysis of lignin-based resins

The bond strength development of the lignin-phenol-formaldehyde resins prepared with various organosolv

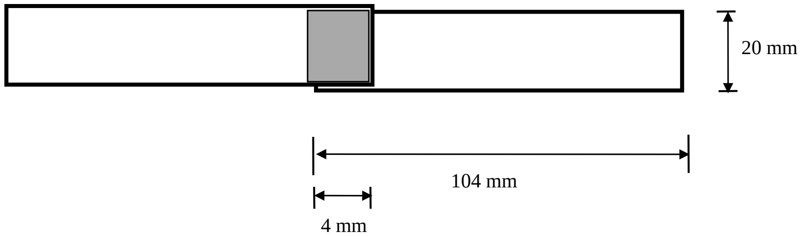

Fig. 1. Lap-joint specimen.

Table 1
The bond strength values of lignin-phenol-formaldehyde resins (pressed at 180°C)

Time (s)	Failure stress (N mm^{-2})					
	PF-com	Resin 5 (5%)	Resin 10 (10%)	Resin 20 (20%)	Resin 30 (30%)	Resin 40 (40%)
30	5.05	4.3	3.9	4.32	2.93	1.45
60	5.96	6.6	5.7	6.24	3.5	1.83
90	6.7	6.7	6.2	6.56	4.2	2.10
120	7	6.9	6.4	6.94	4.94	2.13
240	6.9	6.9	6.6	6.9	6.2	2.74

Table 2
The bond strength values of LPF 20, LPF 25 and LPF 30 resins (pressed at 180°C) using lap-shear test

Time (s)	Failure stress (N mm^{-2})		
	LPF 20	LPF 25	LPF 30
60	6.22	5.55	5.18
90	6.73	6.64	6.50
120	No failure	No failure	6.74
180	No failure	No failure	6.56
240	No failure	No failure	No failure

lignin contents (5%, 10%, 20%, 30% and 40%) were analysed using the ABES tester. Initial experimental results showed that there was no bond strength deterioration observed up to a 10% lignin substitution level. Resins 5 and 10 (5% and 10% substituted respectively) had similar bond strength values and showed the same trends to PF-com resin (Table 1).

Resin 20 (20% replacement of phenol) cured at a faster rate and developed high bond strength. Resin 20 was equivalent in bond performance to the standard PF-com resin at 180°C. Increasing the press cycle time beyond 60 s did not improve the bond strength for Resin 20 at this temperature. For wood strips, this work indicated that 20% of phenol in the PF resin could be replaced with organosolv lignin without detrimentally affecting adhesive properties. However, as can be seen from Table 1, Resin 40 (40% phenol replacement) exhibited a dramatically low failure stress, compared to the other resins. This resin showed a distinctly poorer performance.

The bond strength values of phenolated-lignin formaldehyde resins (LPF 20, LPF 25 and LPF 30) (≈20–30% substitution with phenolated-lignin and 41% formaldehyde solution) are illustrated in Table 2. The bond strength of these resins built up faster, and achieved higher final bonding at 60 s. Failure stress reached ≈6.50 N mm^{-2} in a short time. There was an adhesive failure in these samples. There was no bond failure observed for these three resins after 90 s of bond formation. During the experiment, in some cases, it was also noted that before the bond failed, the veneer strips failed (material failure) with most of the LPF 20, LPF 25 and LPF 30 bonded strips. Since there was no bond failure between the strips, it is an example of how strong an interaction it was, although high temperatures were needed.

To see the effect of lignin substitution level on the bond strength, the failure stresses of LPF 20, LPF 25, LPF 30 are plotted and shown in Fig. 2. It can be seen from Fig. 2 that the higher the lignin content, the poorer the bond quality. However, the bond strength is still tolerable, at 60 s press cycle time and 180°C press temperature. In addition, when the press cycle time was increased, the high lignin content resins gave better bond strength, similar to the PF-com resin. The performance of all three of these phenolated-lignin-formaldehyde resin formulations was adequate with respect to a standard or by comparison with PF-com.

3.2. Physical and mechanical properties of experimental panels

3.2.1. Internal bond strength

The bonding ability of resins made with different additions of organosolv lignin Resin 20, Resin 40, LPF 20, LPF 25 and LPF 30 was tested and compared with that of a PF-com resin at 180°C at 15 min press time (6 and 12 mm particleboards). The internal bond data of the particleboards made using these resins are illustrated in Fig. 3. Increasing levels of lignin in the resin resulted in decreasing IB values. Particleboards of 12 mm made at 180°C press temperature and 15 min press time using Resins 20 and 40 showed a reduction in IB value of 21% and 33%, respectively, compared to PF-com resin. Similar trends were observed in 6 mm thick boards. The measured IB values of boards bonded using with phenolated-lignin-formaldehyde resins are also summarised and compared with those of boards bonded PF-com resin in Fig. 3. The IB of 12 mm boards made with LPF 20 (20% lignin) were equal to or better than those bonded with PF-com resin. The IB of boards bonded with LPF 20 resin was significantly ($P<0.05$) higher than that of boards bonded with LPF 30 resin. Typically, IB tended to decrease with increasing addition of lignin. Particleboards bonded with LPF 30 resin

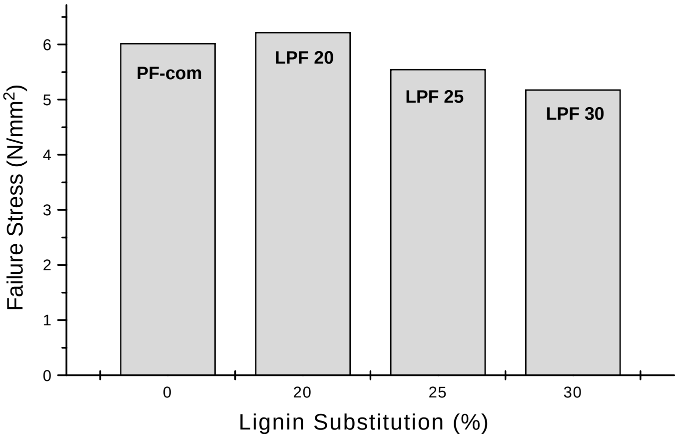

Fig. 2. Effect of lignin substitution in phenolated-lignin formaldehyde resins (press cycle time was 60 s and press temperature was 180°C).

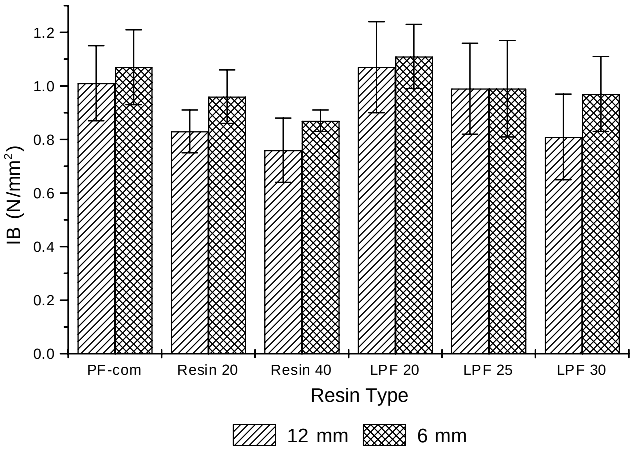

Fig. 3. The internal bond strength of 12 and 6 mm thick boards made with lignin-based resins.

(30% lignin) showed 20% reductions in the IB values compared to particleboards bonded with PF-com resins. It was evident that increasing lignin substitution level had a more detrimental effect on IB values for 12 mm boards compared with 6 mm thick boards. On the basis of the data it can be concluded that, with respect to IB, phenolated-lignin-formaldehyde resins can be used successfully as adhesives in particleboards. Satisfactory strength can be achieved at substitution levels of lignin for phenol up to 30%.

3.2.2. *Modulus of elasticity (MOE)*

The measured MOE of panels bonded with PF-com and lignin-based resins are summarised in Fig. 4. The presence of lignin had little influence on the MOE of 12 mm boards. For 6 mm boards the presence of lignin in resins typically resulted in increases in MOE, with the exception of Resin 40, where a reduction was observed.

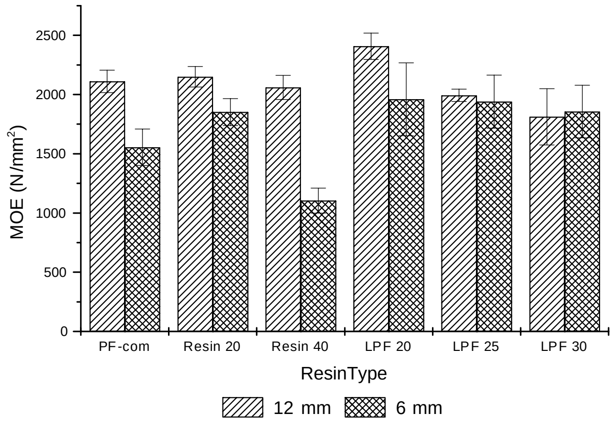

Fig. 4. MOE values of 12 and 6 mm thick boards.

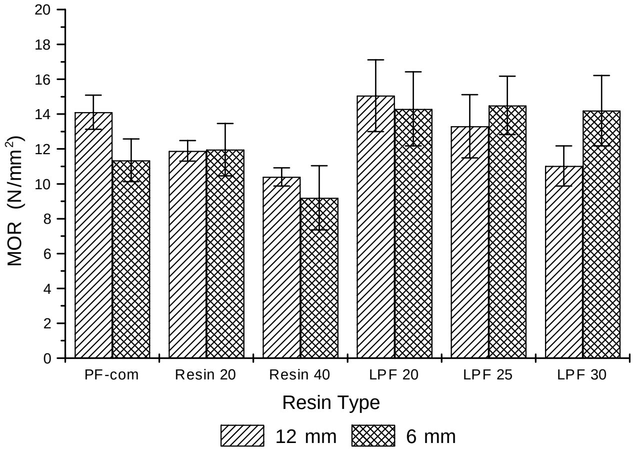

Fig. 5. MOR values of 12 and 6 mm thick boards.

3.2.3. *Modulus of rupture (MOR)*

Fig. 5 shows the effect of lignin substitution level in resins on the MOR of panels. Boards of 12 mm bonded with PF-com exhibited better bending strength (at least 18% higher) than both Resins 20 and 40. Boards of

6 mm bonded with Resin 20 exhibited similar MOR strength values to boards bonded with PF-com resin. Boards of 6 mm bonded with Resin 40 exhibited significantly lower MOR compared to all other resins. There was no significant ($P > 0.05$) difference in MOR between 12 mm panels bonded with LPF 20 and PF-com. For 12 mm panels it was evident that as the lignin substitution level increased, MOR tended to decrease. In 6 mm thick panels, lignin appeared to have little significant influence on MOR.

3.2.4. *Thickness swelling and water absorption*

Figs. 6 and 7 show the effects of resin type on the thickness swelling of after 2 h and 24 h immersion in water. For 12 mm thick panels, there were modest increases in swelling as the lignin content of resins increased. Thickness swelling was largely unaffected by the presence of lignin in resins in 6 mm thick panels.

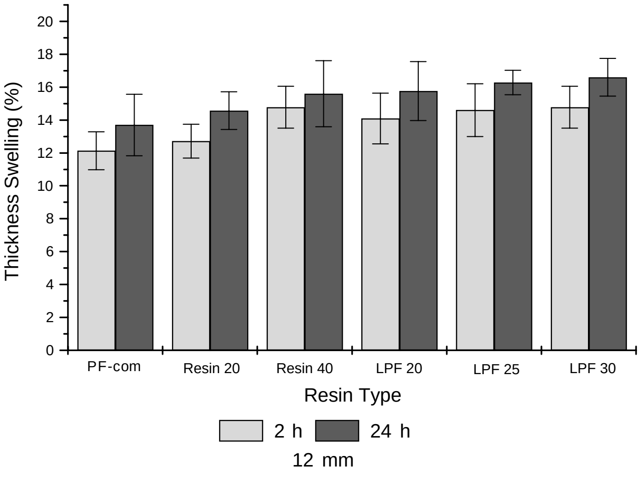

Fig. 6. Thickness swelling of 12 mm thick boards.

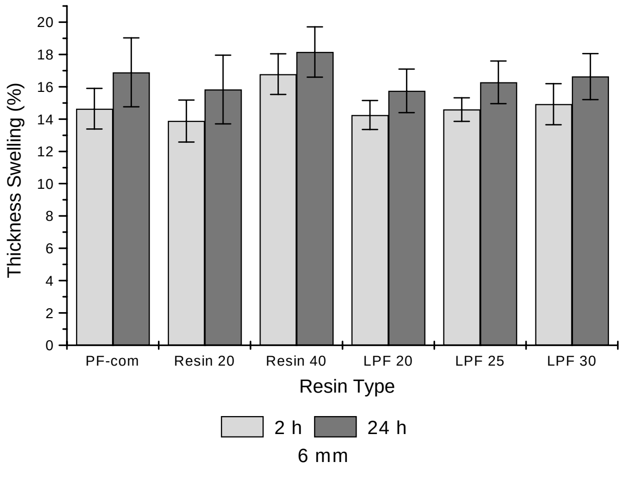

Fig. 7. Thickness swelling of 6 mm thick boards.

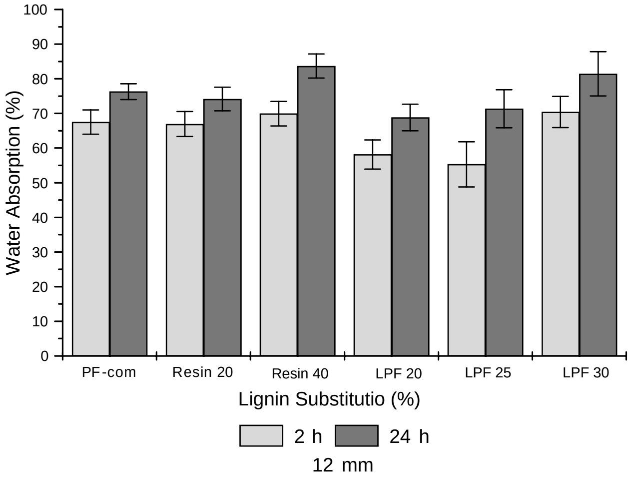

Fig. 8. Water absorption of 12 mm thick boards.

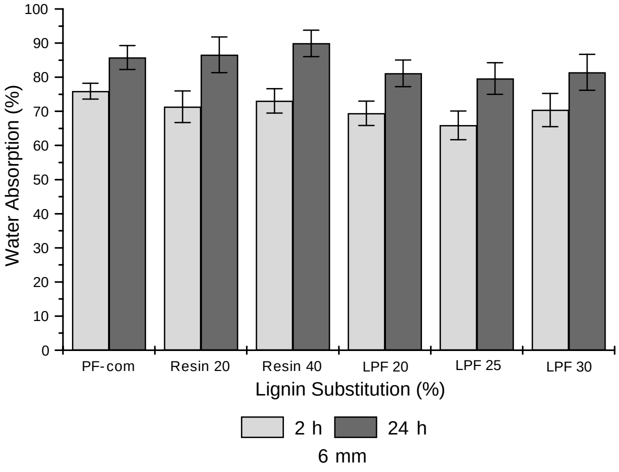

Fig. 9. Water absorption of 6 mm thick boards.

Table 3
Internal bond values before and after 2 h boil test

	IB (N mm^{-2})			
	PF-com	Resin 20	LPF 20	LPF 25
Before boiling	1.01	0.83	1.07	0.99
After boiling	0.45	0.31	0.43	0.41
Retention (%)	45	37	40	41

The water absorption of panels is shown in Figs. 8 and 9. Apart from 12 mm panels bonded with LPF 20 and LPF 25, which absorbed less water than PF-com bonded panels, water absorption appeared to be relatively unaffected by the presence of lignin in resins.

3.2.5. *Two hour boil test*

Data for retained IB after a 2 h boil test are summarised in Table 3 and Fig. 10. Panels bonded with Resin 20 resins exhibited poorer retention of strength

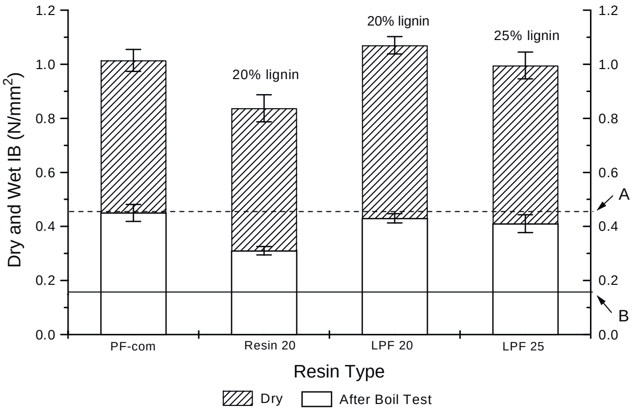

Fig. 10. Influence of the resin type on IB strength after boil test. Arrows A and B show the required minimum IB value of load bearing boards in dry conditions and load bearing boards in humid conditions after boil test [13,14].

compared with panels bonded with the commercial resin. However, panels bonded with phenolated-lignin resins showed comparable performance to the panels bonded with the commercial resin.

4. Conclusions

Phenolated-lignin-formaldehyde resins impart better mechanical and physical properties than substitution by unmodified lignin. Properties comparable to those of boards bonded with PF-com resin were achieved in boards bonded with resins with up to 30% substitution of phenol by phenolated-lignin. There is a relationship between increased addition of lignin and reduction in board mechanical properties. However, it has been demonstrated that organosolv lignin could be used to replace 20–30% of the phenol in PF resins used to bond particleboards, without adversely affecting bond properties. Particleboards of 12 mm bonded with Resin 20, LPF 20, LPF 25 exhibited comparable mechanical properties to particleboards bonded with PF-com resin. IB values of 12 mm boards bonded with Resin 20, LPF 20, LPF 25 gave statistically similar results in dry tests as well as comparable IB strength retention values with PF-com after boil test.

The work has indicated that phenolated-lignin-formaldehyde resins (up to 30% substitution level) can be used successfully as a wood adhesive for constructing particleboard. The performance of these panels is comparable to those of panels made using PF-com resin. The study demonstrates that renewable resources such as lignin have potential as a substitute for phenol in PF resin systems.

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