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## Properties of particleboard produced with liquefaction-modified phenol-formaldehyde adhesive

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**Abstract:** *Pinus densiflora* wood flour was liquefied in the presence of phenol, using sulfuric acid as a catalyst. Liquefaction-modified phenol-formaldehyde (LPF) resins were synthesized as particleboard binder. The physical characteristics of the laboratory-synthesized control PF resin and the LPF resins are described. Laboratory particleboard (PB) was made using Korean hybrid poplar (*Populus alba* × *glandulosa*) particles with the control PF resin and LPF resins. The physical strength and dimensional stability of the PBs were tested according to ASTM D 1037-99 and KS F 3104 standard procedures. PB test results show that the physical and mechanical strength properties differed significantly according to resin type; however, all PBs made in this study exceeded the minimum strength requirement of the Korean Standard KS F 3104 for PB type 8.0.

**Key words:** Particleboard, hybrid poplar, adhesive, resin

### Introduction

Currently, there is renewed interest in the development of abundant, renewable, biomass resources for the production of polymeric materials. According to the Food and Agriculture Organization of the United Nations, yearly growth of world forests is approximately 7-9 billion m<sup>3</sup> of biomass (FAO 1966). Lignocellulosic biomass, such as wood and bark, can be liquefied easily in a variety of organic solvents using acid catalysts to produce homogeneous liquefaction materials (Alma et al. 2001; Lee and Liu 2003). These liquefied wood solutions consist of many carbohydrate- and lignin-derived components with useful applications. Liquefied wood solutions have been used successfully as wood adhesives (Lee and Liu 2003).

Phenol-formaldehyde (PF) resin is a thermosetting resin traditionally used as a binder of wood composite products, such as plywood, oriented strand board (OSB), and fiberboard, on account of its excellent durability, water resistance, and thermal stability. In addition, wood-based panels bonded with PF resins have almost no formaldehyde emission problem; however, PF resin prices are closely related to the price of phenol, which is derived from petroleum, and its price has increased during the past decade. The increase in PF prices has led to research into alternative sources of adhesives or chemical feedstock (Lee and Liu 2003).

Several studies have been reported on the substitution of phenol in PF resins with lignocellulosic biomass materials, such as lignin, tannin, and

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carbohydrates, in the search for new methods of preparing PF resin (Alma et al. 2001; Lee and Liu 2003). A phenol-liquefied wood solution was investigated as a partial replacement for phenol in PF resins (Lee and Liu 2003); however, the parameters for synthesizing PF resins require optimization for wood composite products, such as plywood, OSB, and particleboard (PB).

*Populus* species have been planted in Korea since the early 1960s. The Korean hybrid poplar *Populus alba* × *glandulosa* covered an area of more than 10,000 ha in 1973 (FAO 1979). Until 1975, more than 20,000 ha of *Populus* species were planted each year, and more than 70,000 ha were planted annually from 1976 to 1980 (Koo 2001). In total, over 935,000 ha of *Populus* species have been planted to date (Koo 2001).

The present study compared the properties of PBs bonded with liquefaction-modified phenol-formaldehyde (LPF) resins with those of panels bonded with a laboratory-synthesized control PF resin adhesive. The Korean hybrid poplar *Populus alba* × *glandulosa* was used as the raw material for PB.

**Materials and methods**

**Liquefaction of wood**

*Pinus densiflora* wood flour (-100-mesh) was used as raw material for liquefaction. Liquefaction was carried out with phenol (99%), using 3 different weight charge ratios of phenol:wood (72:28, 67:33, and 62:38) based on oven-dried wood weight (Lee and Liu 2003). Sulfuric acid (95%) was used as the

catalyst at 2.8%, based on the phenol weight, in all experiments. Phenol was charged into a stirred reactor and heated to 60 °C. After adding the sulfuric acid with continuous stirring, wood flour was added over a period of 30 min. The reaction temperature was increased to 165-170 °C for 1 h and the temperature was maintained for 1 h until liquefaction was complete. The liquefied wood solution was cooled and stored until needed.

**Determination of the formaldehyde reactivity of the liquefied wood**

The formaldehyde reactivity of the liquefied wood solution was determined using the method described by Wooten et al. (1988). The number of moles of formaldehyde that reacted with 50 g of a liquefied wood solution at 60 °C in the presence of a catalytic amount of NaOH was determined over a 4-h period. The amount of free formaldehyde in the sample was determined using the hydroxylamine hydrochloride method (Walker 1964). For the purpose of LPF resin synthesis, the number of moles of formaldehyde that reacted after 2 h was selected as the relevant amount to use in the condensation step of LPF resin synthesis.

**PF resin synthesis**

Table 1 summarizes the PF resin formulation procedure used in the present study. A control PF resin was formulated in the laboratory with a phenol:formaldehyde mole ratio of 2.1 and a sodium hydroxide:phenol mole ratio of 0.36. The general cooking procedure was similar to that outlined by Oh et al. (1994) and Oh (2009).

Table 1. Control PF and LPF resins formulations used in this study.

|                    | PF resin<br>(control) | LPF resin I<br>(72/28) | LPF resin II<br>(67/33) | LPF resin III<br>(62/38) |
|--------------------|-----------------------|------------------------|-------------------------|--------------------------|
| ---- (g) ----      |                       |                        |                         |                          |
| H <sub>2</sub> O   | 119                   | 174                    | 209                     | 244                      |
| Phenol (99%)       | 377                   | -                      | -                       | -                        |
| Liquefied solution | -                     | 400                    | 400                     | 400                      |
| NaOH (50%)         | 116                   | 152                    | 160                     | 168                      |
| HCHO (37%)         | 676                   | 562                    | 519                     | 476                      |
| Urea               | 38                    | 38                     | 38                      | 38                       |
| Total              | 1326                  | 1326                   | 1326                    | 1326                     |

Three LPF resin types (I, II, and III) were prepared from 3 phenol-liquefied wood solutions (3 different phenol:wood ratios): PF resin type I from a phenol:wood ratio of 72:28, PF resin type II from a phenol:wood ratio of 67:33, and PF resin type III from a phenol:wood ratio of 62:38 (Table 1). Three different LPF resin types were similarly synthesized with the control PF resin as follows.

Water, sodium hydroxide (50% solution), and a phenol-liquefied wood solution were charged into a stirred reactor and the reaction mixture was heated to 65–70 °C. A formaldehyde solution (37% conc.) was then added dropwise to the reaction mixture over a period of 30 min, while keeping the reaction temperature in the same range by intermittent cooling using an ice bath. After the addition of formaldehyde, the reaction temperature was maintained in the same range for approximately 10 min, and then was increased gradually to 85 °C over a period of 30 min. The reaction temperature was maintained until the resin reached “H” viscosity on the Gardener-Holdt (G-H) viscosity scale. A small amount of urea was added to the resins at the end of synthesis, which were then cooled to room temperature and stored until property analysis and use.

### Resin analysis

The viscosity of the resins was measured using a Brookfield RVF viscometer, spindle number 1 at 2.09 rad s<sup>-1</sup> (20 rpm) of rotation. The free formaldehyde

content was measured using the hydroxylamine hydrochloride method (Walker 1964). Resin solids levels were determined by heating 1 g of resin on an aluminum pan at 125 °C for 2 h.

### PB manufacture

*Populus alba* × *glandulosa* wood particles (12–3.5-mesh) were dried to a 4%–5% moisture content prior to use. Single-layer homogeneous PBs were manufactured using the processing parameters shown in Table 2.

### PB performance test

Test specimens were cut from boards. Internal bond (IB), modulus of elasticity (MOE), and modulus of rupture (MOR) values were determined in accordance with the American Society for Testing and Materials (ASTM) Procedure D 1037-99 (2002). Panel water absorption and thickness swelling properties were observed after 24 h of soaking.

### Statistical analysis

Panel property test results were analyzed using the Statistical Analysis System (SAS) package program (SAS Inst. 1994). Analysis of variance (ANOVA) was used to determine the differences between the panels bonded with each resin type. Significant differences ( $P < 0.05$ ) were further compared using the t-test for least significant differences (LSD) in the SAS program (Steel and Torrie 1980).

Table 2. Panel manufacturing parameters.

| Parameter            | Conditions                                  |
|----------------------|---|
| Panel dimension      | 250 × 250 × 6.3 mm                          |
| Mat moisture content | 8% to 9%                                    |
| Wax & resin content  | 1% and 7%, based on oven-dry wood weight    |
| Catalyst             | None  |
| Resin flow rate      | 120 mL min <sup>-1</sup>                    |
| Target density       | 705 kg m <sup>-3</sup>                      |
| Press temperature    | 190 °C                                      |
| Press time           | 5 min                                       |
| Replication          | 4 boards per condition (total of 16 boards) |

**Results**

**Reaction of formaldehyde with liquefied wood solution**

The phenol-liquefied wood solution for formaldehyde was determined to formulate LPF resins for use in bonding PB. The phenol:wood (62:38) liquefaction solution had significantly lower formaldehyde reactivity than the other 2 liquefaction solutions (Table 3).

**Resin properties**

The PF resins made in the present study had very low free formaldehyde content (less than 0.25%), as expected (Table 4). The resin solids level ranged from 43.2% to 47.1%. The resin viscosity ranged from 150 to 220 mPa s, which is suitable for resin application with a compressed air sprayer.

**PB performance test**

The PB density in this study ranged from 708 to 715 kg m<sup>-3</sup> (Table 5). LSD test results show that panel density was not significantly affected (P < 0.05) by resin type.

The IB of all panels ranged from 0.478 to 0.902 N mm<sup>-2</sup> (Table 5). The IB of the panels bonded with the control PF resin was higher than that of the panels bonded with the LPF resins. LSD test results show that IB differed significantly according to resin type. The control panel (bonded with the control PF resin) had the highest IB value in the present study; however, the panels bonded with the 3 LPF resins (LPF resin type I, type II, and type III) had equivalent IBs.

The MOE range for all panels was 2307 to 2733 N mm<sup>-2</sup> (Table 5). LSD test results for MOE show that the control panels were significantly different from the panels bonded with 3 LPF resin types; however, the MOE values of the panels bonded with the 3 LPF resin types were similar.

The MOR for all panels ranged from 15.3 to 18.9 N mm<sup>-2</sup> (Table 5). LSD test results for MOR show that the control panel was similar to the panels made from the 3 LPF resin types; however, there were no significant differences in MOR between the panels made with 3 LPF resins. These differences suggest that the control PF had the best bending properties (MOE

Table 3. Formaldehyde reactivity of a phenol liquefied wood solution at 60 °C.

| Reaction time (hours) | LPF resin I (72/28) | LPF resin II (67/33) | LPF resin III (62/38) |
|-----------------------|---------------------|----------------------|-----------------------|
|                       | ---- (mole) ----    |                      |                       |
| 1                     | 1.72 <sup>a</sup>   | 1.73                 | 1.70                  |
| 2                     | 1.76                | 1.78                 | 1.72                  |
| 3                     | 1.74                | 1.76                 | 1.69                  |

<sup>a</sup> Average of 2 values.

Table 4. Properties of control PF and LPF resins synthesized.

|                       | PF resin | LPF resin I | LPF resin II | LPF resin III |
|-----------------------|----------|-------------|--------------|---------------|
| Solid content (%)     | 46.3     | 47.1        | 43.2         | 45.2          |
| Free formaldehyde (%) | 0.20     | 0.21        | 0.18         | 0.24          |
| Alkalinity (%)        | 4.18     | 4.15        | 4.26         | 4.58          |
| Viscosity (mPa s)     | 150      | 160         | 220          | 210           |

Table 5. Test results of particleboards made from control PF and LPF resins synthesized.

| Resin type         | Density<br>(kg m <sup>-3</sup> ) | Internal bond                   | MOE    | MOR    | 24-h Thickness swell | 24-h Water absorption |
|--------------------|----------------------------------|---------------------------------|--------|--------|----------------------|-----------------------|
|                    |                                  | ---- (N mm <sup>-2</sup> ) ---- |        |        | ---- (%) ----        |                       |
| PF resin (control) | 715 A                            | 0.902 A                         | 2733 A | 18.9 A | 18.1 B               | 42.3 B                |
| LPF resin I        | 711 A                            | 0.478 B                         | 2307 B | 15.3 B | 23.0 A               | 51.0 A                |
| LPF resin II       | 708 A                            | 0.510 B                         | 2407 B | 16.0 B | 21.9 A               | 48.6 A                |
| LPF resin III      | 710 A                            | 0.483 B                         | 2345 B | 15.8 B | 24.3 A               | 51.4 A                |

MOE: Modulus of elasticity, MOR: Modulus of rupture.

Means with the same capital letter are not significantly different (\*P < 0.05).

and MOR) among all the panels tested. MOE and MOR decreased as the quantity of liquefaction solution increased.

Thickness swelling of all the panels ranged from 18.1% to 24.3% in the 24-h test (Table 5). LSD test results for thickness swelling after 24-h water soaking show that the control panel had a significantly higher thickness swelling value than the panels made from the 3 LPF resins. On the other hand, the panels made from the 3 LPF resins had equivalent 24-h thickness swelling values.

Water absorption for all the panels ranged from 42.3% to 51.4% in the 24-h test (Table 5). LSD test results for water absorption after 24-h water soaking show that the control panel absorbed significantly less water than panels made from the 3 LPF resins; however, the 24-h water absorption values of the panels made from the 3 LPF resins were similar.

## Discussion

The 60 °C reaction temperature of formaldehyde with liquefied wood solution used in the present study favors the formation of hydroxymethyl groups. However, methylene groups can form at a low rate with extended reaction times (Wooten et al. 1988). The formaldehyde reactivity of the liquefied wood solution was relatively high (more than 1.1 mol of formaldehyde), which is similar to lignin from some pulping sources (Wooten et al. 1988). In the present study the amount of formaldehyde that reacted for 2 h was used as the relevant value for resin synthesis formulation.

All the tested properties of the resins were in the acceptable range for bonding PB. Generally, for industrial use a small amount of urea is added during the final stage of control PF and LPF resin synthesis in order to capture the residual free formaldehyde without impairing resin performance, and to decrease resin viscosity and cost. Thus, resins can be polymerized to a higher molecular weight than otherwise possible using a given synthesis procedure (Sellers 1985; Oh et al. 1994).

The panel density variation was quite low in the present study, suggesting that the panel mat forming operation was suitable. Uniform mats are essential for comparing the bonding performance of fabricated panels. Normally, panel density positively affects the strength properties of panels (Sellers 1985).

It is well known that, after hot pressing, panel hot stacking probably improves IB (Sellers 1985); therefore, IB differences between panels bonded with the 3 LPF resins could be reduced by hot stacking. However, the panels made in the present study exceeded the minimum strength requirements for IB and MOR, as per Korean Standard KS F 3104 for PB type 8.0 (KSA 2002).

The differences in thickness swelling and water absorption observed in the present study were attributed to the presence of extraneous materials in the cured LPF resin matrix of the PBs, such as carbohydrate-derived components, which can adversely affect the dimensional stability (thickness swell and water absorption) of PBs.

## Conclusions

Liquefaction-modified phenol-formaldehyde (LPF) resins were synthesized as PB binders and compared with a laboratory-synthesized control PF resin. The Korean hybrid poplar *Populus alba* × *glandulosa* was used for raw material to fabricate PB.

Performance test evaluations show that the PBs that were bonded with synthesized resins exhibited good physical and mechanical properties; however,

the physical and mechanical properties differed significantly according to resin type. Overall, the LPF resins can be used successfully as PB binders. The Korean hybrid poplar *Populus alba* × *glandulosa* is also a suitable raw material for the manufacture of PB.

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