FORMULATION OF LIGNIN PHENOL FORMALDEHYDE RESINS AS A WOOD ADHESIVE

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Abstract

This work describes the potential of reducing phenol with lignin in phenol formaldehyde resin formulation. The physical and chemical properties between lignin phenol formaldehyde resin (LPF) and commercial phenol formaldehyde resin (CPF) were compared. Phenol had been replaced by lignin [that was extracted from black liquor of oil palm empty fruit bunch (EFB)] in synthesizing resin with a ratio lignin to phenol 1:1. The IR spectra showed that there were similarities in functional groups between LPF resin and CPF resin. The comparison of physical strength properties via tensile strength test between LPF resin and CPF resin showed that the newly formulated resin has higher bonding strength compared to commercial resin. Kinematics viscosity test showed that LPF resin has lower kinematics viscosity compared to CPF resin in 21 days storage time. SEM images for both resin showed similarities in the effect of resin penetration into wood's vessel existed.

Abstrak

Kajian ini dijalankan untuk mengkaji potensi lignin sebagai pengganti kepada fenol dalam penghasilan resin fenol formaldehid. Seterusnya perbandingan sifat fizikal dan kimia dikaji antara resin lignin fenol formaldehid dengan resin fenol formaldehid komersial. Lignin yang diekstrak daripada likuor hitam tandan kelapa sawit kosong telah dijadikan sebagai pengganti kepada fenol dalam penghasilan resin dengan nisbah lignin-fenol 1:1. Spektrum IR bagi resin lignin fenol formaldehid menunjukkan persamaan kumpulan berfungsi dengan resin fenol formaldehid komersial. Perbandingan kekuatan tensil melalui ujian kekuatan resin menunjukkan resin lignin fenol formaldehid mampu membuat pengikatan yang lebih kukuh dengan substrat berbanding dengan resin fenol formaldehid komersial. Daripada ujian kelikatan kinematik yang dilakukan menunjukkan resin lignin fenol formaldehid mempunyai kelikatan yang lebih rendah berbanding dengan resin fenol formaldehid komersial walaupun setelah melalui proses penyimpanan selama 21 hari. Imej SEM bagi kedua-dua resin menunjukkan corak perekatan yang hampir sama dan wujud kesan penembusan resin ke dalam liang vesel kayu.

Introduction

Adhesives are required in many woods processing industries as such as particleboard, wood panels, fiber board and plywood. Among the various adhesives used for the production of these articles, phenol formaldehyde (PF) resin is the most common type of adhesive [1]. With the increasing use of composite materials such as strand board, flake board and composite lumber, the demand for adhesive is increasing. These adhesives are in adequately supply today, but the scarcity of petroleum products could affect the future cost and availability of these petroleum based adhesive [2].

To avoid this scenario, it is imperative to explore the possibility of replacing petroleum based raw materials. Therefore, investigations aimed at using natural products as substitutes for phenol gained favor on not only environment but also economic grounds. Extensive studies have been made on the use of lignin as a PF resin substitute. Lignin has an aromatic and highly cross-linked structure, similar to the network of PF resins. They may reduce the cost of PF resins because of the high cost and toxicity of phenol.

Experimental

The raw material used in this study was black liquor. It is a waste substance from the pulping process of oil palm empty fruit bunch (EFB) long fiber supplied by School of Industrial Technology, Universiti Sains Malaysia.

During the pulping process, the pH of black liquor was measured as 12.45. The soda lignin was precipitated from the concentrated black liquor by acidifying it to pH 2 using 20 % sulfuric acid. The precipitated was filtered and washed with pH 2 water, which was prepared using the same acid as in the earlier step. The soda lignin was then dried in an oven at 55 °C for 24 hours prior to further analysis [3].

Lignin-phenol-formaldehyde (LPF) resin was prepared in two steps which are preparation of lignin-phenol adduct (LP) and lignin-phenol-formaldehyde resin. The lignin phenol mixture was mixed (lignin to phenol 1:1) and stirred at 40 °C to obtain a homogenous mass. The resulting adduct was found to contain 50 % moisture. Then, 20 % of LP adduct, additional phenol (LP adduct to phenol 1:1), formaldehyde solution and methanol were mixed using mechanical stirrer. When the temperature was raised to 80 °C, 11.6 % NaOH dissolved in distilled water was added to the mixture and the reaction was continued for 4 hours [4].

The IR spectra of lignin, lignin-phenol-formaldehyde (LPF) resin and commercial-phenol-formaldehyde (CPF) resin were analyzed using FT-IR Perkin Elmer System 2000. This analysis was carried out to determine functional groups found in LPF and CPF resins.

Close contact joint or adhesive tensile strength of all samples was tested according to ASTM D 5266, the American Society for Testing and Materials (1999) standard procedure for estimating the percentage of wood failure in adhesive bonded joints.

Resins penetration test into wood structure were analyzed using Scanning Electron Microscope (SEM) model Leosupra 50VP FESEM.

Finally, capillary viscometer model *Micro-Ubbelohde Viscometer* was used to measure the viscosity of these resins.

Results and Discussion

Formulation of lignin-phenol-formaldehyde (LPF) resin was analyzed using FT-IR to compare against commercial-phenol-formaldehyde (CPF) resin. Figure 1 depicts the infrared (IR) spectra of lignin, LPF and CPF resins.

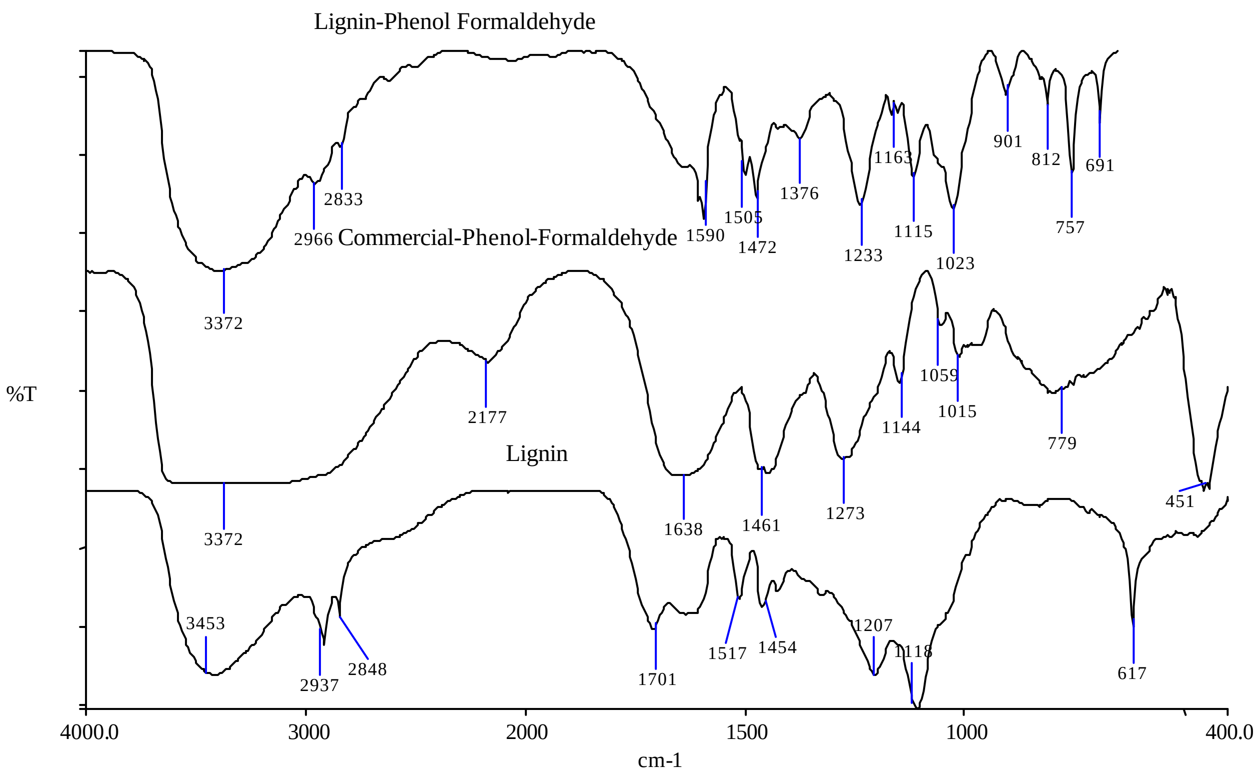

Figure 1: IR spectra of lignin, LPF and CPF resins

The IR spectrum of lignin shows absorption at 1513 cm^{-1}, 1458 cm^{-1} and 1421 cm^{-1}. The absorption bands for LPF and CPF resins were showed the presence of OH functional group at 3327 cm^{-1}. Besides, stretching vibration of aromatic compound can be seen in a wide adsorption band at 1638 cm^{-1} to 1505 cm^{-1} for both resins. The presence of these functional groups is the products of the reaction between phenol-lignin adduct and formaldehyde [5,6].

Basically, phenol will react with formaldehyde to make three bonding to produce ortho, para-hydroxymethylphenol as shown in Figure 2.

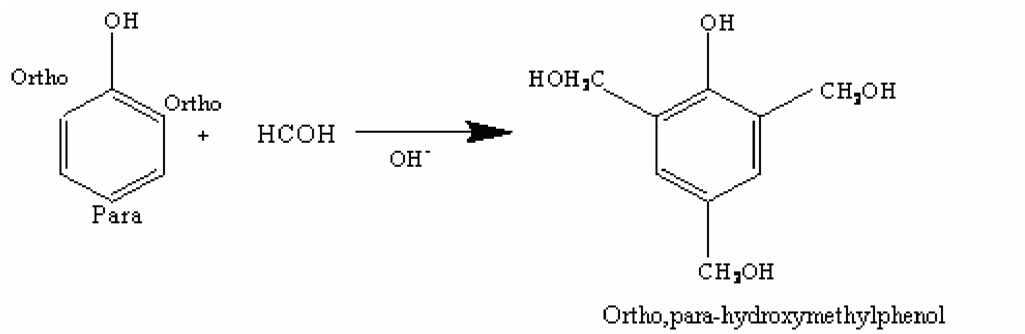

Figure 2: Reaction of ortho, para-hydroxymethylphenol

At the same time, formation of ortho-hydroxymethyllignin will form when lignin reacts with formaldehyde (Figure 3). During the reaction, some ortho or para-hydroxymethylphenol will react with ortho-hydroxymethyllignin to produce a network polymers of lignin-phenol-formaldehyde (LPF) resin as shown in Figure 4.

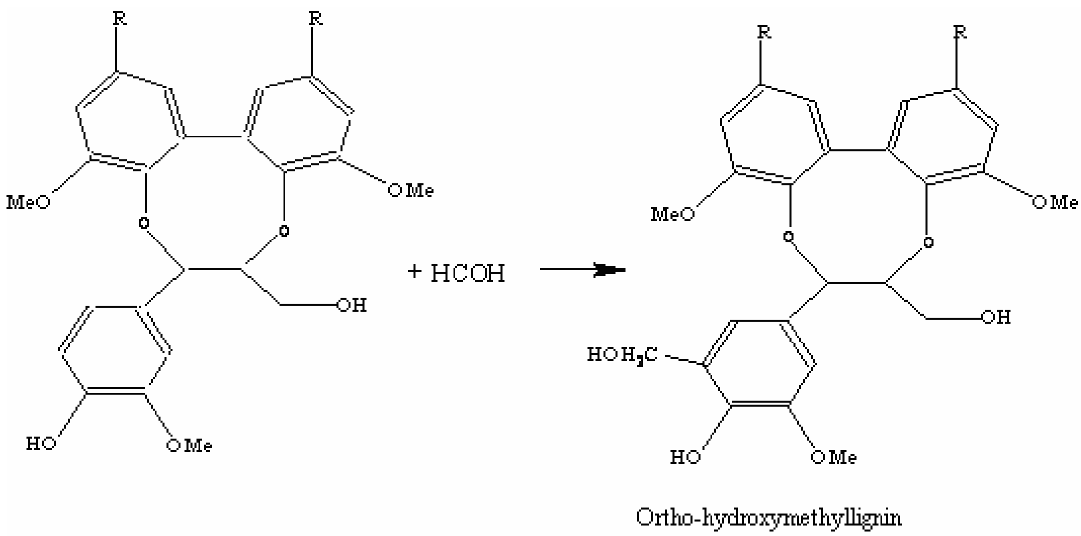

Figure 3: Reaction of ortho-hydroxymethyllignin

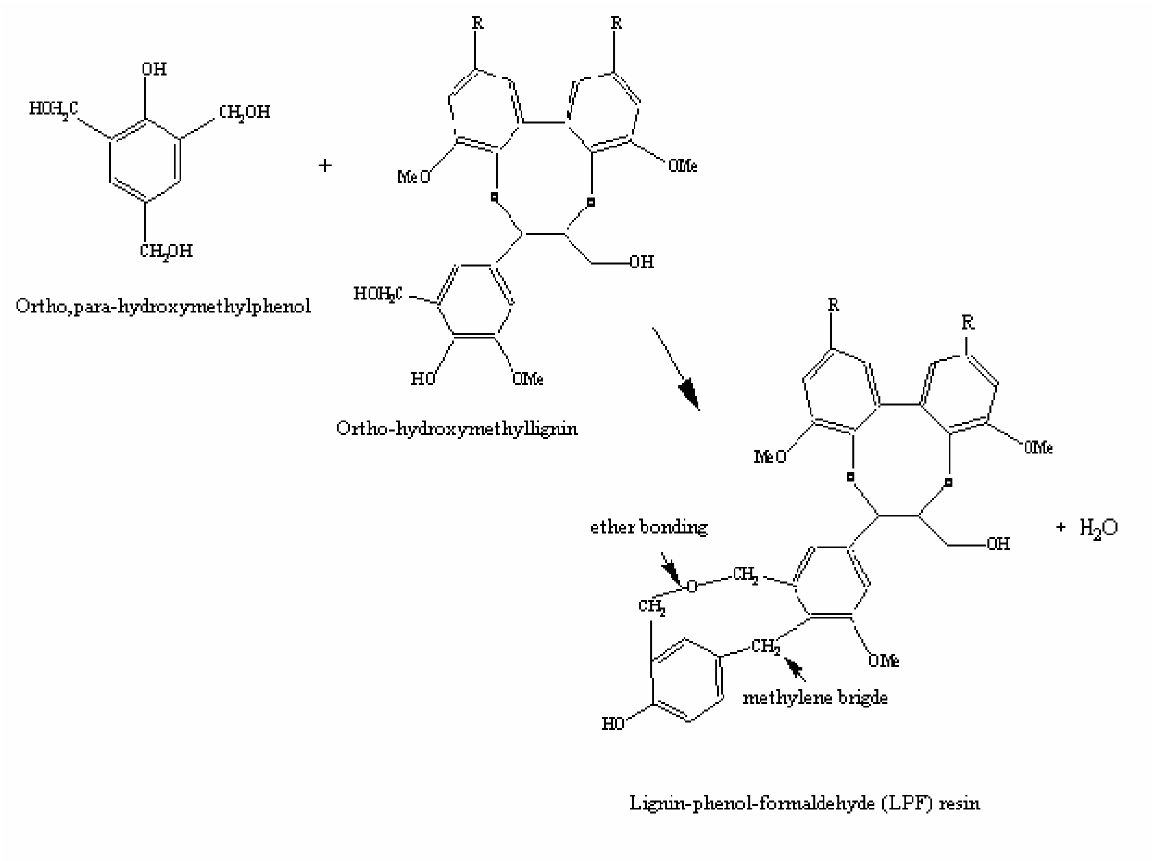

Figure 4: Reaction of lignin-phenol-formaldehyde (LPF) resin

Further reaction will lead to the formation of methylene bridges and water elimination. The bonding strength formed by LPF was studied via tensile strength test on wood substrate as shown in Table 1 and 2. The results indicated that LPF resin is far stronger than that of the CPF resin.

Table 1: Tensile strength test using LPF resin as an adhesive

Samples	Maximum load (N)	Ultimate Tensile Strength (MPa)	Tensile stress (Nm^{-2})
1	3823.50	15.39	1.84
2	2944.85	11.78	1.32
3	1866.08	7.51	0.33
Mean	2878.14	11.56	1.163

Table 2: Tensile strength test using CPF resin as an adhesive

Samples	Maximum load (N)	Ultimate Tensile Strength (MPa)	Tensile stress (Nm^{-2})
1	2536.78	5.15	0.53
2	2269.43	4.55	1.45
3	2930.61	7.15	2.28
Mean	2578.94	5.61	1.42

The ability of phenol formaldehyde to capture more loads before the adhesive broken apart is probably due to the presence of lignin and phenol. Based on the image shown in Figure 5, the LPF resin dispersion into wood structure is equally distributed. Besides, there is resin penetration into wood vessel pores to attach the resin with the wood structure [7]. In addition, the image of resin spreading pattern between the two surface layers of wood clearly shown that LPF resin is widely distributed due to its lower viscosity compared to CPF resin. This help to make the bonding formed is more firm [8]. In Figure 6, the image of wood that was adhered with CPF resin is presented. The bonds formed are not strong due to the present of crack as shown clearly in the Figure 6.

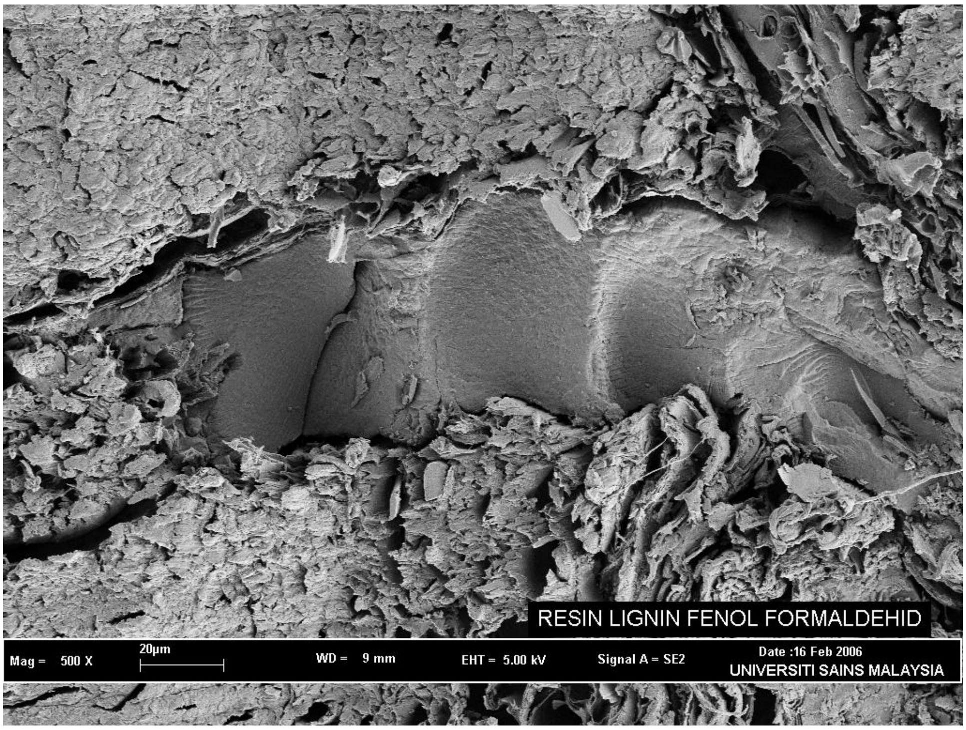

Figure 5: SEM image for LPF resin penetration effect in wood vessel

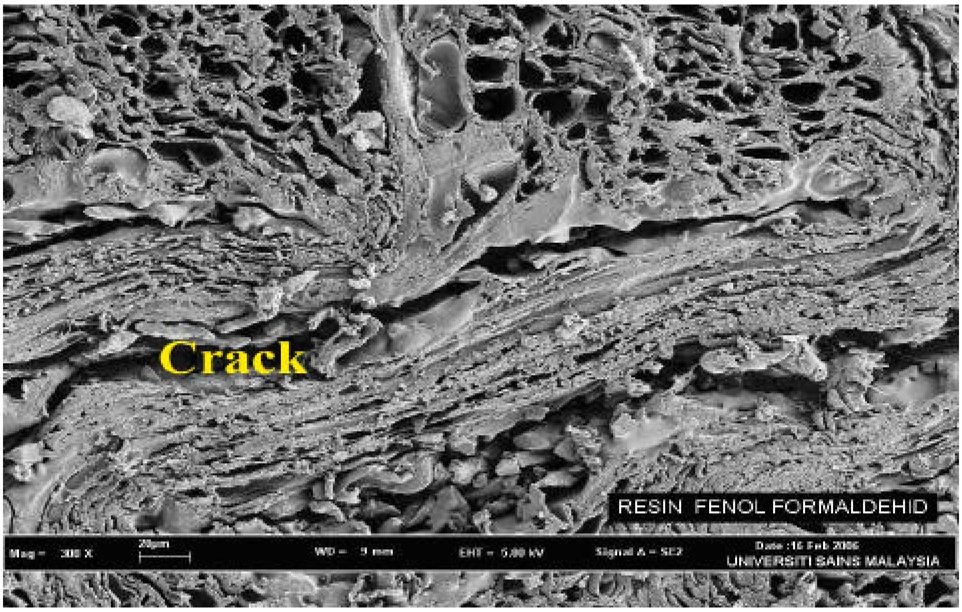

Figure 6: SEM image for CPF resin penetration effect in wood vessel

Table 3: Results for viscosity test for LPF and CPF resins

Days	LPF resins		CPF resins	
	25 ºC	29 ºC	25 ºC	29 ºC
7	0.9656 cSt	0.8204 cSt	6.1781 cSt	5.7156 cSt
14	0.9908 cSt	0.8612 cSt	9.9864 cSt	8.6264 cSt
21	0.9746 cSt	0.8756 cSt	9.9884 cSt	9.1148 cSt

Table 3 shows the kinematics viscosity of LPF resin which give lower value compared to CPF resin after kept for three weeks. Results show that kinematics viscosity for both resin increase against time. CPF resins show higher reading level of viscosity because these resins have higher density compared to LPF. The viscosity of a liquid depends on its density. Viscosity of liquid will increase as the density increases [9]. High density will hinder resin movement and cause resin to freeze rapidly while contaminated to air.

Besides, different temperature shows different viscosity reading for both resins. At 29 ^{o}C, both resins share a lower viscosity compared to that of 25 ^{o}C. Viscosity of the fluid decreases as the temperature increases because cohesive force between fluid molecules decreases and the kinetics force increased between molecules [10]. The tendency of resin to become viscous would hider the resin handling process on the substrate, especially on ply wood [11,12]. Therefore, low kinematics viscosity of lignin phenol formaldehyde resin would be better because this resin has longer shelf life or storage time.

Conclusion

The formulation of a lignin-phenol-formaldehyde (LPF) resin has been successfully developed as an alternative to the existing production of commercial-phenol-formaldehyde (CPF) resin. Physical and chemical properties comparison between LPF and CPF resins had been made. LPF resin had shown similar physical and chemical characteristics against CPF resin via FT-IR, viscosity test, tensile strength test and SEM analysis.

From the study, up to 50 % of phenol in commercial-phenol-formaldehyde (CPF) resin can be substituted by lignin in the production of lignin-phenol-formaldehyde (LPF) resin. Besides, LPF resin can be stored longer period of time compared to CPF resin due to its lower viscosity.

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