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Article

# Isolation, Characterization, and Utilization of Coconut Pith Lignin in a Biobased Adhesive

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## Abstract

In the study, lignin was extracted from coconut pith (CP) using soda pulping method, utilizing Response Surface Methodology to optimize key process parameters. The dependent variables were the extraction temperature and time, and NaOH concentration, with the yield (g lignin/10 g material) as the response. The statistical results identified extraction temperature ( $q = 0.0044$ ) and time ( $q = 0.0035$ ) as significant factors. NaOH concentration, though not significant individually ( $q = 0.757$ ), exhibited interaction effect with time ( $q = 0.006$ ). The theoretical optimal extraction conditions were 159.0 °C, 169.0 min, and 2.1% NaOH, under which an actual coconut pith lignin (CPL) yield of 1.806 g/10 g CP was achieved. Characterization of CPL showed a composition of 92.96 ± 0.32% acid-insoluble lignin. To demonstrate its applicability, CPL was incorporated into a phenol-formaldehyde (PF) adhesive formulation, substituting the phenol component. The Adhesive Shear Strength Test demonstrated that the lignin-formaldehyde (LF) adhesive had a mean failing load of 0.819 kg/cm<sup>2</sup>, nearly half that of the prepared PF adhesive at 1.78 kg/cm<sup>2</sup>. The FT-IR spectra of the CPL and LF adhesives revealed notable differences in the 1750 – 1000 cm<sup>-1</sup> region, suggesting distinct structure and bond formations. These findings illustrate the potential of CPL as a sustainable phenol substitute in industrial adhesive formulations and in other applications.

**Keywords:** coconut pith; lignin; soda pulping; phenol-formaldehyde; adhesive

## 1. Introduction

Phenol is a highly valuable industrial chemical and among natural sources, lignin has emerged as one of its most promising natural alternatives [1-3]. Lignin is the second most abundant natural polymer after starch and is primarily composed of substituted phenolic compounds derived from p-coumaryl, coniferyl, and synapyl alcohols [4,5]. It is a common plant polymer that provides structural reinforcement, strengthening the woody parts [6]. It is biosynthesized through enzyme-mediated dehydrogenative polymerization of above-mentioned phenolic compounds that are incorporated into the polymer as the phenylpropanoids p-hydroxyphenyl, guaiacyl, and syringyl [7].

Industrially, lignin is derived as a by-product of pulp and paper manufacturing, extracted mainly through the kraft and sulfite processes [8]. Other more recent extraction technologies have also been developed such as the use of the biomass process, solvents, and soda pulping. Soda pulping is a widely practiced method for non-wood feedstocks such as straw, sugarcane bagasse, and flax [9]. Through this method, lignin may be recovered by precipitation from the effluents or excess liquors generated by pulp mills followed by liquid-solid separation and drying [10].

Lignin has commercial applications and more recent studies have shown that these could further expand [11]. Kraft lignin can be sulfonated to various degrees depending on the process and can be utilized in applications depending on its solubility, molar mass distribution, and hydrophilic character [12]. Among the potential applications of kraft lignin, its use as dispersant and additive for concrete mixtures have shown the best results [13]. In a previous study, softwood lignin was applied as a dispersant and flocculant. Aside from Kraft lignin, other types such as organosolv lignin have also been studied [14]. It has been proposed that this lignin type can be used as partial replacement for phenolic resins that serve as binders in the manufacture of friction products or lubricants [15].

This material has been successfully utilized on a commercial scale by a North American manufacturer of automotive brake pads and moldings.

According to previous reports, coconut husks contain about 33–53% lignin [16, 17]. These values are generally higher than those from common hardwoods (23–30%) and softwoods (26–34%) [18]. Coconut husks hold marginal economic value and are often regarded as waste by many coconut farms and related industries; thus, their conversion into usable products is highly beneficial. The Philippines has nearly half a billion coconut trees and generates an estimated 1.8 million tons of shells and 4.1 million tons of husks annually [19]. While shells and husks are partially utilized in charcoal production alongside wood and bamboo, they still represent a substantial and underutilized lignocellulosic waste stream.

In this study, the researchers aimed to optimize the soda pulping extraction process for isolating coconut pith lignin (CPL). The extracted lignin was subsequently utilized in the formulation of a phenol–formaldehyde (PF) adhesive to evaluate its potential as a sustainable alternative to phenol

## 2. Materials and Methods

### A. Materials

CP was purchased from a local supplier, dried at 60 °C for about 12 hrs or until the moisture content was  $\leq 20\%$ , and was then sifted using a USA Standard Testing Sieve No. 16 prior to use. Sodium hydroxide (NaOH) and sulfuric acid ( $\text{H}_2\text{SO}_4$ ) were purchased from Duksan Pure Chemicals. Sodium sulfite ( $\text{Na}_2\text{SO}_3$ ) anhydrous was bought from Loba Chemie Pvt. Ltd. Phenol ( $\text{C}_6\text{H}_5\text{OH}$ ) and Urea ( $\text{CO}(\text{NH}_2)_2$ ) were sourced from Techno Pharmchem. Formaldehyde ( $\text{CH}_2\text{O}$ ) was procured from RCI Labscan. All chemicals used in the study were analytical grade and were utilized without further processing.

### B. Soda Pulping Extraction of CPL

Lignin was extracted from the CP via soda pulping method. Exactly 10 g of CP was weighed in a 1 L glass reaction vessel with lid & steel clamp. About 200 mL of NaOH solution was added and the mixture was refluxed with stirring for a set time and temperature, immediately after which approximately 400 mL of distilled water was added. The mixture was mixed then vacuum filtered using a Whatman Filter No. 1. The residue was washed thrice with 100 mL portions of distilled water and all filtrates were collected. The filtrate's pH was then slowly adjusted to pH 2.0 using 20%  $\text{H}_2\text{SO}_4$  and was boiled for approximately 1 hr. The mixture was allowed to cool to room temperature and the resulting precipitate was vacuum filtered using Whatman Filter No. 1. The precipitate was washed thrice with 100 mL portions of distilled water and was dried in a Memmert Oven Model U10 at 70 °C for about 16 hrs. After drying, the extracted lignin was weighed and its yield was calculated. To maximize the yield, process optimization was performed using Stat-Ease® Design Expert Version 7.0. Response Surface Methodology (RSM) was used with Central Composite Design (CCD) having three independent factors (time, temperature, and NaOH concentration) and one response (Yield). Table 1 shows the design summary employed to optimize the soda pulping of CP.

**Table 1.** Design summary for soda pulping of CP showing the three (3) independent factors and the chosen response.

Study Type		Response Surface			
Initial Design		Central Composite			
Design Model		Quadratic			
Factor	Name	Units	Type	Low Actual	High Actual
A	Time	min	Numeric	60.00	170.00
B	Temp	°C	Numeric	120.00	160.00
C	%NaOH	%	Numeric	2.00	5.00
Response		Units			
Yield		g lignin / 10 g material			

### C. Preparation of PF Adhesive

The preparation of the adhesive was based on the method described in a previous study with modifications [20]. Exactly 21.96 g phenol was added with 14.6 mL formaldehyde and heated at 80 °C for 1 hr. Then, 11.9 mL formaldehyde and 1.80 mL 30% NaOH were added and the mixture was heated at 80 °C for 1 hr. Afterwards, the same amount of formaldehyde and 30% NaOH were again added followed by heating at 80 °C for another 1 hr. About 28.80 g urea and 5.10 mL 30% NaOH solution were added and the resulting mixture was heated at 87 °C for 30 min. After heating, the resulting PF adhesive was immediately cooled down to room temperature and was stored in a chiller until further analysis or use.

### D. Preparation of Lignin-Formaldehyde (LF) Adhesive

To prepare the LF adhesive, the procedure for preparing the PF adhesive was followed with modifications wherein phenol was substituted by CPL on a per weight basis. Prior to the synthesis, CPL was dissolved in a 30% (w/w) NaOH solution at a CPL-to-NaOH weight-to-volume ratio of 1:8. The mixture was subjected to gentle heating at 50 °C for 1 hr to ensure complete dissolution of the lignin. Once dissolved, the appropriate amount of formaldehyde was added to the solution, and the mixture was heated at 80 °C for 1 hr. Subsequently, a second portion of formaldehyde was introduced, and the heating process at 80 °C was repeated for another 1 hr. A third batch of formaldehyde was then added, followed by an additional 1-hr heating period at 80 °C. Finally, urea was incorporated into the mixture, and the reaction was allowed to proceed at 87 °C for 30 min. Upon completion of the synthesis, the resulting LF adhesive was immediately cooled to room temperature and stored in a chiller until further analysis or application.

### E. Chemical and Physical Analyses

#### 1. CP and CPL characterization

##### a. Moisture Content

The moisture content of all samples was analyzed using an IR moisture analyzer. The sample was analyzed by drying at 105 °C and the equipment was set and operated based on the recommendation of the manufacturer.

##### b. Ash Content

The ash contents of the samples were analyzed using a Nabertherm L9/S Furnace. Exactly 1 g of sample was placed in a pre-weighed crucible with cover and was heated to 575 °C for at least 4 hrs. After heating, the ash was allowed to cool down to ambient temperature in a desiccator prior to weighing.

##### c. Lignin Content

The lignin content of the samples was analyzed using the procedure of a previous work [21]. Exactly 1 g of sample was treated with 15 mL of 72% H<sub>2</sub>SO<sub>4</sub> at 37 °C for 2 hrs. The mixture was then diluted to 3% H<sub>2</sub>SO<sub>4</sub> and refluxed at 80 °C for 4 hrs. Afterwards, the sample was filtered using Whatman Filter No. 1. The residue (insoluble lignin fraction) was dried using a Memmert Oven Model U10 at 100 °C for about 16 hrs. The dried sample was weighed and reported as the insoluble lignin. The filtrate (soluble lignin fraction) was analyzed using a Shimadzu UV Spectrophotometer UV mini-1240 at 280 nm. The soluble lignin was calculated using the formula:

$$\text{Soluble lignin} = \frac{A}{110} \times \frac{\text{dilution}}{\text{sample weight}} \times 100\% \quad (1)$$

The total lignin content was calculated by adding the results of the insoluble and soluble lignin fractions.

##### d. Lignin Phenolic Hydroxyl Content

The phenolic content of the samples was measured employing an earlier established method [22]. Approximately 0.1 g of sample was dissolved in 100 mL pH 12 buffer solution (12.4 g boric acid in 2 L 0.1 N NaOH). Then, a 2 mL aliquot was pipetted and diluted to 50 mL with the pH 12 buffer solution. Another 2 mL aliquot was added with 2 mL of 0.1 N H<sub>2</sub>SO<sub>4</sub> and diluted to 50 mL with pH 6 buffer solution (495 mL of 0.2 N KH<sub>2</sub>PO<sub>4</sub> and 113 mL of 0.1 N NaOH diluted to 2 L with distilled H<sub>2</sub>O). The absorbance of each solution at 280 nm to 400 nm (with 5 nm intervals) was measured using a Shimadzu UV Spectrophotometer UV mini-1240. The difference between the measured absorbances per wavelength was then calculated. The phenolic hydroxyl group content of the lignin samples was calculated using the  $\Delta\alpha_{max}$  value of the sample and the following equation:

$$\%phenolic\ hydroxyl = \Delta\alpha_{max} \times \frac{17}{41} \quad (2)$$

#### e. Thermogravimetric Analysis (TGA)

About 0.5 g of sample was submitted to a third-laboratory laboratory for TGA measurements. The samples were analyzed using Perkin Elmer STA 6000 by heating from 30 to 600 °C and then 600 to 900 °C with heating rate of 20 °C/min. Nitrogen was used at 50 mL/min from 30 to 600 °C while oxygen was used from 600 to 900 °C.

#### f. Fourier Transform-Infrared (FT-IR) Analysis

About 1.0 g of sample was submitted to a third-laboratory laboratory for FT-IR analysis. The testing protocols for FT-IR of the said laboratory were followed.

### 2. Adhesive Characterization

#### a. Viscosity Measurement

The viscosity of the samples was measured using a Cannon-Fenske Capillary Viscometer. A Koehler KV3000 Series Kinematic Viscosity Bath was used to maintain the temperature at 40 °C and time the viscosity measurements. Prior to taking measurements, the samples were allowed to equilibrate at 40 °C in the viscosity bath.

#### b. Adhesive Shear Strength

The samples were submitted to a third-party laboratory to test their adhesive strength. To prepare the samples, two strips of hard cardboard each measuring 5 × 1 (l × w) inch were glued together using the prepared adhesives with an overlap of 0.5 in on one end. Ten (10) pairs were prepared for each adhesive sample and were analyzed for shear strength based on the ASTM D1002 standard.

## 3. Results

### A. Extraction of CPL

In the study, CPL was extracted using soda pulping. To maximize the yield, process optimization was conducted using RSM with CCD (see Table A1 in Appendix A for the three (3) variables investigated, their combinations, and their respective experimental responses). Table 3.1 summarizes the ANOVA results of the process optimization while Table 3.2 presents the R-Squared, Adjusted R-Squared, and Predicated R-Squared of the model generated from the experimental data.

**Table 2. 1.** ANOVA results of lignin extraction using soda pulping.

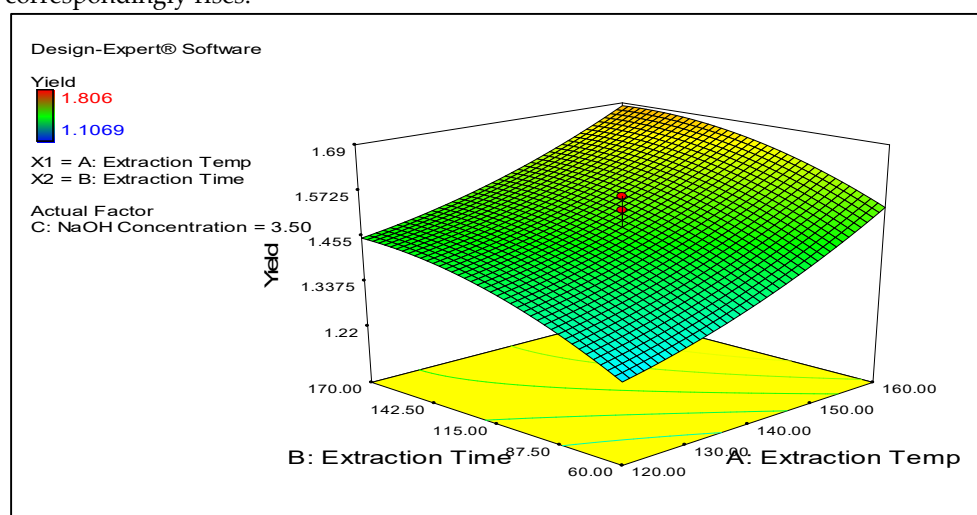
Source	Sum of Squares	Df	Mean Square	F Value	q-value Prob > F
Model	0.37	9	0.041	13.73	0.0050
A-Time	0.072	1	0.072	24.27	0.0044
B-Temp	0.080	1	0.080	26.86	0.0035
C-% NaOH	3.188E-004	1	3.188E-004	0.11	0.7570
AC	3.454E-003	1	3.454E-003	1.16	0.0037
Lack of Fit	2.640E-003	3	8.800E-004	0.14	0.9256

**Table 2. 2.** R-squared values of the model generated for the conducted soda pulping.

R-Squared	0.9611
Adjusted R-Squared	0.8911
Predicated R-Squared	0.8112
Adequate Precision	15.887

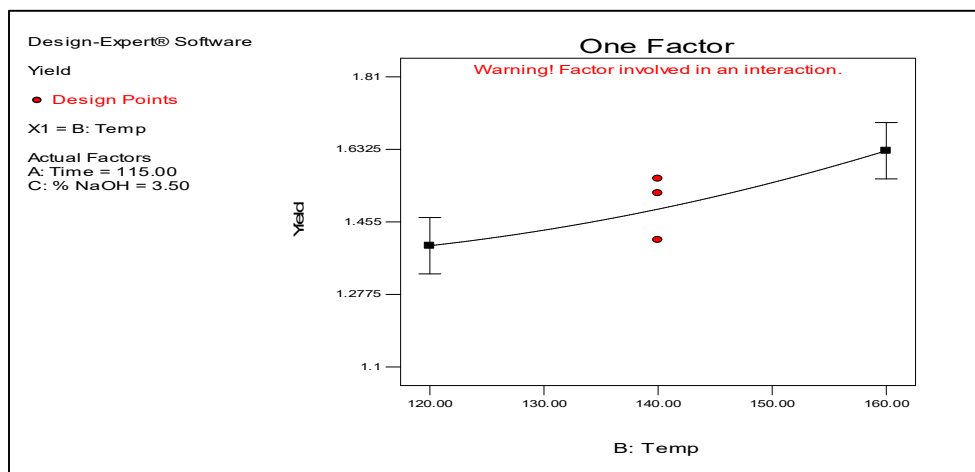
The results show that the quadratic model generated ( $\rho = 0.005$ ; lack of fit is  $\rho = 0.9256$ ), including the variables time and temperature, are significant ( $\rho = 0.0044$  and  $0.0035$ , respectively). The R-squared values (see Table 2.2) propose that the model is a good fit for the data. Unexpectedly, the NaOH concentration has a  $\rho$ -value  $> 0.05$ , which contradicts findings commonly reported in the literature. This indicates that the range covered in this study did not have considerable effect on the CPL yield. The Predicted R-Squared of 0.8112 is in reasonable agreement with the Adjusted R-Squared (0.8911), which means that the model has good predictive capability. The Adequate Precision measures the signal to noise ratio and a value higher than 4 is desirable. In this case, its value is 15.887, demonstrating adequate signal.

Figure 1 shows a 3D contour map demonstrating the effect of the experimental time and temperature on the CPL yield. The map illustrates that as the time and temperature increase, the yield also correspondingly rises.

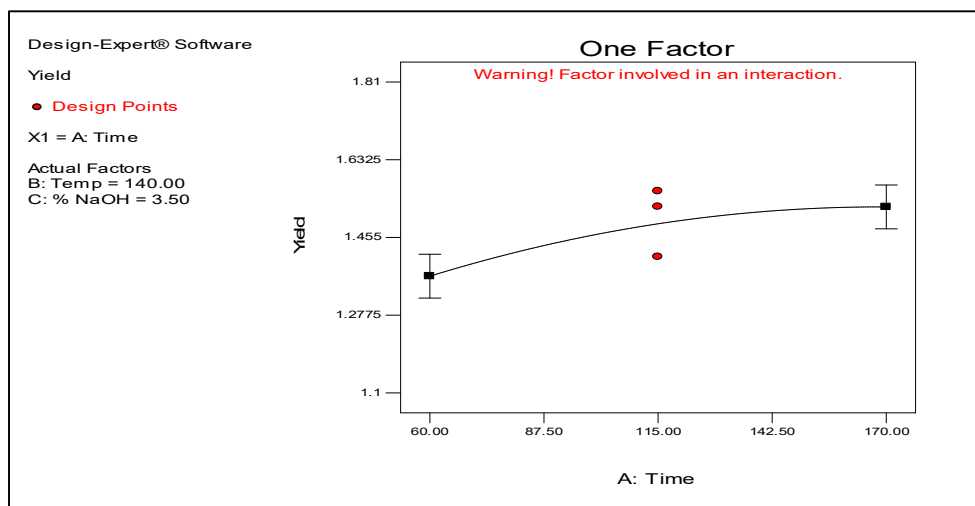


**Figure 1.** 3D surface plot showing effects of temperature ( $^{\circ}\text{C}$ ) and time (min) on lignin yield (g lignin / 10 g material).

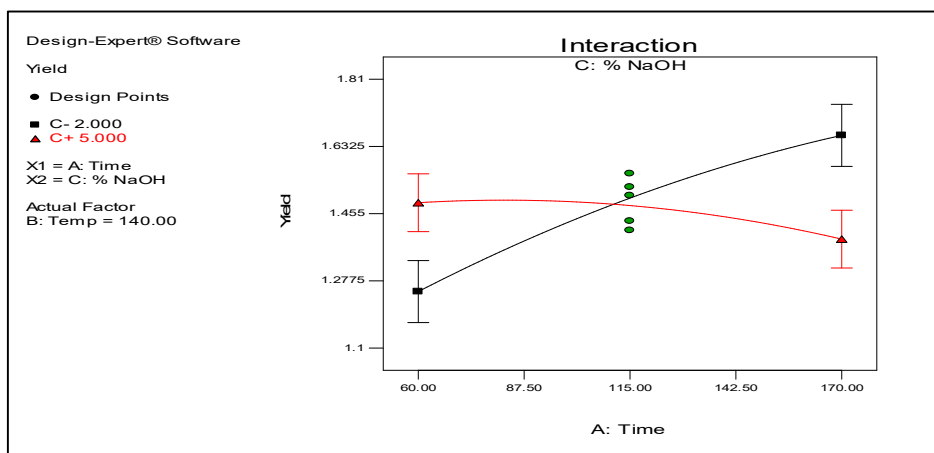
Figures 2 and 3 illustrate how temperature and time individually affect the yield. As shown, the yield exponentially increases with the temperature. In contrast, the yield increases for a certain period but seemingly levels off after 170 min. The results also demonstrate that temperature and time do not interact to affect the yield, suggesting that both variables can be manipulated independently to obtain a target result. Meanwhile, time and NaOH concentration shows significant interaction as illustrated by their plots in Figure 4. Conversely, temperature also interacts with NaOH concentration as shown in Figure 5 but this interaction does not have a significant effect on the yield based on the ANOVA results (data not shown).



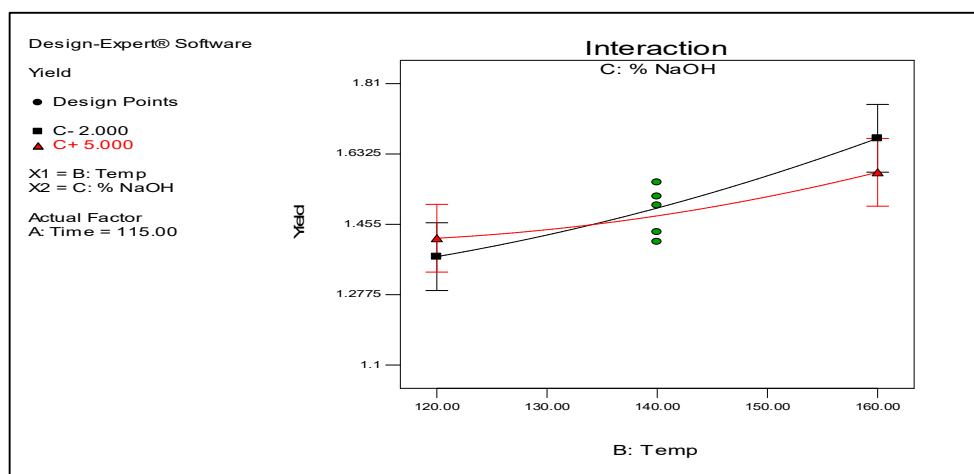
**Figure 2.** 2D plot showing effect of temperature (°C) on lignin yield (g lignin / 10 g material)



**Figure 3.** 2D plot showing effect of time (min) on lignin yield (g lignin / 10 g material)

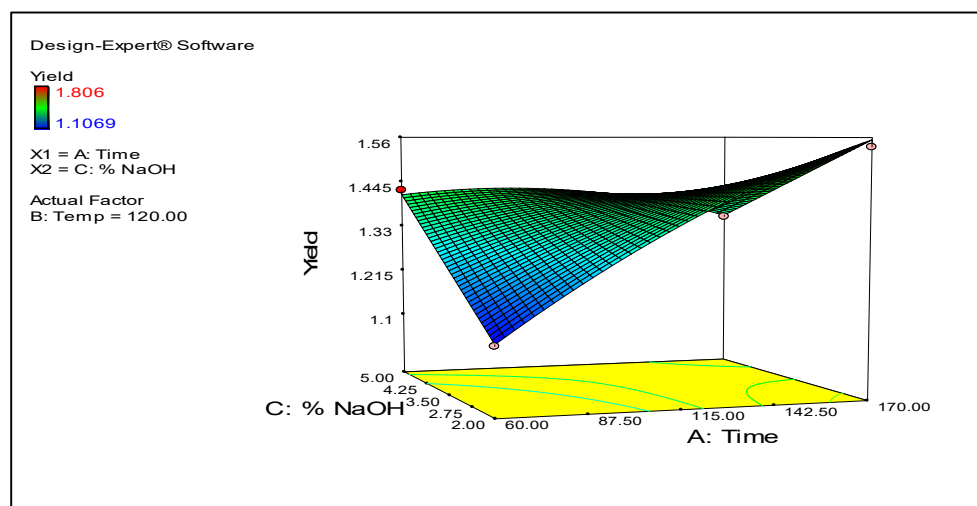


**Figure 4.** 2D plot showing interaction between time (min) and NaOH concentration (%) and their effect on the lignin yield (g lignin / 10 g material).



**Figure 5.** 2D plot showing interaction between temperature (°C) and NaOH concentration (%) and their effect on lignin yield (g lignin / 10 g material)

The interaction between NaOH concentration and time presents interesting results. The 3D surface plot in Figure 6 shows that time and NaOH concentration affect the yield. An increase in both factors generally results in an improved yield. However, when their interaction is taken into account, an increase in both factors' values does not necessarily produce the same result (see Figure 4). Conversely, a reduced yield might be expected especially at higher NaOH concentration and time. This result implies that a high NaOH concentration can lead to the possible degradation or modification of the lignin resulting to lower yield after acid precipitation with H<sub>2</sub>SO<sub>4</sub>.



**Figure 6.** 3D surface plot showing effects of NaOH concentration (%) and time (min) on lignin yield (g lignin / 10 g material)

The model generated follows Equation (3) given below:

$$\text{Yield} = +1.50 + 0.12 * A + 0.083 * B - 5.535E-003 * C - 8.425E-003 * AB - 0.026 * AC - 0.12 * BC + 0.029 * A^2 - 0.044 * B^2 - 0.010 * C^2 \quad (3)$$

where A is time, B is the temperature, and C is the NaOH concentration. AC, AB, AC, and BC represent the interactions of the variables.

To obtain maximum yield, the theoretical optimization (see Table A2 in Appendix A) suggested a temperature and time combination of 160.0 °C and 168.11 min using 2.12% NaOH. The theoretical maximum yield was calculated at 1.808 g lignin / 10 g material while the actual yield was 1.806 g / 10 g material (99.8%), demonstrating that the model captured the actual conditions affecting the yield of the optimized lignin extraction process.

## B. Characterization of CP and extracted CPL

The physicochemical properties of coconut peat and CPL are presented in Table 3. Upon delivery, the fresh coconut pith was immediately dried using an air convection and a final moisture content of  $18.40 \pm 0.62\%$  was recorded. The ash content of the dried coconut peat was  $6.68 \pm 0.13\%$ . The insoluble lignin content was at  $45.02 \pm 1.72\%$  which is relatively higher compared to other sources. For example, typical hardwoods contain approximately 21–31% lignin, while softwoods range from 27–32% [23]. Herbaceous materials such as grasses generally exhibit 10–20% lignin, and residues like wheat straw, rice straw, and corn stalks often range even lower, between 10–20% [24]. Lignocellulosic coconut coir is reported to contain 37–42% lignin, which underscores the inherently high lignin level of coconut-derived materials [25].

The extracted CPL was a dark brown crystalline solid (ground) as shown in Figure 7. The ash content of the extracted CPL was  $1.15 \pm 0.04\%$ , which was much lower than that of the raw material. This result is desirable because a high ash content could interfere with the utilization of lignin as a potential industrial chemical. Using the optimized soda pulping process, the study obtained CPL with  $92.96 \pm 0.32\%$  purity, slightly higher (82.8%) than the results reported in the literature [16]. The phenolic hydroxyl content of CPL was  $0.80 \pm 0.04\%$ , which was slightly lower compared with those reported in literature for technical and native lignin. Typical phenolic hydroxyl contents for lignin derived from hardwoods and softwoods range between 1–3%, depending on the isolation method and botanical source whereas organosolv and kraft lignin may exhibit even higher values due to structural modifications during pulping [23, 26, 27]

**Table 3.** Physicochemical properties of CP and CPL.

Property	CP	CPL
Moisture Content (%)	$18.40 \pm 0.62$	n/a
Ash (%)	$6.68 \pm 0.13$	$1.15 \pm 0.04$
Insoluble lignin content (%)	$45.02 \pm 1.72$	$92.96 \pm 0.32$
Acid soluble lignin (%)	$4.05 \pm 1.40$	$0.0088 \pm 0.0021$
Phenolic hydroxyl content	n/a	$0.80 \pm 0.04$

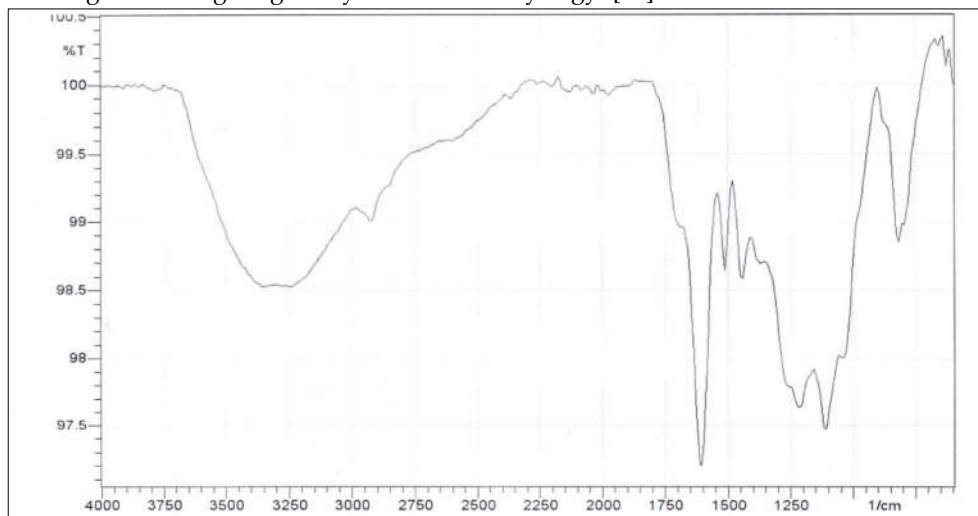


**Figure 7.** Photo of ground extracted CPL.

TGA was also conducted on CPL to demonstrate how it specifically loses mass when heat is incrementally applied. The Thermogravimetric and Derivative Thermogravimetric Analysis (TG-DTA) curves are shown in Figure A1 and the data are presented in Table A3 of Appendix A. The wide range of decomposition temperature indicates that CPL is structurally heterogenous. Moreover, the sample presented a high peak temperature which suggests excellent thermal stability.

Figure 8 presents the FT-IR spectrum of CPL which shows a strong and broad band at around  $3700\text{--}3000\text{ cm}^{-1}$ , indicating phenolic and aliphatic OH groups. There is also a band at about  $2924\text{ cm}^{-1}$  which suggests C–H stretching in alkyl sidechains. The bands at  $1605$  and  $1512\text{ cm}^{-1}$  are also

noticeable and may be attributed to the C=C stretching in the structure's aromatic skeleton [28]. The band at  $\sim 1443\text{ cm}^{-1}$  suggests aromatic skeletal vibration particularly C-H in-plane deformation. The band at  $1211\text{ cm}^{-1}$  may be related to C-C, C-H, and C=O stretches in the CPL guaiacyl ring; in addition, bands at  $1111$  and  $818\text{ cm}^{-1}$  might be due to the C-H deformations of the syringyl units [29]. The FT-IR data suggests that CPL extracted in this study has a combination of guaiacyl and syringyl units similar to what other studies were able to attain [28, 30]. Others have reported that their extracted lignin had higher guaiacyl content than syringyl [31].



**Figure 8.** FT-IR spectrum of CPL extracted via soda pulping method

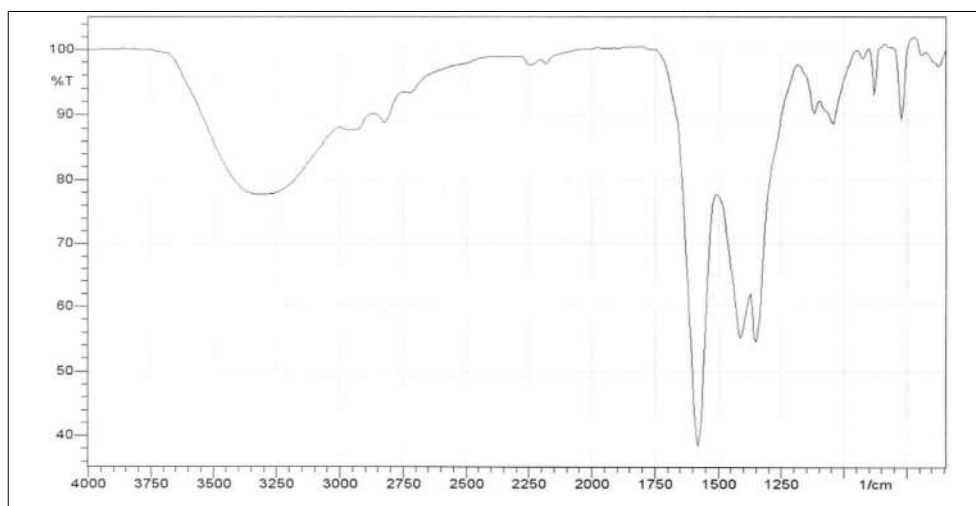
#### Characterization of LF Adhesive

One objective of the study was to replace phenol with lignin in adhesives to demonstrate how it can eventually serve as a sustainable alternative to the said petroleum-based chemical. In this study, a standard PF adhesive and LF formulation were prepared and their viscosity and adhesive shear strength were tested. Generally, LF exhibited lower viscosity and adhesive shear strength values than PF (see Table 4). A low-viscosity adhesive can be utilized in various applications requiring high flowability, surface wetting, and precision, examples of which include adhesives used for bonding, sealing, potting, encapsulation and impregnation (Low viscosity adhesives, sealants and coatings). Although the adhesive strength of LF is nearly half that of PF, the result proposes that native and crude CPL has enough phenolic character to replace phenol to a certain extent.

**Table 4.** Shear Strength and Viscosity of PF and LF.

Type	Viscosity ( $\text{mm}^2/\text{s}$ )	Mean Failing Load (Shear Strength, $\text{kg}/\text{cm}^2$ )
PF	$33.18 \pm 0.25$	$1.78 \pm 0.50$
LF	$10.90 \pm 0.07$	$0.82 \pm 0.21$

The FT-IR spectrum of the LF adhesive in Figure 9 shows a broad band at  $3700\text{--}3000\text{ cm}^{-1}$ . This may be assigned to the O-H stretching of the phenolic and aliphatic hydroxyl groups and can be indicative of strong hydrogen bonds and residual OH groups that are important for adhesive interactions [23, 26]. Noticeable absorptions around  $2970$  and  $2824\text{ cm}^{-1}$  correspond to the aliphatic C-H stretching of  $-\text{CH}_2-$  and  $-\text{CH}_3$  groups from the lignin side chains and methylene bridges formed during condensation [23, 26]. A strong peak at  $1582\text{ cm}^{-1}$  represents aromatic skeletal vibrations, thus suggesting a phenylpropane backbone that contributes to rigidity and thermal stability [32]. Additional aromatic ring vibrations at  $1412\text{ cm}^{-1}$  and a syringyl-associated band at  $1350\text{ cm}^{-1}$  reflect guaiacyl/syringyl substitution patterns in the lignin matrix [23, 26]. The band at  $1119\text{ cm}^{-1}$  further supports syringyl substitution while that at  $1041\text{ cm}^{-1}$  indicates C-O stretching of alcohols and ether linkages [32].



**Figure 9.** FT-IR Spectrum of formulated LF Adhesive

Other studies have reported that absorptions in the 1458-1350  $\text{cm}^{-1}$  region, associated with methylene (-CH<sub>2</sub>-) deformations, and in the 1257-1149  $\text{cm}^{-1}$  range, assigned to Ar-O-C ether linkages, can help provide evidence of crosslinking reactions between lignin phenolic groups and formaldehyde, and confirm adhesive network formation [33, 34]. In addition, a low-frequency absorption at about 787  $\text{cm}^{-1}$  (aromatic C-H out-of-plane bending) may help confirm the diverse aromatic substitution patterns and the coexistence of guaiacyl and syringyl units [23, 26]. Comparing the spectra shown in Figures 9 and 10, noticeable distinctions can be observed particularly around 1600-800  $\text{cm}^{-1}$ , signifying differences in their chemical structure or imply bond formation.

#### 4. Discussion

The aim of the study was to extract CPL and utilize it in an application to demonstrate its capacity as an industrial material. Coconut pith is considered an attractive resource because it has been shown to contain a considerable amount of lignin compared with other woody materials. CPL was extracted using soda pulping due to its relatively simpler and milder conditions compared to other common methods like Kraft and sulfite pulping. Moreover, although previous reports on CPL extraction exist, optimization was performed using a theoretical model to pinpoint the specific conditions for maximum yield and to assess how the different variables interact.

Using the experimental data, a quadratic model (Equation 3) was generated, which provided a specific set of theoretical temperature, time, and NaOH concentration to reach a calculated maximum yield. The actual vs. theoretical yields were nearly identical, demonstrating the model's accuracy, which means it can be applied to target specific responses with a set of preferred variables. It is noteworthy that the yield increases with temperature and time, although the increase with time leveled off after 170 minutes. While similar results have been reported, in this specific study, the NaOH concentration was not a significant factor for CPL extraction, in contrast with findings presented in other works. This result demonstrates that within the selected range of NaOH concentration (2 – 5%), one can expect the same amount of CPL.

The data interestingly revealed that NaOH concentration interacts with time, and this interaction significantly affects the CPL yield. Specifically, at higher NaOH concentrations, time negatively impacts the response, meaning the yield decreases over longer periods. Conversely, when the concentration is lower (roughly about 2%), the yield correspondingly increases with time. Therefore, during production, the manufacturer should prioritize the minimum NaOH concentration to ensure that the yield continues to improve as the reaction time is extended. Although the direct cause of this specific behavior was not investigated in the study, it can presumably be attributed to either the degradation of CPL at higher NaOH concentrations or the high alkali condition negatively affecting the solubility of the extracted CPL.

In the current study, the extracted CPL was applied into a PF adhesive by direct substitution of the phenol portion. This was done to evaluate the performance of CPL as a sustainable alternative to the petroleum-based chemical given that lignin is a phenolic natural polymer. Data from the conducted shear strength test illustrates that the LF adhesive possessed just half the adhesiveness of PF. This may be attributed to the lower phenolic content of CPL, leading to a lesser degree of polymerization with formaldehyde. Nonetheless, it was capable, at least to some degree, of forming an adhesive and partially replacing phenol. This assumption is supported by the FT-IR spectrum of CPL and the LF adhesive, showing key differences and distinct bands. However, to fully realize the potential of lignin as a direct substitute for phenol or other petroleum-based compounds, further research is necessary, focusing mainly on altering the lignin's chemical structure to improve its functionality and reactivity.

## 5. Conclusion

This study demonstrated the successful extraction of lignin from CP using the soda pulping method, with process optimization achieved through RSM. The optimized conditions yielded a high proportion of acid-insoluble lignin, confirming CP as a rich lignocellulosic source. When applied as a phenol substitute in PF adhesives, LF adhesive exhibited a shear strength comparable to conventional PF resin, highlighting its potential as a sustainable alternative. FT-IR analysis further highlighted structural differences between PF and LF adhesives, particularly within the hydroxyl and aromatic vibration regions, suggesting unique bond formation during polymerization. Collectively, these findings position coconut pith-derived lignin as a promising renewable resource for adhesive production, supporting both material performance and sustainability objectives in industrial applications.

## 6. Patents

The technology presented in this article was awarded with a Utility Model Registration with Registration No. 2/2022/051178 and Registration Title "Coco peat Lignin-based Phenol Formaldehyde Adhesive" last 04 January 2023 by the Bureau of Patents, Intellectual Property Office of the Philippines.

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## Abbreviations

The following abbreviations are used in this manuscript:

ANOVA	Analysis of Variance
CCD	Central Composite Design
CP	coconut pith
CPL	coconut pith lignin
FT-IR	Fourier Transform-Infrared
LF	lignin-formaldehyde

## Appendix A

**Table A1.** Details of the runs conducted including their resulting experimental yield.

Extraction Temperature (°C)	Extraction Time (minutes)	NaOH Concentration (% w/v)	Yield (g lignin / g material)
173.6	115.0	3.5	1.5781
140.0	115.0	3.5	1.5608
120.0	170.0	2.0	1.5382
140.0	207.5	3.5	1.5244
120.0	60.0	2.0	1.1069
140.0	115.0	3.5	1.4109
140.0	115.0	1.0	1.5028
120.0	170.0	5.0	1.3431
140.0	W22.5	3.5	1.2241
160.0	60.0	5.0	1.6223
160.0	60.0	2.0	1.6103
140.0	115.0	6.0	1.4355
120.0	60.0	5.0	1.4237
140.0	115.0	3.5	1.5254
106.4	115.0	3.5	1.3719
160.0	170.0	2.0	1.8060
160.0	170.0	5.0	1.5308

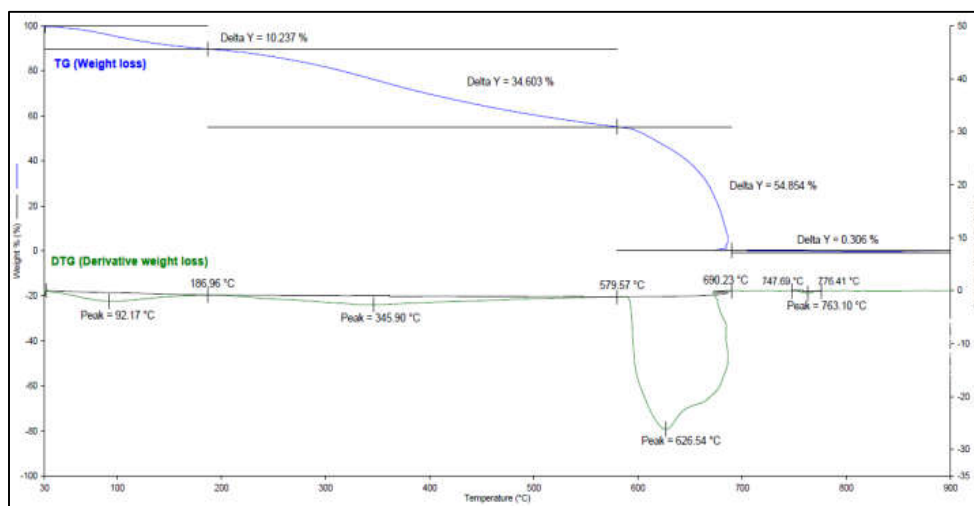
**Table A2.** Theoretical temperature and time combination suggested by the software based on the model generated for maximum yield.

Name	Goal	Lower Limit	Upper Limit	
Time	is in range	60	170	
Temp	is in range	120	160	
% NaOH	is in range	2	5	
Yield	maximize	1.107	1.806	
Solution				
Number	Time	Temp	% NaOH	Yield

1	168.11	160.00	2.12	1.808
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**Table A3.** TG-DTA data for CPL showing the corresponding weight loss per temperature range applied on the sample.

Peak	Peak Temperature (°C)	Weight loss (%)	Temperature Range (°C)
1	92.17	10.237	30.00 – 186.96
2	345.90	34.603	186.96 – 579.57
3	626.54	54.854	579.57 – 690.23
4	763.10	0.306	747.69 – 776.41



**Figure A1.** TG-DTG curve of CPL

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