

## Full Length Article

## Enabling biomass combustion and co-firing through the use of Lignocol

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## ABSTRACT

We have developed a biomass-derived solid fuel, referred to as Lignocol, that can reduce sulfur and nitrogen emissions from coal-fired power plants without suffering the ash fouling and loss of boiler capacity usually associated with co-firing biomass. To produce Lignocol, lignocellulosic biomass was pyrolyzed to vapors that were condensed as distinct fractions of bio-oil. The heavy ends of bio-oil, consisting primarily of anhydrosugars from holocellulose and phenolics from lignin in the biomass, was subjected to a water extraction to produce separate streams of water-insoluble phenolics and water-soluble sugars. The phenolics were cured at 105–220 °C to evaporate water and expedite cross-linking reactions that solidified the phenolic liquid into Lignocol. The higher heating value (HHV) and particle density of Lignocol were similar to several commercially significant coals in the United States while the nitrogen and sulfur contents were only 0.26 wt% and 0.02 wt%, respectively, much lower than typically found in coal. The ash content of Lignocol was less than 0.5 wt% compared to 3–10 wt% for coals. Lignocol was structurally stable in the presence of water and only leached 8.52 ppm and 9.03 ppm of aromatics at pH 4.2 and 5.0, respectively. Lignocol was combusted at 650 °C in a 100 g/h fluidized bed reactor to determine feeding characteristics. Neither melting in the feeder system nor agglomeration in the reactor were observed. Based on these observations, Lignocol has excellent prospects for high levels of co-firing with coal in power plants and replacing wood pellets in dedicated biomass-firing while avoiding the usual challenges of burning biomass.

## 1. Introduction

Coal remains an important energy resource worldwide [1]. However, combustion of coal releases a number of air pollutants chiefly sulfur dioxide (SO<sub>2</sub>), nitrogen oxides (NO<sub>x</sub>), mercury and particulate matter [2]. It is also a major source of greenhouse gas emissions into the atmosphere, predominantly as carbon dioxide [3].

Co-firing biomass with coal has the potential to reduce most of these emissions [4]. However, co-firing has several operational disadvantages [5]. Its lower bulk density and high moisture content, compared to coal, results in higher transportation costs. Differences in physical properties complicates direct blending of biomass with coal, making necessary separate storage, handling and feeding of biomass. The lower heating value and moisture content of biomass compared to coal results in boiler derating. Relatively high values of alkali exacerbates ash fouling while some forms of biomass contain high levels of chlorine, which increases corrosion of boiler tubes. Finally, the composition of ash in biomass is distinct from coal ash and may not meet ASTM specifications for concrete applications. Accordingly, co-firing can increase the cost of

ash disposal.

We propose to mitigate these problems of burning biomass by first processing it into solid fuel known as Lignocol that has potential to dramatically reduce moisture and ash content while increasing fuel density and heating value compared to raw biomass. The process for its production is illustrated schematically in Fig. 1. Lignocellulosic biomass is subjected to fast pyrolysis to produce bio-oil, biochar, and non-condensable gases [6]. The bio-oil is fractionated into several streams including heavy ends consisting of a mixture of phenolic oil derived from lignin in the biomass and sugars from plant polysaccharides [7]. A simple liquid-liquid extraction separates the phenolic oil from the sugar. The phenolic oil, a viscous reactive liquid, is cured to solid Lignocol. The biochar, representing 12–15 wt% of the pyrolysis products, is a carbonaceous solid that could also be used as boiler fuel. However, because much of the original ash in the biomass reports to the biochar, it might be more productively used as soil amendment where these inorganic compounds, especially the potassium and phosphorous, serve as plant nutrients [8] and the carbon serves as carbon sequestration agent [9].

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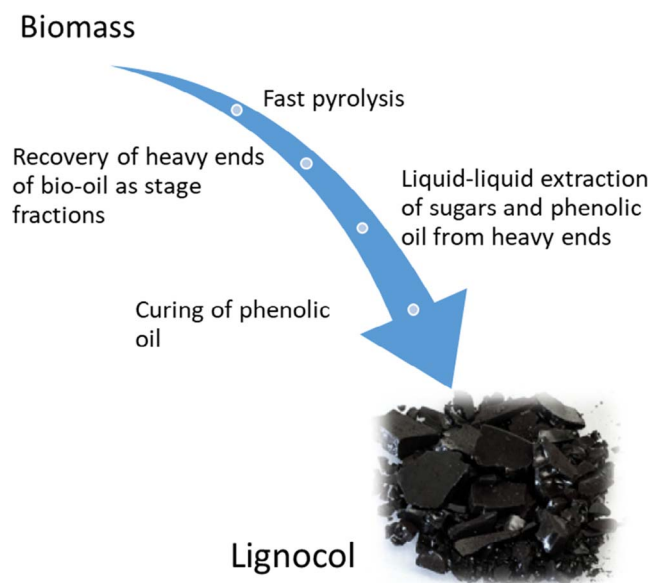


Fig. 1. Process for the production of Lignocol from lignocellulosic biomass.

The goal of this study is to investigate the properties of Lignocol as a solid boiler fuel and compare them to those of several types of coals in the United States. Red oak, high lignin content biomass, was pyrolyzed in a continuous, 8 kg/h process development unit to produce bio-oil fractions from which the phenolic oil was separated and cured into Lignocol. A wide range of properties were evaluated including density, heating value, elemental and proximate analysis, trace element composition, grindability, stability and leaching when exposed to water, and combustion behavior.

## 2. Materials and methods

### 2.1. Biomass characterization and reactor configuration

Red oak was used for the production of Lignocol. The ultimate and proximate analyses and heating value of the red oak is given in Table 1. As-received biomass was passed through a 60 hp hammer mill equipped with a 3 mm screen to produce particles ranging from 200  $\mu\text{m}$ –3 mm. The red oak particles were pyrolyzed in an 8 kg/h fluidized bed reactor operated at 500 °C. The biomass feed rate was 7.78 kg/h with 114 SLPM nitrogen flow and 2.5 SLPM helium flow.

The reactor was fitted with a fractionating bio-oil recovery system that collects the bio-oil into distinct stage fractions (SF) according to boiling point (Fig. 2). The reactor is described in detail by Rover et al. [7].

The bio-oil was 60–70% of the products of fast pyrolysis of red oak. The first two stages of the reactor (SF 1 and SF 2), the heavy ends, typically condense 40–50% of the produced bio-oil, and consist mostly of monomeric sugars and phenolic monomers, dimers and oligomers. These two fractions contain the vast majority of the lignin derived bio-oil. The middle fractions (SF 3 and SF 4) collect the monomeric

Table 1

Ultimate and proximate analyses and higher heating values (HHV) for the red oak feedstock. Ultimate analysis is on dry basis (db) with the oxygen calculated by difference.

Ultimate Analysis (wt% db)		Proximate Analysis (wt%)	
Carbon	50 $\pm$ 0.43	Moisture	7.1 $\pm$ 0.90
Hydrogen	4.8 $\pm$ 0.06	Volatiles	79 $\pm$ 0.48
Nitrogen	0.40 $\pm$ 0.00	Fixed carbon	13 $\pm$ 1.1
Sulfur	0.03 $\pm$ 0.004	Ash	0.33 $\pm$ 0.08
Oxygen	45 $\pm$ 0.49	HHV (MJ/kg)	19 $\pm$ 0.09

materials such as phenol, condensing approximately 10% of the produced bio-oil. The light ends (SF 5 and SF 6), representing 45% of the bio-oil, collect light oxygenates (i.e. furfural, hydroxymethylfurfural, acetic acid, etc.) and the water originating from the biomass and reaction water produced during pyrolysis. On average the water content in SF 5 is 60–80 wt%. Further descriptions of the condensed fractions and the chemical constituents of the bio-oil can be found in Rover et al. [7].

### 2.2. Test methods

Liquid-to-liquid extraction using water as the solvent is applied to separate the water-soluble material from the water-insoluble phenolic compounds. A 1:1 (w/w) mixture of water to SF 1 and SF 2 is mechanically stirred with a drill press equipped with a stainless steel open paddle for 10–15 min. The samples are placed on a shaker table (MaxQ 2506, Thermo Scientific®, Hanover Park, IL) for 30 min at 250 motions  $\text{min}^{-1}$  and centrifuged (accuSpin 1R, Thermo Scientific®, Hanover Park, IL) at 2561g force for 30 min. The water-soluble portion is then decanted. The remaining water-insoluble fraction, referred to here as phenolic oil, is the basis for Lignocol.

To cure the liquid phenolic oil into solid Lignocol, samples ranging from several grams to 300 g were heated in a laboratory oven (IsoTemp, Thermo Scientific, Hanover Park, IL, U.S.) to 105–220 °C for  $\leq$  2 h. Cured samples were broken into lumped Lignocol with a hammer. Since many boilers are based on burning pulverized coal, we also evaluated mechanical pulverization of Lignocol in a ball mill at 400 rpm for 30 min with 1 min intervals between 5 min reverse rotation. The ball milled Lignocol was sieved using a 75  $\mu\text{m}$  sieve to determine whether it would agglomerate, which would affect its combustion behavior in a pulverized coal boiler.

Moisture content of the phenolic oil bio-oil was determined by a MKS 500 Karl Fischer Moisture Titrator (Kyoto Electronics Manufacturing Co., Ltd, Kyoto, Japan) using ASTM E203 Standard Test Method for Water Using Karl Fischer Reagent. The Lignocol was weighed after curing with comparisons made to the original wt% moisture in the phenolic oil.

The proximate analysis was accomplished using thermogravimetric analysis (TGA) on a Mettler-Toledo TGA/DSC 1 STARe System. The sample size ranged from 15 to 25 mg. The program was set to ramp from 25 °C to 105 °C at a rate of 10 °C/min and held for 40 min. The temperature was then ramped to 900 °C at a rate of 10 °C/min and was held for 20 min. Nitrogen was flowed through the system at 100 mL/min. The nitrogen flow was replaced with 100 mL/min of air and the samples were held for 20 min at 900 °C.

Higher heating values were determined with a Parr 6400 Calorimeter (Parr Instrument Company, Moline IL). Benzoic acid was used as the standard. Trials were done in duplicate and standard deviation determined.

Ultimate analysis was done utilizing an Elementar, vario MICRO cube (Elementar, Hanau, Germany) elemental analyzer, with oxygen determination by difference. A minimum of three trials was performed with standard deviation calculated.

Density of Lignocol was measured by volume displacement. Ten trials were performed with standard deviation determined.

Inductively coupled plasma (ICP) was performed on the Lignocol to determine inorganic metal content using an optical emission spectrometer (OES) Optima 8000 (PerkinElmer, Shelton, CT). Approximately 1.0 g of the Lignocol was used for the analyses. Concentrated nitric acid (10 mL) was added to each sample and digested in an Anton Parr Microwave Reaction System (Ashland, VA) following a modified microwave assisted pressure controlled Environmental Protection Agency (EPA) 3052 digestion method. After digestion was complete, the samples were diluted to a total volume of 100 mL with distilled water and filtered with 0.45  $\mu\text{m}$  glass microfiber followed by testing [10]. Six replicates of the Lignocol and a blank were analyzed. The standards

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