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Co-processing coal and torrefied biomass during direct liquefaction

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ABSTRACT

Co-liquefaction of coal and torrefied biomass was performed in a mechanically stirred and electrically heated Parr reactor to evaluate effects of co-liquefaction and torrefaction on product yields and properties. The test samples were coal, red maple, yellow-poplar, torrefied red maple and torrefied yellow-poplar. Sample mixtures of coal-red maple, coal-yellow-poplar, coal-torrefied red maple, and coal-torrefied yellow-poplar were prepared in the weight ratio of 1:1. About 10 g of sample and 100 ml of tetralin were placed inside the reactor for each test. The reaction temperature was 300 °C and the hydrogen gas pressure was set at 600 psi (cold). The results showed that oil yields obtained from co-liquefaction of either coal-torrefied red maple, or coal-torrefied yellow-poplar mixtures were higher than the weighted average yields from coal and biomass, separately.

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1. Introduction

With decreased petroleum reserves and increased energy demand, direct liquefaction of coal is again drawing attention in some countries [1–4]. Coal contains complex macromolecules, which are built primarily of carbon and hydrogen but it also contains significant amounts of oxygen, sulfur, and nitrogen [5]. Typically, the constituent units of coal are mostly substituted aromatics or hydroaromatics [5]. The hydrogen-to-carbon ratio of coal is significantly lower than that for petroleum and therefore, converting coal into liquid fuels requires the addition of substantial amounts of hydrogen [6]. Direct coal liquefaction is performed in a solvent at elevated temperature and pressure of hydrogen, accompanied by the hydrocracking of the dissolved coal with H₂ and a catalyst [1]. High temperatures (more than 300 °C) cracks coal macromolecules into small radical fragments that are then stabilized by hydrogen atoms provided by a solvent or a hydrogen rich gas [5]. The solvent also acts as hydrogen donor and a carrier of gas-phase hydrogen to the solid coal [7]. The high pressure keeps the solvent and products in the liquid phase at elevated temperatures and maintains a high partial pressure of hydrogen [5]. The use of catalyst is one of the important factors in liquefaction as it can increase reaction kinetics and improve yield of products [8]. The process produces pre-asphaltene, asphaltene, oil fractions and other solid hydrocarbons. However, direct liquefaction of coal is not considered very competitive due to the high cost of the hydrogen donor solvent and high carbon emissions associated with the process [5,7]. One method of reducing the high cost of hydrogen and cutting carbon emissions during direct coal liquefaction is to mix biomass with coal during liquefaction, thereby altering the process to one of co-liquefaction [9].

In co-liquefaction, coal and biomass are mixed and co-fed into a sealed reactor in the presence of a solvent. The most preferred solvent is tetralin due to its high thermal stability and high hydrogen donating ability [9–16]. The reactor is pressurized with hydrogen. The reactor is then heated to a temperature usually between 300 and 500 °C. In the co-liquefaction process, thermal fragmentation of coal produces organic free radicals, which combine with hydrogen supplied from a hydrogen donor solvent or the biomass itself [17].

One advantage of co-liquefying coal and biomass is a reduction in greenhouse gas emissions [7,9]. Another advantage is enhanced oil and gas yields [10]. Previous research has shown that the addition of biomass to coal in co-liquefaction had a positive effect on liquefaction

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product yields [9,15,18,19]. However, several factors, for example, type of biomass, type of coal, biomass pretreatment, type of solvent, temperature, pressure and holding time have direct impact on product yield and product quality [20].

For example, co-liquefaction of bagasse with coal increased the oil yield from 10.9% to 47.8% [15]. Additionally, the combined oil and gas yield for co-processing was reported to be higher than the average arithmetical value of yields obtained from the individual feeds (lignite coal and cellulosic waste material) [9]. Co-liquefaction of coal and biomass not only increases oil yields but also changes the chemical characteristics of the produced oils. For example, the hydrogen-to-carbon ratio in the water-soluble fraction of the liquefaction product increased when coal was co-processed with cellulose [18]. Hua et al. also [21] reported that co-liquefaction of coal and rice straw in the presence of an FeS catalyst had a synergistic effect during co-pyrolysis of Shenfu coal and rice straw. Shui et al. [22] reported that sawdust accelerated thermolysis of Shenhua coal by enhancing volatilization of the coal structure. Most of these investigations have found that the total conversion increased when biomass was processed with coal. However, not much synergy has been observed between coal and biomass, probably, because the biomass has high H/C (hydrogen-to-carbon) and O/C (oxygen-to-carbon) ratios [9]. In addition, Stiller et al. [11] concluded that lignin content of biomass plays an important role during co-liquefaction. They reported that addition of cow manure produced the highest oil yields, among other biomass (sawdust, horse and cow manure and a commercial manure) tested due to its very high lignin content [11]. Similarly, Löwe et al. [23] reported the highest oil yields when coal was processed with lignin during direct liquefaction. Coughlin and Davoudzadeh [24] reported that coal and lignin depolymerize under mild conditions in presence of tetralin solvent to produce a filterable liquid product with yields greater than that of the weighted average liquefaction yields of both components reacted independently. Therefore, biomass may be preprocessed to reduce the H/C and O/C ratios, and increase reactivity and lignin content via torrefaction prior to co-liquefaction [25].

During the torrefaction process, biomass is subjected to moderate heating (200 °C–300 °C) in a low-oxygen environment [26] to decompose hemi-cellulose, completely, and cellulose, partially, thereby producing torrefied biomass [25]. Typically, torrefaction yields 79%–95% torrefied biomass with lower H/C and O/C atomic ratios than the original biomass [25]. The torrefied biomass has high carbon content [27]; low H/C and O/C atomic ratios [28]; high lignin content [20] and brittleness [29]. Biomass type, torrefaction temperature, and residence time have a major influence on the characteristics of torrefied biomass [30,31]. However, the combination of 260 °C and 1.0 h residence time seems sufficient to produce good quality torrefied biomass from woody feedstock [30].

There has been a growing interest to find the compounds associated with liquefaction products using supercritical solvent extraction coupled with HPLC and NMR [32,33], Mass Spectroscopy [34], and Attenuated Total Reflectance Infrared Spectroscopy (ATR-IR) [35–41], and a combination of multiple techniques [42]. Yuan et al. [32] reported that oils produced from coal liquefaction (at 380 °C and 10 MPa hydrogen pressure) contained saturates (22.1 wt%), monocyclic aromatics and dicyclic aromatics (20.3 wt%) and polar compounds (39.4 wt%). Analysis of the pyridine soluble fraction of a distillation residue and a further processed liquid product in a coal liquefaction process using electrospray ionization (ESI) Fourier Transform ion cyclotron resonance mass spectrometry (FT-ICR MS) was reported by Wu et al. [34]. They reported that the residue contained more chemically distinct species of higher average molecular weight, as well as a wider and higher distribution in the number of CH₂ groups, and higher aromatic content than coal liquid.

ATR-IR is a suitable technique to identify functional groups. Previous attempts have been made to identify the functional groups associated with the chemical compounds present in liquefaction products [40,43,44]. These studies have found that aldehydes, alcohols, phenols, amines, esters, carboxylic acids, aliphatic hydrocarbons, ethers and aromatic compounds are mainly present in direct liquefaction products [40,43]. Damin et al. [40] used Fourier Transform Infrared Spectrometry (FTIR) to characterize raw Chinese brown coal and coal that was ultrasonically extracted with carbon disulfide (CS₂). They reported that the raw Chinese brown and extracted coals had a great abundance of amidocyanogen, carbonyl, aromatic and heteroaromatic rings, and single carbon-heteroatom bonds, which was an indication of the possible presence of compounds such as alcohols, phenols, amines, esters, carboxylic acids, aliphatic hydrocarbons, ethers, and aromatic or heteroaromatic compounds. Teng et al. [39] used FTIR to characterize tetrahydrofuran soluble coal liquefaction products and whole process oil from Pittsburgh coal for aromatic and aliphatic hydrogen (Proton) distribution, hydroxyl groups analysis and oxygen, hydrogen, and carbon analyses. The –OH stretch band found between 3200 and 3400 cm⁻¹ was used to determine the phenolic –OH concentration from measurements on the THF-soluble portions of the samples [39]. They also reported that the aromatic-to-total hydrogen ratio measured using FTIR analysis was in agreement with that derived from the ¹HNMR method. Therefore, use of ATR-IR/FTIR provides some key information about functional groups present in the liquefaction products.

Therefore, torrefied biomass is expected to have certain characteristics suitable for enhancing liquefaction of coal, for example low H/C and O/C atomic ratios and high lignin content. The combination of the H/C and O/C atomic ratios are important for direct liquefaction [45,46] because the theoretical efficiency of liquefaction increases with increasing H/C ratio and with decreasing O/C ratio. The objective of this study was to evaluate of effect of co-liquefaction of coal with torrefied biomass on product yields.

2. Experimental procedure

2.1. Materials

Bituminous coal (C) from Kingwood, West Virginia was used for co-liquefaction of coal and biomass. Samples of freshly cut yellow-poplar (YP) and red maple (RM) were collected from the West Virginia University Research Forest. The two types of wood samples were chosen due their different ultrastructural characteristics (pore distribution) and specific gravity, which might play role in co-liquefaction. RM is a ring porous hardwood with green specific gravity of about 0.56 [47] whereas YP is a diffuse porous hardwood with green specific gravity of about 0.46 [47]. The biomass samples were chipped, dried at 103 °C for 24 h, and then ground to less than 1 mm particle size.

2.2. Torrefaction

The dried and ground samples were torrefied at 300 °C for 1 h under a nitrogen atmosphere to produce torrefied red maple (TRM) and torrefied yellow-poplar (TYP) in a thermo-gravimetric analyzer (Model: TGA 701, Leco Corporation, St Joseph, MI). A temperature of 300 °C was selected for maximizing lignin content [48] while obtaining high reactivity biomass and a residence time of one-hour was chosen based

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