



Co-gasification of pine and oak biochar with sub-bituminous coal in carbon dioxide

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ABSTRACT

Pine and oak biochars derived as byproducts of demonstration-scale pyrolysis, and blends of these two feedstocks with Powder River Basin coal, were gasified in a carbon dioxide environment using a modified drop tube reactor (MDTR) and a thermogravimetric analyzer (TGA). The impact of gasification temperature on conversion kinetics was evaluated from the temporal evolution of major product gases in the MDTR as measured using a mass spectrometer. Random pore modeling was conducted to simulate gasification in the MDTR with favorable results. The MDTR and TGA were used to conduct gasification for assessment of non-linear additive effects in the blends. Additive analysis of the blends showed deviation from the experimental blend results, indicating inhibiting effects of co-gasifying the biochar and coal. Inhibitory effects are more significant for oak than pine and more pronounced in the TGA at lower gasification temperatures. Results are discussed in the context of feedstock and reactor type.

1. Introduction

Increasing concerns about emissions associated with traditional fossil fuel sources, like coal, have driven a need to develop alternative energy sources that reduce greenhouse gas emissions without compromising energy availability and security. Of available low carbon energy options biomass is attractive because it is widely available and, unlike wind and solar energy, it does not suffer from intermittency issues. Biomass can be utilized as a feedstock for energy production through several major thermochemical conversion routes, including gasification. Gasification is a partial oxidation process whereby a solid carbonaceous feedstock, such as biomass, is converted to a gaseous mixture of carbon monoxide (CO) and hydrogen (H₂), known as synthesis gas or syngas, as well as carbon dioxide (CO₂) and water (H₂O). This product gas is subsequently combusted for power production or upgraded to higher value liquid products (Basu, 2013; Digman, et al., 2009). Extensive research has been conducted on the gasification of coal, particularly in a CO₂ environment to produce a CO₂-rich product stream with the eventual goal of integrating CO₂ capture and sequestration for an overall clean conversion process (Bell et al., 2010; Minchener, 2005; Wang and Bell, 2015). Despite the potentially smaller carbon footprint of coal gasification as compared to direct combustion, coal gasification does not achieve the larger goal of reducing fossil fuel consumption.

In order to reduce fossil fuel use, biomass gasification has been

explored as a fuel-flexible route of power production from renewable feedstocks. In addition to a potentially significant reduction in net carbon emissions as compared to coal, biomass offers a number of additional advantages as a feedstock for gasification, including lower ash and sulfur contents as compared to coal (Howell et al., 2017; Taba et al., 2012; Ding et al., 2014), making the clean-up of gasification product gases easier and less expensive. A major shortcoming of widespread power production from biomass gasification, however, is the existence of some uncertainty in feedstock availability and composition due to seasonal variability and other natural factors (Taba et al., 2012; Ding et al., 2014). Thus, in consideration of the advantages and disadvantages of gasifying coal or biomass alone, co-gasification of coal and biomass feedstock blends has emerged as a viable strategy to reduce fossil fuel consumption and emissions (Taba et al., 2012). In order to advance this power production strategy, a thorough understanding of feedstock properties and co-conversion behavior is required.

Raw biomass has some qualities that limit its direct use as a fuel, including high moisture and volatile contents and low energy density (Chen et al., 2011). High moisture and low energy density are closely related, indicating an advantage in drying biomass prior to use (Howell et al., 2017; Phanphanich and Mani, 2010). The generally high volatile content of biomass has been shown to render biomass more reactive than coal, which can cause troublesome time scale discrepancies during

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their co-conversion (Chen et al., 2011; Sahu et al., 2014). All of these traits have been shown to be improved through thermal pretreatment of the biomass, either through mild pyrolysis, called torrefaction, or moderate-to-severe pyrolysis. Increased pretreatment severity has been shown to increase the C:H and C:O ratios of the biomass, rendering the pretreated biomass composition to be more similar to coal (Medic et al., 2012; Couhert et al., 2009). Furthermore, in addition to pyrolysis being explored as a biomass pretreatment process, pyrolysis is receiving significant attention as a primary process because valuable bio-oil products can be produced; thus, gasification provides a highly beneficial outlet for biochar which is produced as a pyrolysis byproduct (Brewer et al., 2009). While several process parameters can affect the gasification process, temperature and feedstock compositions have been found to be among the most impactful factors in determining conversion rates and product yields (Taba et al., 2012; Lahijani et al., 2015). Feedstock properties, including volatile and mineral contents, have been found to particularly impact the gasification behavior of fuel blends (Tilghman and Mitchell, 2016). Biomass reactivity, reflective of volatile content, has been found to significantly affect the reactivity of biomass-coal fuel blends, with increased pyrolysis pretreatment temperature translating to lower reactivity of fuel blends (Gao et al., 2010).

The impact of mineral content, particularly alkali and alkaline earth metals (AAEM), on catalytic or inhibitory behavior in coal gasification has been widely studied. Several studies have examined the effects of mineral content on co-gasification of different biomass feedstocks with coal as a result of the differing compositions of the feedstocks (Gao et al., 2010; Kajitani et al., 2010). Observations of the presence or absence of synergistic effects in co-gasification of fuel blends has not been consistent among studies, however (Mallick et al., 2017; Masnadi et al., 2015; Zhu et al., 2008). High concentrations of alkali ash components, such as potassium, have been observed to have both synergistic and inhibitory effects in a study of co-conversion of switchgrass with fossil fuels of varying ash contents, depending on the fuel mixture (Habibi et al., 2013). Co-gasification of coal and pine sawdust has been observed to have increased reactivity as compared to simple algebraic additive prediction, with increased synergy observed at higher biomass fractions and process temperatures (Jeong et al., 2014). In contrast, a study of co-gasification of bituminous coal and cedar bark did not show a synergistic increase in reactivity (Kajitani et al., 2010). An extensive summary of the various additive, synergistic and inhibitory effects observed in the literature has been presented by Mallick et al. (2017).

A number of reactor apparatuses have been used in gasification studies, including thermogravimetric analyzers (Habibi et al., 2013; Wei et al., 2017), fixed bed reactors (Jeong et al., 2014), fluidized bed reactors (Aluaddin et al., 2010), and entrained flow reactors (Gao et al., 2010). Drop tube furnaces are commonly used in experimental studies because they mimic the high heating rate conditions that fuels would undergo in a pulverized fuel reactor in commercial plants (Gao et al., 2010). Kinetic modeling of coal, biomass, and biomass-coal blend gasification has been conducted using the volume reaction model, which assumes homogeneous reaction throughout a reacting particle; the shrinking core model, which assumes a progressing reaction front from the external surface towards the core of a reacting particle; and the random pore model (RPM), which considers the development and growth of pore surfaces in the surface area available for reaction. The RPM approach has been found to perform well in modeling of gasification behavior, and has been used to derive kinetic parameters in coal gasification and co-gasification of coal and biomass (Kajitani et al., 2010; Jeong et al., 2014).

In the present study, biochars produced from two biomass feedstocks, pine and the lesser studied oak, were gasified alone and blended with sub-bituminous Powder River Basin coal. The biomass feedstocks utilized in the present study are rather unique among gasification studies, as they were produced as a byproduct of a demonstration-scale pyrolysis process; thus, the pyrolysis process was designed for the production of bio-oils, not char, leaving a moderately pyrolyzed char

available for energy production via gasification. Additionally, the coal utilized in the present study was not charred prior to gasification in order to test a realistic feedstock and feedstock mixture that might be implemented in an industrial gasifier. Gasification was conducted over a range of temperatures in a CO₂ environment to compare feedstock behavior and elucidate the impact of temperature and fuel blending on conversion kinetics, and the presence or absence of non-additive behavior. A modified drop tube reactor (MDTR) was used to explore high heating rate behavior, and kinetics were assessed by random pore model analysis. A thermogravimetric analyzer (TGA) was utilized to assess conversion behavior with close contact between biomass and coal throughout the duration of the reactions. Given the inconsistency in findings of synergy or inhibition in the literature, these two reactor apparatuses were used together in this study for a thorough and simultaneous assessment of synergistic and inhibitory behavior in gasification of the fuel blends. Assessment of additive and non-additive behavior in the fuel blends during conversion in the MDTR and the TGA were evaluated and compared.

2. Materials and methods

2.1. Sample preparation

The biochar used in this study was collected after pyrolysis of biomass feedstocks in the Thermochemical Process Development Unit at the National Renewable Energy Laboratory (NREL) in Golden, CO. The primary purpose of the pyrolysis reactor was to collect bio-oils produced by pyrolysis, while the remaining biochar used in this study was a byproduct of the process. The two biomass feedstocks pyrolyzed for this study were pine and oak. Biomass feedstock was fed into an entrained flow reactor using heated nitrogen as a carrier gas. Solid char was produced as volatile species were released, and the solid products were separated outside of the reactor using cyclonic separators. The collected biochar was then passivated off-line using a low percent air. The solid char product was collected and provided for this study. The pyrolysis temperature and processing conditions for both feedstocks were 480 °C with a residence time of approximately 3.5 s, which are representative of conditions used for bio-oil production by NREL. Samples were sieved to collect particles between 106 and 250 μm and stored in sealed glass containers inside a desiccator prior to gasification tests in order to preserve their composition.

The coal used in this study was sub-bituminous coal from the Powder River Basin provided by Idaho National Laboratory (INL). The coal was dried and sieved to the same size range as the biochar particles, 106–250 μm, and then stored in a sealed glass bottle in a desiccator in order to preserve its composition. Sieved coal and biochar particles were blended at a 50/50 wt ratio prior to co-gasification experiments in order to create pine biochar-coal and oak biochar-coal blends. The blends were mixed by manual agitation before each gasification test to ensure a uniform distribution of coal and biochar particles in each test sample. Experimental test repeatability further supported that sample composition consistency and blend homogeneity were achieved.

2.2. Modified drop tube reactor

Gasification experiments were carried out in a modified drop tube reactor (MDTR) as detailed in Wang and Bell (2015). The reactor consisted of an outer tube made from Inconel 600 (33 mm ID × 38 mm OD × 1067 mm length) and an inner tube made from quartz (28 mm ID × 31 mm OD × 241 mm length). Below the quartz frit in the inner tube, upon which the sample was dropped, the quartz tube diameter reduced to 4 mm ID × 6 mm OD. The total reactor length was 1524 mm. Each sample was loaded into the upper portion of a loading chamber which was separated from the reactor by a ball valve. Once the reactor was heated to the desired gasification temperature, the ball valve was opened, dropping the test sample into the reactor. A

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