



Research article

Co-Hydrothermal Carbonization of coal-biomass blend: Influence of temperature on solid fuel properties



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ABSTRACT

Co-Hydrothermal Carbonization (Co-HTC) was performed on a blended feedstock of coal and miscanthus. The main goal of this work was to evaluate the synergistic effects of miscanthus on coal during Co-HTC. Fuel quality was assessed for all hydrochars by evaluating mass yields, energy content, ultimate analysis, and proximate analysis. Calculation of combustion parameters showed experimental ignition and burnout indices of Co-HTC 260 °C hydrochar were 29.0% and 26.5% lower than theoretical, non-interacting indices, respectively. Hydrochars shared the benefits of low sulfur and low ash content of miscanthus but maintaining higher energy content of coal. Hydrochars produced at 260 °C had energy contents as high as coal ($27.3 \pm 0.6 \text{ MJ kg}^{-1}$) and 73% less ash content and 74% less sulfur than raw coal as a result of the more acidic environment produced by miscanthus decomposition. Furthermore, hydrochars were homogeneous as miscanthus-derived hydrochar was formed on coal surface according to SEM imaging and verified by the reduced pore width from nitrogen adsorption. Co-HTC hydrochars were pelletized in a single-press pellet press. Both mass and energy densities of Co-HTC pellets were increased with the increase of Co-HTC temperature. For instance, energy densities of pelletized Co-HTC hydrochars were increased to 32.4 GJ m^{-3} , whereas HTC coal contains energy density of 28 GJ m^{-3} .

1. Introduction

Energy demand continues to increase rapidly as the population of the world grows. The current world population of 7.5 billion is expected to reach nearly 10.3 billion in 2040 while energy demand is projected to reach 42 quadrillion BTU in 2040 [1,2]. Meeting the energy demand without further contribution to global warming requires high output from our current fossil energy technologies as well as the contribution of renewable and sustainable resources. In the U.S., coal-fired power plants accounted for approximately 40% of electricity generation in 2015 and coal consumption has been expected to increase by 17.6% quadrillion BTU to 2040 [2]. Biomass, on the other hand, is the largest and most abundant source of renewable energy [3]. Biomass contributes zero to net negative greenhouse gas emission, as plants uptake carbon dioxide during its growth [3]. Therefore, co-firing biomass with coal in existing coal-fired power plants has been shown to reduce coal-derived harmful pollutants and greenhouse gas emissions [4]. However, overall efficiency is sacrificed due to low energy content and low bulk density of biomass. Moreover, the wet nature of biomass as well as seasonal availability create more challenges for co-firing options. Therefore, viable co-firing requires biomass pretreatment to produce a

higher density and hydrophobic feedstock, which is compatible with coal.

Hydrothermal carbonization (HTC) is a promising wet biomass treatment process for producing biofuel and upgrading solid fuel [5]. The process conveniently uses residual water as the reaction media, due to its solvent properties at high temperatures and pressures [6]. The moisture content of the feedstock varies between 30 and 90%, depending on biomass type, harvesting time, and storage facility [3]. As HTC requires all the biomass submerged in water, biomass with low moisture content might require additional water [7]. Subcritical water at around 200–260 °C has a very high ionic constant and low dielectric constant, which results water to be more reactive and behaves similarly to a non-polar solvent [8]. As a result, a residence time of 5–30 min of treating low fuel quality biomass, solid-hydrophobic hydrochar is formed along with liquid and gaseous byproducts [9,10]. Hydrochar is quite hydrophobic, friable, and more energy dense [10,11]. Moreover, the exothermic nature of the HTC reaction has shown to have minimum to no net energy input for biomass upgrading in the lab-scale setting [12]; the energetics of fully scaled up processes have not yet been fully understood. Although hydrochar itself shows significantly higher potential to replace coal, well established coal-processing technologies

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and the maturity of coal infrastructure has made coal more favorable in cost and availability thus making it more suitable to use in the power plants.

In addition to being useful for upgrading biomass, HTC treatment of coal can also be benefitted by having energy content increased while decreasing other undesirable impurities such as fraction of total ash and oxygen containing volatile fractions [13,14]. Specifically, others have shown that performing HTC of low rank coals (e.g. lignite) between 150 °C and 350 °C successfully upgrades their rank as a result of decreasing oxygen and moisture contents [14–16]. Previous literatures discussed HTC of sub-bituminous coal, but solid fuel properties were not entirely investigated over ranging reaction temperatures [17,18]. To the authors' knowledge, higher rank coals like bituminous and anthracite coals have not been examined under hydrothermal treatment, especially in the temperature range of 150–260 °C.

Co-Hydrothermal Carbonization (Co-HTC), where coal and biomass blends are treated with subcritical water, has some additional advantages than HTC of individual coal and biomass. Mild acidic conditions may increase the sulfur removal efficiencies while upgrading the low rank coal [19]. HTC of biomass produces acidic solutions by dehydration and decarboxylation reactions [9]. Therefore, Co-HTC might further upgrade the coal synergistically using acids produced by biomass during HTC. On the other hand, presence of metal or porous structure influence the condensation-polymerization reactions, therefore, overall hydrochar yield increases [20]. Coal's surface can be a potential condensation site during Co-HTC. As a result, coal will enhance the overall hydrochar formation from biomass during Co-HTC. Furthermore, Co-HTC might offer a better homogeneity of coal-biomass blends and potentially reduce the expensive solid-solid mixture of two solids with different densities.

Very few researchers have looked into the HTC of mixed feedstocks, and even less have considered HTC of biomass and coal mixtures. Zhang et al. showed the potential of hydrochar customizability from hydrothermally treating two feedstocks simultaneously. Positive synergistic effects were observed in terms of ash reduction, nutrient concentration, an increase of functional group densities, which allude to interaction during treatment [21]. Nonaka et al. examined the characteristics and effects of different biomass to coal ratios at a single reaction temperature of 300 °C [22]. The study concluded that the solid products become more hydrophobic after HTC treatment and chemical composition and fuel quality were independent of the mixing ratio. Although, coal is not often pelletized, however, HTC produces fine powder that requires pelletization to increase mass and energy densities as well as to enhance storability and transportability [23]. Improper storability and transportability results in higher occupation of volume which can lead to higher costs, ultimately making certain technologies not economically viable. It has been shown that pelletization of HTC biomass products not only increases durability and storability, but also significantly increases its mass and energy densities [24]. Pelletization of lignite coal and treated biomass have also been reported as beneficial, as fuel quality was increased and treated biomass was able to hold the coal in an interlocked matrix, resulting a strong and durable pellet [25]. Pelletization of certain fuels depends on a variety of parameters such as moisture content, glass transition phase, effective binders, etc. [26,27]. Among them, pelletization of coal heavily depends on effective binder, as hydrophobic coal does not show glass transition behavior [28]. Whereas, biomass-derived hydrochar are considered as effective solid binder [27].

Therefore, the goal of this study was to examine the fuel quality of solid products produced by Co-HTC of coal and miscanthus at various HTC temperatures. The main objective of this study is to reveal the synergistic effects including sulfur content, ash content, and hydrochar yield from Co-HTC compared to HTC of individual feedstocks. Nitrogen adsorption/desorption on the hydrochars are also examined to study physical interactions during Co-HTC. Furthermore, the effects of pelletization of hydrochars from Co-HTC and HTC were examined

considering biomass derived hydrochar acting as a solid binder.

2. Materials and methods

2.1. Materials

Miscanthus (*Miscanthus × giganteus*) and Clarion type #4A coal were the feedstocks for this study. Miscanthus was grown in Ohio University's Ridges Land Lab in the summer of 2016 and was harvested at the surface of the soil with a moisture content of 55%. The long stalks of miscanthus were chipped with a Del Morino Harvester (model number DM 100, Caprese Michelangelo – Italy). To attain a smaller particle size, the miscanthus was grinded with a Retsch ZM 200 grinder (Newtown, PA) using a number two blade. The chopped miscanthus, from the Retsch grinder, was then sieved for 30 min and everything between a size 20 and 60 mesh was collected and used as biomass in this study.

Meanwhile, Clarion type #4A coal is a bituminous coal with a moisture content of 10% that was obtained from Sands Hill Mining LLC., originated from Southeast Ohio. The coal was also grinded on the Retsch grinder similar to the miscanthus. The smaller coal produced was sieved for 30 min and all coal between a 40 and 60 mesh was collected. After both feedstocks were sieved, they were dried in an oven at 105 °C for 24–48 h and stored in a Ziploc bag prior to Co-HTC.

2.2. Experimental methods

All HTC and Co-HTC experiments were performed in a 500 mL 4575 Parr reactor (Moline, IL). Reaction temperature was controlled with a model 4848 Parr proportional-integral-derivative (PID) controller. Reaction pressure was not controlled rather monitored during the experiment. All HTC and Co-HTC experiments consisted of treating a 1:10 feedstock to deionized water ratio, which were all stirred at 120 rpm from initial reactor heating to reactor cooling to ensure product homogeneity. It is important to note that the Co-HTC experiments still used a 1:10 feedstock to deionized water ratio; however, the feedstock mixture consisted of a 50 wt% coal to 50 wt% miscanthus blend. The 1:1 coal to miscanthus blend was chosen for experimental simplicity as Nonaka et al. showed that hydrothermal treatment of different biomass-to-coal ratios were independent of chemical composition and other fuel properties [22].

HTC were performed, individually, on coal slurries and miscanthus slurries at temperatures of 200 °C, 230 °C, and 260 °C, while Co-HTC were performed on the coal-miscanthus slurry for the same reaction temperatures. In each experiment, the reactor was heated to the desired reaction temperature and held for a 30 min residence time. After the residence time was completed, heating was turned off and the reactor was cooled down to 75 °C by passing tap water through the reactor's internal cooling jacket. It took approximately 15–20 min to cool the reactor temperature from 260 °C to 75 °C. Gaseous products produced during the reaction were vented in a fume hood. The hydrochar was filtered using vacuum filtration with a Whatman 114 filter paper (25 µm) from the HTC process liquids. In order to remove finer hydrochar, the filtered process liquids were further filtered with vacuum filtration but using a Fisherbrand Q2 filter paper (1–5 µm). The filtered process liquids were refrigerated in no. 1730 Corning snap-seal containers while the separated hydrochars were dried at 105 °C in an oven for 24 h, then stored in the same type of snap-seal containers used for the process liquid. Co-HTC experiments were triplicated to ensure the reproducibility of the experimentation.

2.3. Characterization of hydrochars

The higher heating value (HHV) of untreated feedstocks and dried hydrochars were determined using a Parr 6200 adiabatic oxygen-bomb calorimeter (Moline, IL). Benzoic acid was used for the calorimeter's

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