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The effect of torrefaction on syngas quality metrics from fluidized bed gasification of SRC willow

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Short rotation coppice willow is proposed as a dedicated energy crop in the Canadian prairie region. A coppice willow hybrid known as SV1 (*Salix Dasyclados*) grown at the University of Saskatchewan was torrefied in a continuous torrefaction reactor at four temperatures (240, 260, 270 and 280 °C). The torrefied and control samples were then ground and gasified in a fluidized bed reactor at 900 °C with air and steam. The samples were characterized for ultimate composition and lignocelluloses. A unique HR-TGA method was used to determine the fraction of hemicelluloses, cellulose, lignin and ash in the torrefied and control samples. Syngas quality was evaluated based on gas yield and tar concentration. Tars were measured using a flame ionization detector and gas chromatograph. The syngas yield was found to increase from 2.02 to 2.47 m^3/kg_{SV1} between the non-torrefied and heavily torrefied samples. Tar yield was observed to decrease from 17.26 g/m^3 (mean for the control and the 240 °C conditions) to 9.21 g/m^3 (mean for the 260, 270 and 280 °C conditions) a reduction of 47%. The change in syngas quality coincides with the degradation of hemicelluloses below approximately 12% dry weight. More severe torrefaction had no additional effect on the syngas quality metrics.

1. Introduction

Torrefaction is a promising pretreatment for underutilized biomass resources in North America. A mild form of pyrolysis, torrefaction involves heating biomass in the absence of oxygen to between 230 and 300 °C [1]. The structure of the biomass changes as oxygenated compounds are volatilized. The main effect of interest is the reduction in the polymer hemicellulose from the biomass structure. Hemicelluloses begin to volatilize at approximately 250 °C [2]. Hydrolysis first causes hemicelluloses to depolymerize, followed by a series of acid and radical reactions which release acids and water [3]. The water and acids formed by these reactions are then available to also depolymerize cellulose and lignin, but to a much lesser extent. Overall, the result is a lower O/C and H/C ratio in the solid product that in turn, increases the energy density and reduces the required comminution energy. Three recent reviews summarize the state-of-the-art of torrefaction and the benefits to biomass processes [4–6]. In short, torrefied biomass can be conveyed, stored and pulverized in a similar manner to coal, a common fuel for electricity generation.

Independent of torrefaction, researchers have shown short-rotation coppice willow (SRCW) to yield significantly more energy and output significantly less GHG emissions [7]. Djomo et al. reviewed 26 studies and found, on average, SRCW to yield 36 times more energy per unit of fossil fuel energy input and 24 times lower GHG emissions compared to coal. Co-firing ratios in existing coal-fired power infrastructure are limited to approximately 10% due to the behavior of biomass in suspension burning systems [8]. Recently, a 100% substitution of coal with torrefied biomass in a pulverized coal-fired boiler was suggested with no loss of boiler efficiency or fluctuation in load [9]. With consideration of the associated operational problems (slagging, fouling and corrosion), the modeled reduction in CO_2 and NO_x emissions presented by Li et al. [9] is promising.

Beyond co-firing, torrefied biomass has been proven to be a valuable feedstock for gasification leading to a wider range of energy applications. Replacement of fossil derived transportation fuels with synthetic fuels from biomass-to-liquids processes is of particular interest owing to the lack of viable alternatives [10]. Early investigation into the area of gasification of torrefied biomass considered the benefits of handling and fluidization as a result of the characteristics of torrefied biomass. The reduction of net

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hemicelluloses through torrefaction was reported to result in ground biomass particles with a lower length-to-diameter ratio capable of smooth fluidization required for entrained flow (EF) gasification [11]. More specifically, particles could be pulverized to behave like Geldart type A powders as required for fast fluidization [12]. By studying torrefied wood gasified in a circulating fluidized bed (CFB), an EF reactor and an oxygen-blown EF reactor, Prins et al. [12] were able to conclude that, in addition to the improved fluidization characteristics, decoupling torrefaction reactions from the gasifier increases the efficiency of high temperature gasification. The increase in available energy is, however, dependent on the use of both the char and volatile gases from torrefaction in the gasification process. More recent studies have expanded the understanding. In a steam injected EF gasifier at 1400 °C, Couhert et al. [13] were able to produce 7%g/$g_{dry\ wood}$ more H_2 and 20%g/$g_{dry\ wood}$ more CO from torrefied versus non-torrefied beechwood, consistent with the increase in carbon and hydrogen fraction in the feedstock. In a similar study, Chen et al. [14] gasified sawdust without the application of steam. The study showed improvements in the syngas quality, but a reduction in the gasification efficiency unless the torrefaction gases were consumed in the gasifier.

Few studies relating to the gasification of torrefied biomass consider the 'cleanliness' of the product or syngas. Production of clean syngas accounts for the majority of capital expenses in modern biomass-to-liquids processes, as high as 70% [15]. The main impurities in syngas are tars, particulates, nitrogen and sulphur. Tar in particular is both costly to remove from the gas stream and damaging to downstream equipment surfaces that are below the dewpoint temperature of the vapour mixture. The dewpoint temperature can be as high as 450 °C [16]. Tar formation has been linked to biomass composition by multiple studies. Milne et al. [17] defined primary tars as those derived from macromolecules cellulose, hemicellulose and lignin. Others have gasified individual components to connect tar yield and tar species to carbohydrates and lignin [18,19]. Finally, researchers have shown a decrease in tar yield by comparing syngas from a torrefied and non-torrefied feedstock [20–22]. Dudyński et al. reported a reduction of tar mass from 0.0210 kg/kg_{fuel} to 0.0138 kg/kg_{fuel} in an industrial fixed-bed gasifier between untreated sawdust and torrefied pellets [22]. However, the gasification temperatures were inconsistent between tests and definitive conclusions relating tar concentration to torrefaction were not possible.

In this work, a continuous torrefaction reactor was used to torrefy short-rotation coppice willow samples at four temperatures. The resulting biomass was analyzed and gasified in a bubbling fluidized bed gasifier with steam and air under fixed and steady-state conditions. The scope presupposes a techno-economic benefit to producing torrefied SRC willow for co-firing with coal and includes value-added processing via gasification. The objective is to correlate lignocellulose composition with syngas quality using rapid analysis techniques. The resulting syngas analysis provides insight into syngas quality and tar yield based on biomass composition prior to gasification.

The novelty in this work is in application of rapid analysis techniques for process development. Analysis of syngas from fluidized bed gasification is used as feedback to determine the conditions in an upstream torrefaction process. The data and methods presented here can be used to change the willow composition during the torrefaction process in order to produce low-tar syngas from fluidized bed gasification. By changing the feedback information from tar yield and H_2/CO ratio to, for example, heating value of syngas or hydrophobicity of the willow, torrefaction parameters for other applications could be established.

2. Materials and methods

Lignocelluloses are controlled using a continuous torrefaction system and measured using a high-resolution thermogravimetric method developed by the State University of New York College of Environmental Science and Forestry (SUNY-ESF). The torrefied and non-torrefied willow is gasified in a 75 mm diameter bubbling fluidized bed with steam and air. Tar yield is quantified by comparing a gas-chromatogram to a total hydrocarbon concentration from a flame ionization detector similar to the method of Moersch et al. [23].

2.1. Biomass

Coppice willow is a promising feedstock for bioenergy applications in Northern regions. A plantation of 28 varieties of hybrid willow was established in 2007 on the University of Saskatchewan campus (Saskatoon, Canada). For this work, the shrub willow hybrid named 'SV1' (*Salix Dasyclados*) was selected. It is a well-known hybrid with disease and pest resistance and has produced high yields in a variety of soil conditions [24]. In the 2nd, three-year cycle, after 1 year of growth, the willows were harvested by hand. The stems were collected and sorted. Stems approximately 1.25 cm in diameter or less were cut to 2.5 cm lengths using a rotary cutting machine [25]. The equipment produces approximately pellet sized segments of willow that flow easily through a screw auger system. The cut biomass was then stored at 4 °C to prevent significant degradation. In storage the stems dried to approximately 10% moisture content. Cut pieces were then further dried at 150 °C in a continuous, moving bed torrefaction unit (CTU) operating as an air-dryer. Dried willow pieces retained a moisture content of approximately 5%. Moisture content between 3 and 20% was necessary to prevent adverse effects of reduced mass and energy yield and reduced energy density as a result of accelerated polymer decomposition reactions [2]. All moisture content is reported on a wet basis.

SV1 willow pieces were torrefied at four temperatures (240, 260, 270 and 280 °C) in the CTU using nitrogen gas as the heating medium rather than air. Development of the reactor is the topic of a parallel project and details will be published elsewhere. The direct heating method and moving bed allows for lower residence time relative to other torrefaction processes. Typically, torrefaction times range from 10 to 60 min whereas in this work, samples were processed for 10 min including drying, intermediate heating, heating and torrefaction as described by Bergman and Kiel [11]. Earlier work showed the effects of residence time within the range of interest (10–30 min) did not have an appreciable impact on biomass characteristics and the residence time was fixed at 10 min for these experiments [26]. The particles reached peak temperature in the last third of the reactor length. Peak particle temperature has a pronounced effect on biomass compared to the effect of residence time [2]. Torrefied and control (untreated) samples were then ground using a #1 Wiley mill operated by a 1 HP variable speed drive at 878 rpm with a 1.88 mm mesh screen.

2.2. Characterization

The relative lignocellulose composition was determined using high-resolution thermogravimetric analysis (HR-TGA). The analysis method was developed by SUNY-ESF for the purposes of breeding shrub willow (*Salix* spp.) for biomass and environmental applications [27,28]. In the early method development, Serapiglia et al. compared macro-molecular composition using well established wet chemical methods (TAPPI standard T 204 om-88 and T 222 om-88) to the high-resolution TGA method [27]. The resulting linear