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Strength, storage, and combustion characteristics of densified lignocellulosic biomass produced via torrefaction and hydrothermal carbonization

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HIGHLIGHTS

- Densification characterization of the raw and pretreated miscanthus is proposed.
- HTC pretreated miscanthus shows improved grindability and reduced ash yield.
- Energy density and O/C–H/C ratios of HTC-260 °C pellets are comparable to lignite.
- HTC pellets show improved hydrophobicity and resistance against water immersion.
- Torrefied pellets show low mass density and durability even compare to raw pellets.

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ABSTRACT

Lignocellulosic biomass has the potential to produce sustainable clean-green energy and other bio-based materials. However, due to the inferior physicochemical properties compared to coal, biomass is not regarded as an ideal feedstock for industrial applications. The work presented in this study evaluates the feasibility of two different thermal pre-treatments, torrefaction and hydrothermal carbonization (HTC), followed by densification. The densified and pretreated samples obtained from miscanthus feedstock were characterized in terms of the strength, storage, and combustion properties for energy applications. The results showed that both the thermal pre-treatments are promising methods for upgrading biomass. However, the HTC pellets showed considerably superior physicochemical properties when compared to the raw and torrefied pellets. The mass density (mass per unit volume) and volumetric energy density (HHV per unit volume) of the pellets produced via HTC at 260 °C was significantly higher (1036 kg/m^3, 26.9 GJ/m^3) compared to raw pellets (834 kg/m^3, 15.7 GJ/m^3) and torrefied pellets (820 kg/m^3, 16.7 GJ/m^3). Moreover, the HTC pellets showed improved hydrophobicity, reduction in ash content, reduction in alkali and alkaline earth metal content, and a considerable increase in the carbon content. Based on these results, the HTC pellets have potential for the heat and power applications, including replacing coal in the existing coal-fired power plants without any significant modifications.

1. Introduction

Transformation towards developing a renewable and sustainable energy resource has gained tremendous attention due to the decline in the supply and environmental concerns associate with consumption of fossil fuels. Among all the renewable energy options, lignocellulosic biomass is the only carbon neutral energy resource that can be converted into any form of fuel including solid, liquid, or gas, which has made biomass an attractive fuel source for the energy production [1]. In addition to many other advantages, the use of biomass as a supplement to fossil fuel reduces the greenhouse gases (GHGs) and other harmful-toxic carcinogenic emissions. However, the lignocellulosic have poor structural heterogeneity, non-uniform physicochemical properties, low bulk density, low carbon content, high oxygen content, low energy density, high fibrous nature, high alkali and alkaline earth metal composition, high moisture content, and hydrophilic nature. These inferior properties results in highly inefficient transportation, handling, storage, combustion, and its conversion to bio-based

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materials for bioenergy development [2]. In order to address the these limitations, lignocellulosic biomass has to be pre-processed or pre-treated before it is utilized as an efficient energy resource [3].

Compaction or densification of biomass into a regular shape product(s) like pellets, briquettes, and cubes is one way to increase the bulk density and overcome handling difficulties. Pelletizing, the densification of biomass into pellets, has obtained a great deal of worldwide interest in recent years as an efficient technique in improving the logistics of biomass. Densification of agricultural (straw and grasses) and woody (chips) biomass into pellets can increase the bulk density from 40–200 to 600–800 kg/m^3 [4]. Thus, the densification of biomass can significantly reduce the overall transportation and handling costs associated with biomass processing. However, owing to the hydrophilic nature of biomass, pellets produced from raw biomass tends to shatter when they come in contact with water or relatively high humidity conditions. The presence of high moisture content in biomass feedstock/pellets can also influence fungal growth, which can cause the material to decompose during storage.

The chemical reactions (generally oxidation) or anaerobic microorganism activity in biomass feedstock/pellets can produce heat at a sufficient rate that it can cause self-heating of the biomass stockpile. This can lead to self-ignition or other harmful toxic gaseous emissions [5]. Therefore, the pellets produced from raw biomass are not meant for long term storage (either indoor or outdoor) without the use of environmentally controlled storage structures. The construction of these structures can increase the overall storage cost associated with biomass feedstock or pellets.

Densification in combination with the thermal pre-treatments like torrefaction is often proposed as an alternate to improve the physicochemical properties of biomass [6,7]. During torrefaction, biomass is heated in an inert atmosphere at temperatures of about 200–300 °C for residence times of 30 min to a couple of hours. This process results in approximately 30% mass loss, with only 10% of the energy contained within the biomass lost in the form of gases. Therefore the specific energy density of the torrefied solid product is increased [7]. As such, the pellets produced from torrefied biomass are more cost competitive than the regular "white pellets" (raw pellets) as they have an increased bulk energy density (i.e. energy per unit volume).

Other advantages associated with the torrefaction process include reduced moisture content, improved resistance to water damage and microbial growth, and increased friability which makes torrefied pellets easier to grind [8]. While torrefied pellets represent a significant improvement over the conventional white pellets, these pellets present handling issues due to their weak strength and low durability. This can cause them to break apart easily and generate dust, which causes a risk of explosion. The bulk energy density and grindability of the torrefied pellets are not comparable to that of coal and more importantly the high inorganic metallic content in ash still remains a significant challenge for biomass combustion [9,10]. As the torrefaction process is unable to remove the alkali and alkaline earth metals from biomass ash, the use of torrefied biomass in conventional pulverized coal boiler systems is highly inefficient [11]. A research has demonstrated that torrefied biomass was only able to replace 50% of the coal used in a coal fired boiler. This was due to the lower heating value (HHV) of biomass, fouling issues and low grindability compared to coal [12].

Addition of binding agents can improve the durability of torrefied pellets. However the addition of such binders may increase the overall manufacturing cost of the pellets and may also negatively impact the combustion behavior. Pre-drying the feedstock may be required before torrefaction because the energy input and quality of end product significantly depend on the moisture content of the feedstock. The drying methods for biomass are highly energy consuming processes that require a significant financial load in the torrefaction and pelletization process, which makes the torrefaction of wet biomass like food waste impractical [13,14]. In order to improve the densification characterization of biomass without the expense of binders or adhesives, there is a need to develop an effective technique to produce pellets that have potential to replace coal at thermal power plants without any modification to the system.

A relatively new approach of hydrothermal carbonization (HTC), also referred to wet torrefaction, could potentially address these limitations of biomass. HTC is performed at the temperature range of 180–260 °C during which biomass is submerged in water and is heated in a confined system under pressure (2–6 MPa) for 5–240 min [15]. As the process itself is carried out in the presence of water it thus eliminates the pre-drying requirement of feedstock. The HTC process results in the formation of three different products: solid (hydrochar), liquid (aqueous soluble) and gaseous (mainly CO_2) products. The properties and percentage distribution of the final products strongly depends upon the process conditions [16]. Although both reaction time and temperature have been observed to influence the physicochemical characteristics of products, the reaction temperature remains the governing process parameter [17]. Hydrochar is the desired product in the HTC process, which exhibits unique and superior physicochemical properties compared to biochar (from pyrolysis and torrefaction), along with several value-added industrial applications [18].

Hydrochar is highly hydrophobic and friable, and also has the increased percentage of lignin and aqueous soluble materials compared to raw biomass. It is expected that using hydrochar for densification purposes can improve the pelletability of the biomass [19]. Secondly, since the process is carried out in the presence of liquid water, it can demineralize the elemental inorganic composition by precipitating the minerals in the liquid by-product stream. Reduction of the alkali and alkaline earth metal content from biomass would potentially mitigate the challenges such as slagging, scaling, and fouling in boilers during biomass combustion. The lack of energy intensive drying processes, high conversion efficiency, and a relatively low operating temperature and residence time range are significant advantages offered in the HTC process compared to other conventional thermal pre-treatments like torrefaction [20].

Previous research has primarily focused on woody biomass in developing sustainable energy production. However, purpose grown energy crops like miscanthus also represents a significant share in the bioenergy development, as these crops grow quickly and require less maintenance [21]. An extensive variety of literature is available on torrefaction and densification of woody and agricultural biomass. However, no study exists that has examined the comparative assessment of such crops for producing high energy dense products via HTC and torrefaction pre-treatments. The primary goal of the work presented in this study is to compare the physicochemical properties and densification characterization of raw, torrefied, and HTC pretreated miscanthus feedstock in terms of the energy density, hydrophobicity, compression strength, and durability.

2. Materials and methods

2.1. Materials

To compare and evaluate the densification performance of HTC and torrefied biomass, miscanthus (*Miscanthus* × *giganteus*, '*Nagara*') feedstock was considered in this study. The feedstock was harvested in May 2013 and collected from a privately owned farm in Drumbo, Ontario. Prior to the HTC and torrefaction experiments, the feedstock was manually chopped into samples of