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Comparative evaluation of torrefaction and hydrothermal carbonization of lignocellulosic biomass for the production of solid biofuel

Harpreet Singh Kambo, Animesh Dutta *

School of Engineering, University of Guelph, Guelph, Ontario N1G 2W1, Canada

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ABSTRACT

The work presented in this study demonstrates the potential of using hydrothermal carbonization (HTC) on miscanthus feedstock for the production of a carbon-rich solid fuel, referred to as hydrochar, whose physicochemical properties are comparable to that of coal. The effects of the processing conditions on the mass yield, energy yield and higher heating values (HHVs) were examined by varying the reaction temperature (190, 225, and 260 °C), the reaction time (5, 15, and 30 min) and the feedstock-to-water ratio (1:6 and 1:12). The results show that the reaction temperature is the most significant parameter governing the physicochemical properties of biomass. Increasing reaction temperature reduces the mass yield; however, it also significantly enhances the energy density of solid products. The hydrochar samples produced at 260 °C show a maximum energy density of 26–30 MJ/kg, with 66–74% of energy retained in the solid product. In comparison, the energy density, grindability, and hydrophobicity of the solid samples produced via torrefaction (a conventional thermal pre-treatment) were considerably lower than the hydrochar samples, even if the reaction time was kept much higher than HTC. Furthermore, the inorganic metallic composition of miscanthus feedstock almost remained unaffected after torrefaction; however, it was significantly reduced (30–70%) via HTC.

1. Introduction

The increase in global population, industrialization, and the tremendous amount of pressure on demand and consumption of energy resources are the main cause of the depletion in availability of fossil fuels, and deterioration of environmental health and ecological balance. For instance, coal is the major source of energy production for thermal power plants. Its reliability, maturity of technology, and its well understood nature by the traditional electricity producers are some of the positive strengths favoring the use of coal. However, burning of coal results in smog, soot, acid rain, global warming, and toxic air emissions, which includes the release of heavy metals, such as mercury and arsenic, that are present as trace elements in coal [1]. These counterproductive consequences intensify an interest in seeking alternative energy resources for sustainable power production [2].

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 energy production [3]. Biomass, if properly managed, offers several advantages, the most important being a renewable and sustainable energy feedstock. In addition, biomass usage can significantly mitigate net carbon emissions when compared to fossil fuels [4]. It is true that burning biomass produces pollutants including sulfur dioxide and nitrogen oxides, however the sulfur dioxide produced is 90% less than the amount produced from burning coal. In addition, the quantities of atmospheric pollution produced are insignificant compared to other pollution sources [5]. However, using biomass as a fuel source is associated with certain difficulties. It has inferior physicochemical properties in comparison to coal such as low bulk density, hydrophilic nature, low calorific value, poor grindability, and high alkali and alkaline earth metal composition the transportation, handling, storage, and combustion of biomass is highly inefficient and expensive [6,7]. Raw biomass is often moist and prone to off-gassing and self-heating due to chemical oxidation and microbial activity, therefore it has limited storage life [8]. The presence of alkali and alkaline earth metallic composition in biomass hinders its application as a solid fuel. The alkali metals react with sulfur to form alkali sulphates, which adhere on the combustor surface affecting heat transfer rates and also resulting in the corrosion of turbine blades [9]. Silica in biomass reacts to form alkali silicates that melt or soften at relatively low

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* Corresponding author at: Room No RICH 3509, School of Engineering, 50 Stone Road East, University of Guelph, Guelph, Ontario N1G 2W1, Canada. Tel.: +1 519 824 4120x52441; fax: +1 519 836 0227.

E-mail address: adutta@uoguelph.ca (A. Dutta).

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temperatures (the melting temperature can be lower than 700 °C, depending on composition) and results in the agglomeration of fluidized bed systems [10,11]. Calcium does not cause corrosion but can form deposits that are very difficult to remove from turbine blades. When using biomass for combustion, additives are often used to manage notorious behaviors of alkali metals. However, supplement addition of such additives further increases the operational cost. Moreover, lignocellulosic biomass is highly fibrous in nature and is therefore difficult to grind, where the small fine particles of biomass are generally required during gasification or its co-firing with pulverized coal [6]. Therefore, in order to overcome these aforementioned limitations, lignocellulosic biomass has to be pre-processed or pre-treated before it is utilized as an efficient energy resource.

Recent research interests in the carbonization of biomass and using the final solid product as a substitute to coal has gained remarkable attention [6,12]. Torrefaction is a conventional thermal pre-treatment and is often proposed as an alternative to improve the physicochemical properties of biomass [12–16]. 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. The physicochemical properties of biomass such as grindability, hydrophobicity, and HHV of torrefied biomass (here referred to as biochar) are improved to some extent, however, the high inorganic metallic content in ash still remains a significant challenge for biomass combustion [17–19]. In addition, the torrefaction process is highly inefficient when dealing with wet biomass i.e. feedstocks with high moisture content [20,21].

A relatively new approach of hydrothermal carbonization (HTC) also referred to wet torrefaction, shows promising results. 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 [22]. The process itself is carried out in the presence of water and therefore is not affected by the high moisture content of feedstock [23,24]. 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 depend upon the process operating conditions [25–28]. Hydrochar is the desired product in the HTC process, which in certain ways exhibits unique and superior physicochemical properties compared to biochar (from pyrolysis and torrefaction), along with several value-added industrial applications [22,29]. However, thermal energy is typically required for the post-treatment of HTC streams, i.e. separation of the solid and liquid products, but due to loss in cell structure of biomass this energy requirement can be significantly lower compared to conventional drying methods used for raw biomass [28]. The lack of energy intensive drying processes and high conversion efficiency are few significant advantages offered in the HTC processing of biomass compared to other conventional thermal pre-treatments like torrefaction [22,30–32].

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 bioenergy development, as these crops grow quickly and require less maintenance [33]. There is a wealth of scientific literature available on the torrefaction and HTC of woody and agricultural biomass [14,22,34–37]. However, very few studies have examined the comparative assessment of purpose grown crops for producing energy dense fuel via HTC and torrefaction pre-treatments. In addition, the composition of alkali and alkaline earth metals present in the raw biomass has considerable impact on the combustion behavior of the pre-treated biomass. The HTC process is carried out in the presence of water, and therefore can wash away (fully or partially) these inorganic elemental compositions from biomass into liquid by-product stream, which would reduce the overall ash content. However, to date, very few studies have investigated the comparison of the fate of inorganic compositions in hydrochar and biochar [18,38]. Further experimental analysis is required to validate the results. The primary goal of the work presented in this study is to evaluate the performance of HTC of miscanthus feedstock by: (1) investigating the effect of reaction time and temperature on mass and energy yield of hydrochar; (2) studying the effect of process operating conditions on the structural and chemical properties of hydrochar; (3) examining the composition of alkali and alkaline earth metals prior to and after HTC pre-treatment; (4) comparing the results with the biochar produced via torrefaction.

2. Material and methods

2.1. Materials

To compare and evaluate the performance of HTC and torrefaction, miscanthus (*Miscanthus* × *giganteus*, '*Nagara*') feedstock collected from a privately owned farm in Drumbo, Ontario was considered in this study. The initial moisture content of raw miscanthus feedstock was 10.95%. Prior to the treatment, the feedstock was manually chopped into samples of lengths ranging from 20 to 25 mm for uniformity. These prepared samples were then stored in a sealed plastic bag until treatment. To compare raw and pre-treated (HTC and torrefied) miscanthus samples to conventional fossil fuel, a coal sample from another study was used as a reference material [39].

2.2. Experimental setup

2.2.1. Hydrothermal carbonization (HTC)

HTC of biomass was carried out in a 600 ml Parr bench top reactor (Moline, IL) fitted with the glass liner (762HC3). To examine the effect of different operating conditions on biomass, the experiments were conducted at different reaction temperatures (190, 225, and 260 °C), residence times (5, 15, and 30 min). The temperature of the reactor system was controlled using a Proportional–Integral–Derivative (PID) temperature controller. Prior to the HTC reaction, 10 g of miscanthus feedstock was mixed with deionized water. To study the effect of solid load, for each run the mixture of a water to feedstock ratio of either 6 or 12 to 1 (on mass basis) was loaded into the reaction vessel. The initial moisture content of raw miscanthus feedstock was 10.95%; the supplement amount of water for HTC experiments was added accordingly. was The reactor pressure was not controlled but indicated by the pressure gauge attached to it, which ranged from 1 to 5 MPa. The reactor was heated up to the desired temperature in 20–30 min was then maintained for required time period. Later, to further quench the reactions, the reactor was immersed in cold water and was cooled down to room temperature in about 5–7 min. Once the inside temperature of the reactor dropped to room temperature, the pressure release valve was opened under the fume hood and the gaseous products were allowed to escape. The solid and liquid samples were separated from each other by using a filter paper with a pore size of 20 μm. The separated solid samples were then dried overnight (minimum 16 h) in an oven at 103 °C. It should be noticed that the heating-up time for the reactor to the desired temperature and the cool down period could have significant effect on the properties of product streams. However, to keep the process straightforward and for data analysis purposes the reaction time and temperature for isothermal holding time period. The full detail on the experimental methods and schematic diagram of HTC process can be found in the previous work of this study [40]. The HTC experiments were repeated three times to ensure the reproducibility and consistency.