Oxidative torrefaction of spruce-pine-fir sawdust in a slot-rectangular spouted bed reactor

Ziliang Wang[a,*], Hui Li[a,b], C. Jim Lim[a], John R. Grace[a]

[a] Department of Chemical and Biological Engineering, University of British Columbia, Vancouver V6T 1Z3, Canada
[b] Institute of Biological and Environmental Engineering, Hunan Academy of Forestry, Changsha 410004, PR China

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ABSTRACT

A slot-rectangular spouted bed (SRSB) reactor was employed to torrefy spruce-pine-fir sawdust in an oxidative atmosphere. The effects of torrefaction temperature (240–300 °C) and feed-gas oxygen concentration (3–9 vol.%) on torrefaction performance and solid product properties were investigated. Adding oxygen in the feed-gas led to more uniform temperature distributions in the SRSB reactor, and was a promising way to achieve stable continuous operation. The mass fraction of cyclone-caught torrefied biomass increased, whereas the mass fraction of torrefied biomass remaining in the reactor decreased, with increasing oxygen concentration and temperature. In oxidative torrefaction, biomass weight loss was 15.4–46.4%, while energy yield was 61.2–99.4%. The effects of oxygen concentration on the weight loss, energy yield and torrefied biomass properties were more significant at 300 °C than at 240 °C. The torrefied sawdust also had greater HHV and contained less volatiles, more carbon, less oxygen and less hydrogen than the raw biomass.

1. Introduction

Biomass, a nearly carbon-neutral fuel, is broadly available in nature. Utilization of biomass can reduce fossil fuels consumption and environmental pollution, especially greenhouse gas [1,2]. However, biomass has lower calorific value, higher moisture content and lower energy density than fossil fuels, thereby reducing the potential of biomass as an alternative to fossil fuels [1,3].

Biomass properties can be upgraded by torrefaction [4–8]. In particular, torrefaction leads to higher heating values or energy densities, reduced atomic O/C and H/C ratios, lower moisture content, higher water-resistivity, improved grindability and reactivity, and more uniform properties [5,9]. Moreover, torrefaction as a promising pretreatment method can positively influence subsequent thermochemical processes of biomass, such as pyrolysis [10,11], gasification [12] and combustion [13]. However, torrefaction is usually carried out in nitrogen [9,14], in the absence of oxygen, requiring gas separation to obtain the nitrogen, thereby increasing the operating cost [5]. A more attractive and practical alternative is to recycle flue gas from a combustor [15,16], as a means of reducing the cost of biomass torrefaction. Biomass is then heated at modest temperatures of 200–300 °C in the oxygen-containing environment. The oxygen concentration in the flue gas generally varies from 6 to 14 vol.% [16]. The main reactions of oxidative torrefaction include not only devolatilization and thermal

decomposition of biomass for removal of oxygen, but also exothermic oxidation reactions, which provide the heat needed for thermal degradation of the biomass, thereby reducing the heat demand. Moreover, oxidation reactions are generally faster than devolatilization and decomposition reactions, leading to shortening of the torrefaction duration [5,17]. In addition, to achieve the same weight loss, a lower torrefaction temperature is required for oxidative torrefaction than for non-oxidative torrefaction [17,18].

Oxidative torrefaction has been well investigated in laboratory studies. Uemura et al. [19] reported that oxidative torrefaction takes place in two successive steps, the first being the same as for oxygen-free torrefaction, and the other involving biomass oxidation. Biomass oxidation is dominated by surface oxidation. There exists an upper limit for the air superficial velocity, beyond which thermal degradation of biomass is governed by internal mass transport, rather than by surface oxidation [20]. The influence of oxygen concentration on the solid yield and the torrefied product properties is found to be more marked at higher temperatures [21]. Although, adding oxygen can lead to more uniform heating of the bed, there are temperature-specific limits, beyond which an increase in oxygen concentration likely results in thermal runaway in the packed or fixed beds [22]. An increase in torrefaction temperature leads to reduced combustive limits for oxygen concentration. When combustion flue gas is used to torrefy the biomass, the grindability and hydroscopicity of the solid torrefied products are

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enhanced [15]. The calorific value increases and the solid yield decreases with increasing torrefaction temperature [16].

Spouted beds were originally invented for contacting coarse particles with gases [23], e.g. for drying of wheat. They have excellent gas-solids heat and mass transfer, and have recently found applications in energy conversion [24–28]. Grace and Lim [29] reported that slot-rectangular spouted bed reactors are relatively easy to scale up by extending the column thickness, compared with conventional conical-cylindrical spouted beds. They are capable of handling particles larger than 1 mm in diameter, such as typical biomass particles [30]. Slot-rectangular spouted beds can potentially serve as torrefaction reactors [29,31], but they have not been previously utilized to torrefy woody biomass, except for our previous work [32,33]. These two previous studies focused on the effects of particle size (0.25–0.5 mm, 0.5–1.0 mm and 1.0–2.0 mm), biomass feed rate (220–710 g/h) and temperature (240–330 °C) on SRSB reactor performance and torrefied product properties. The results indicated that particle size plays an important role in determining the residence time and torrefaction performance, especially energy yield and weight loss. It was also found that the torrefaction performance of 0.5–1.0 mm sawdust is better than for 0.25–0.5 mm and 1.0–2.0 mm sawdust. Moreover, the authors suggested that, to quickly achieve a stably continuous operation with stable torrefied biomass entrainment, either lower biomass feed rate or higher torrefaction temperature should be employed, or use of an oxygen-containing gas as the carrier gas, leading to faster weight loss of biomass.

In the present study, the feasibility and reliability of oxidative torrefaction in a continuous slot-rectangular spouted bed facility were assessed. The effects of temperature and feed-gas oxygen concentration on the torrefaction performance and product properties were investigated. The torrefied product properties, including proximate analysis, fibre analysis, elemental analysis, and higher heating value (HHV) measurement were evaluated, while the hydrodynamics of the SRSB reactor were also assessed. At the same time, oxidative and non-oxidative torrefaction of biomass were compared in the slot-rectangular spouted bed reactor.

2. Experimental set-up and material

2.1. Experimental apparatus and operation

The slot-rectangular spouted bed torrefaction facility is shown schematically in Fig. 1. Detailed information on the SRSB reactor can be found in our previous work [32].

At the beginning of each run, 3000 g of glass beads of 1.0 mm diameter were loaded into the reactor. Electrically heated air was utilized to preheat the SRSB reactor to the required temperature. Next, nitrogen replaced air to purge the system for 5–10 min. The nitrogen or a mixture of nitrogen and air was then fed to the SRSB reactor at a pre-determined flowrate and oxygen concentration. The oxygen concentration in the feed-gas was controlled by mixing air and nitrogen at different flowrates, with the two gas flows controlled individually, aided by two rotameters. The total gas flowrate was measured by an orifice flowmeter at elevated torrefaction temperatures. A flue gas analyzer (PS-200, HORIBA) was connected to a gas sampling port (see Fig. 1) to ascertain the oxygen concentration prior to biomass feeding.

After the system had stabilized for 5–10 min, the screw-feeder was turned on to feed sawdust into the reactor. The screw-feeder had been calibrated at room temperature with the SPF sawdust before being employed in the experiments, and the biomass feed rate was calculated as the actual weight of biomass fed divided by the duration of feeding. To prevent hot gas from entering the screw-feeder, 4 L/min nitrogen was fed into it for the entire experimental period. Sawdust was fed into the reactor, contacting hot glass beads as heat carriers, and N_2 or

N_2 + air. There was no immediate discharge of torrefied material. Instead, the sawdust particles lost weight as they were decomposed, devolatilized and oxidized, with some then pneumatically carried over from the reactor. Most entrained particles were captured by a cyclone. The gas then passed to a stainless steel mesh filter of 50 μm apertures to remove the remaining particles. Finally, the off-gas entered an after-burner after being cooled to room temperature.

During the torrefaction experiments, torrefied product samples of ~2 g were taken from a solid sampling port just below the bottom of the cyclone (see Fig. 1) at 5–10 min intervals. To prevent tar from condensing in the cyclone, the cyclone was heated to the same temperature as the spouted bed reactor by a heating tape. The filter was so close to the cyclone that no condensation was observed inside it.

After the SRSB reactor cooled to less than 50 °C, the mixed torrefied sawdust and glass beads were extracted from the reactor by a vacuum cleaner. The glass beads were then recycled after being separated from the torrefied sawdust. Torrefied solid samples from the cyclone and reactor were sealed in zip bags, labelled and stored in a cold storage room (at < 4 °C) for future analysis.

2.2. Material characterization

The experimental biomass particles were spruce-pine-fir (SPF) sawdust from Tolko Industries Ltd., Vernon, BC, Canada. This sawdust was dried, milled and sieved in a Gilson Test-Master Sieving device for 10 min. The 0.5–1.0 mm size fraction was then selected for the experiments, because of three size fractions studied, this fraction performed best in the earlier work [32]. The 0.5–1.0 mm particles account for the highest fraction (37.5 wt.%) of the original raw sawdust. Table 1 gives key properties of experimental material. The inert particles were glass beads of 1.0 mm Sauter mean diameter and 2530 kg/m^3 density.

The loose-packed-bed bulk density of the raw sawdust was measured by half-filling a 500 mL graduated cylinder with a weighed sawdust sample, inverting the covered cylinder and then quickly re-inverting it to measure the sample volume. Moisture content was measured by weighing a sample of raw sawdust before and after holding it in an oven at 105 °C for 24 h. Proximate analyses of the raw and torrefied sawdust were determined by a TGA (Shimadzu, TA-60WS) and a programmable furnace. The detailed operating procedure was provided in our previous study [32]. The volatile matter contents of the solid samples were measured by the TGA. The ash content was determined based on the NREL/TP-510-42622 method [34]. The fixed carbon content was subsequently determined by difference. Fibres were analyzed following Van Soest with a Fibretherm FT12, Gerhardt. Elemental analyses of the raw and torrefied sawdust were provided by a Carlo Erba EA 1108 elemental analyzer, giving carbon, hydrogen and nitrogen elemental contents of the sample in wt.% on a dry basis. The oxygen content was estimated by difference. The higher heating value (HHV) was measured by a bomb calorimeter (Parr 6100), with sawdust pelletized prior to the measurement to ensure controlled combustion. The HHV was determined at least three times for each sample. The relative uncertainties for all measurements were less than 3% for replicates. Note the elemental and fibre analyses for raw and torrefied biomass were repeated, but not for all torrefied products. We randomly selected one of the torrefied products for repetition, and found the repeated results to be very close, with relative error less than 3%.

2.3. Operating conditions

Each experiment took 4–6 h, including a preheating stage of 3–5 h and the oxidative torrefaction period of 50 min. In addition, the reactor would take 30–90 min to cool down from the pre-determined reaction temperature to 200 °C. Three torrefaction temperatures, 240, 270 and 300 °C, were selected based on our previous work for non-oxidative