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Research paper

Pilot scale steam-oxygen CFB gasification of commercial torrefied wood pellets. The effect of torrefaction on the gasification performance

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1. Introduction

Biomass is considered as a potentially carbon neutral energy source. However, due to its price, moisture content, heterogeneous composition and cost of logistics, it is not yet ideal for many thermal conversion applications. Therefore, efforts are being made to develop upgrading processes which convert biomass into a fuel with superior properties in terms of logistics and end-use.

Torrefaction is a thermochemical process, carried out in an oxygen-deficient atmosphere at typically 230–300 °C. During torrefaction the biomass becomes more coal alike; its energy density increases (on mass basis), it becomes more hydrophobic, more brittle and its O/C and H/C molar ratios decrease. Furthermore, if torrefaction is combined with a densification step, the energy density increases on a volumetric basis and its logistics and handling operations are improved [1]. In addition, life cycle assessment studies have shown that torrefied wood offers environmental benefits in global warming impact when it is used for energy applications, such as co-firing with coal for electricity generation [2] and transportation fuels production [3].

Various types of gasification exist based on the applied reactor type. Fluidized bed gasification is a technology which shows benefits in feedstock flexibility and scale-up opportunities. In their...
handbook on gasification, Roracher et al. [4] describe that there are various operational fluidized bed gasification plants globally; such as large scale coal and biomass plants with capacities up to the order of magnitude of 100 MWth output. The gasifier product gas is fired in lime kilns or dedicated boilers, or it is co-fired with coal for power generation or CHP. The characteristics of the fluidized bed gasification of biomass have been studied extensively using smaller scale facilities. In these experimental studies, the focus was put mainly on the cold gas efficiency (CGE), the carbon conversion efficiency (CCE), the permanent gas composition and the tar content [5,6].

So far, only limited studies [7–11] have investigated the effect of torrefaction on permanent gas composition and tar content during fluidized bed gasification of biomass. Furthermore, these studies were restricted to bubbling fluidized bed gasification and the feedstocks used were torrefied on a small scale by the researchers themselves, except for the study by Kulkarni et al. [10] who acquired their feedstock from the American company, New Biomass Energy, LLC. In general, these authors concluded that torrefaction did not have a positive influence on gasification performance, with respect to CCE and CGE. In addition, they reported a limited effect on permanent gas composition and a reduction of the total tar content. Among these studies, only Kwapiszka et al. reported deviating results regarding the effect of torrefaction on the H2 content and on the total tar content. Berreco et al. [8] performed lab-scale steam-oxygen gasification of Norwegian spruce and forest residues at 850 °C. They reported that increasing the torrefaction temperature from 225 to 275 °C resulted in a marginal increase of the H2 and CO contents and a decrease of the total tar content, up to 85% and 66% for forest residues and spruce, respectively. Furthermore, they presented that due to torrefaction the char and gas yields increased; whereas, the CGE did not show a clear trend. Sweeney [7] performed steam gasification of wood at 788 °C but without mentioning the conditions of torrefaction. The author reported the same effects of increasing torrefaction severity as Berreco et al. with respect to the H2 content and tar content. On the other hand, Sweeney reported a reduction in both CCE and CGE due to torrefaction. Woytiuk et al. [11] performed steam-air gasification at 900 °C of willow and torrefied willow at four different temperatures. These authors reported that increasing torrefaction temperature resulted in an increase of the H2 content and a decrease of the tar content by 47%, when the torrefaction temperature reached or exceeded 260 °C. In contrast with studies mentioned above, the CO content remained unaffected. Kulkarni et al. [10] performed air-blown gasification of pine wood at 935 °C. These authors do not report the torrefaction conditions; they concluded that torrefaction led to a decrease in CEG and to minor changes in product gas constituents’ compositions, the H2 content increased and the CO content decreased. Lastly, Kwapiszka et al. [9] performed air-blown gasification of miscanthus × giganteus (M × G) at 850 °C. However, due to the fact that the miscanthus is not a woody type of biomass, their findings are not included in this study.

As presented above, there has been limited and, in several aspects, contradictory research on the effect of torrefaction on the permanent gas composition, CCE, CGE and tar content during fluidized bed gasification of biomass. Furthermore, so far only one publication [10] has considered commercially produced torrefied wood and no studies have evaluated the effect of heavily torrefied conditions (torrefaction at 300 °C) in wood gasification. No research has been carried out, to our best knowledge, on the impact of torrefaction on the steam-oxygen circulating fluidized bed gasification of wood. Thus, the goal of this study is to investigate the influence of torrefaction on permanent gas composition, tar content, CCE and CGE during steam-oxygen circulating fluidized bed gasification of commercial torrefied wood.

2. Materials and methods

2.1. Experimental test rig geometry and analytical methods

The experimental facility at TU Delft consisted of a 100 kWth circulating fluidized bed gasifier (CFBG) followed by a woven ceramic four-candle filter unit (i.e. BWF) operating at 450 °C, and equipped with a gas supply system, a solids supply system and analytical equipment. A schematic of the experimental rig is presented in Fig. 1. Detailed information on the experimental rig has been described elsewhere [12]. Gas and tar were sampled at different locations in the rig. The gas was sampled from the G.A. point downstream the riser and analyzed on-line using a Varian μ-GC CP-4900 equipped with two modules, which measured the volumetric concentration of CO, H2, CH4, CO2 and N2 (1 m CO2 column) and benzene, toluene and xylene, also coded as BTX (4 m CP-Sil5 CB column). The gas composition data from the μ-GC are obtained in intervals of 3 min. In addition, an NDIR analyzer (Hartmann & Braun Ursas 10P) monitors CO2 and CO and a paramagnetic analyzer measures the oxygen concentration (Hartmann & Braun Magnos 6G) with a time interval of 2 s. The water content in the product gas was analyzed via sampling a measured flowrate of product gas for a determined timeframe. The gas was cooled in a condenser immersed in a mixture of ice, water and salt. The weight of the condenser was measured at the beginning and at the end of the test. The tar content of the product gas was sampled from the T.P. point downstream the BWF filter according to the tar standard [13] method. The tar samples were analyzed using an HPLC equipped with a UV and fluorescence detector (Knauer), and a reverse phase column (Kromasil Eternity C18 5 μm 150 × 4.6 mm). 20 μL of filtered sample were injected in the column and a gradient elution with methanol—water was performed for 50 min. The UV detector was set at 254 nm. The quantification was performed by external calibration using triplicate data point and, using standard tar compounds in an appropriate concentration range. All coefficients of determination (R2) exceeded 0.990.

2.2. Biomass feedstock

Four samples of biomass feedstock were tested, two commercial torrefied woods and their parent materials; all samples were in pellet form. Two Dutch companies supplied the fuels, TorrCoal International B.V. and Topell Energy B.V. Topell torrefied pellets (coded as Topell black) consisted of forestry residues torrefied at 250 °C for a less than 5 min with the Torbed® technology, which utilizes a heat carrying medium, blown at high velocities through the bed bottom to acquire a high heat transfer. The Topell black pellets had an outer diameter of 8 mm and a length of approximately 2 cm, and untreated Topell pellets (made from the same residues and coded as Topell white) had an outer diameter of 6 mm and a length of approximately 2 cm. The Torrcoal torrefied pellets (coded as Torrcoal black) consisted of mixed wood, i.e. coniferous and deciduous wood, and residues from Dutch, Belgian and German forests, which were torrefied at 300 °C for less than 10 min in a rotary drum reactor. Both Torrcoal black pellets and untreated Torrcoal wood pellets (coded as Torrcoal white) had an outer diameter of 6 mm and a length of approximately 2 cm. The elemental analysis, proximate analysis and torrefaction degree of the samples are presented in Table 1. The latter was calculated based on the anhydrous weight loss or the reduction of the volatile content upon torrefaction divided by the initial volatile content on an a dry basis. The elemental composition of all feedstocks has been analyzed at the University of L’Aquila, Italy, with a PerkinElmer Series 2 CHNS/O 2400 analyzer. The proximate analysis was
performed via thermogravimetric analysis at the Technical University of Delft. For this purpose a Thermal Advantage SDT Q600 thermogravimetric analyzer (TGA) was used. Detailed information on the TGA procedure has been described elsewhere [14]. Based on the elemental analysis data of the feedstock samples and based on the data for various fuels obtained from the Phyllis2 online database [15], a Van Krevelen diagram (Fig. 2) was drawn that shows the changes in the woody feedstocks due to torrefaction. It is confirmed that torrefaction decreased the O/C and H/C ratios for both wood feedstocks and, even though Topell white and Torrcoal white have approximately spot in the diagram, the higher torrefaction temperature for the Torrcoal black feedstock resulted in lowering both ratios more than for the Topell black feedstock.

2.3. Bed material

Calcined magnesite was used as the bed material in this paper. Calcined magnesite is a mineral consisting mainly of MgO and smaller fractions of Fe₂O₃, CaO, and silica. Detailed information regarding the constituents, price and particle size distribution of the bed material can be found in a previous study from our group [12].

2.4. Gasification parameters

The gasification experiments were performed at approximately 805–852 °C and atmospheric pressure. The experiments were carried out varying the equivalence ratio (ER) and the steam to

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Table 1
Biomass feedstock ultimate and proximate analysis.

| Biomass          | Ultimate analysis, wt% | Proximate analysis, wt% | LHV c,d   | Torrefaction degree f |
|------------------|------------------------|-------------------------|-----------|-----------------------|
|                  | C² | H² | N² | S² | O³   | Moisture⁴ | Volatile matter⁵ | Fixed carbon⁶ | Ash⁷ |         |
| Topell white     | 45.6 | 5.6 | 0.2 | 0.7 | 39.4 | 6.5      | 79.3      | 19.2 | 1.5 | 16.9 | –     |
| Topell black     | 47.6 | 5.4 | 0.3 | 0.7 | 36   | 7.5      | 72.3      | 25.4 | 2.3 | 17.3 | 8.8   |
| Torrcoal white   | 46.6 | 5.8 | 0.2 | 0.8 | 39.7 | 5.9      | 76.8      | 21.8 | 1.4 | 17.3 | –     |
| Torrcoal black   | 53.5 | 5.2 | 0.5 | 0.7 | 34.0 | 4.1      | 66.2      | 32.2 | 1.6 | 19   | 13.8  |

⁴ On a.r. basis.
⁵ O content is calculated by difference.
⁶ Calculated based on [16].
⁷ MJ/kg⁻¹.
⁸ On dry basis.
⁹ %, the torrefaction degree was calculated based on the anhydrous weight loss or the reduction of the volatile content upon torrefaction divided by the initial volatile content on a dry basis.

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3. Results and discussion

All the results presented in this paper are measured during representative steady state time frames. A typical dry gas composition over time graph during steady state operation of the gasifier is presented in Fig. 3.

The permanent gas (vol%) and the tar species concentrations (g Nm⁻³) are presented on a dry and nitrogen-free (dnf) basis. The CO, CO₂, H₂, CH₄, and BTX contents presented are the average values during the steady state operation. Moreover, the standard deviations of these gas species are presented. For water, no standard deviation value is presented due to the nature of the measurement method used. As described above, during the steady state only one measurement for quantification of the water content was performed. The tar yield (g kg⁻¹ daf) is presented on a dry ash-free (daf) basis of supplied feedstock. Finally, key performance indicators based on mass balance calculations are reported.

3.1. Feedstock characterization

The four samples were characterized concerning their slow devolatilization behavior in a N₂-atmosphere. The changes in mass loss rate versus temperature curves, as presented in Fig. 4, are generally reported to be due to changes in chemical composition during torrefaction. For both torrefied feedstocks, the “shoulder” on the left side of the peak has disappeared, which is generally attributed to the (partial) conversion of the hemicellulose fraction in lignocellulosic biomass feedstock [17]. As a consequence, both torrefied feedstocks are expected to contain higher lignin and cellulose contents than their parent materials. Demirbas [18] reported that a higher lignin content results in a higher fixed carbon content, which was found for both feedstocks in this study as well (see Table 1).

3.2. Effect of torrefaction on gasification

Torrefaction had an impact on the product gas composition for Topell and Torrcoal feedstocks, as shown in Figs. 5 and 6. Torrefaction resulted in a decrease in CO₂ (approximately 4% dnf), an increase of CO (approximately 3% dnf), a minimal increase of H₂ (approximately 2.3% dnf) and a minimal decrease of CH₄. The change of each permanent gas species cannot be discussed in isolation from the others due to the chemical reactions taking place in the gasifier simultaneously. The decrease of the CO₂ is attributed to the torrefaction conditions, as the CO₂ is the gas that is released in larger amounts at low temperatures due to hemicellulose devolatilization [17]. On the other hand, main sources for the release of CO are cellulose and lignin, as reported by Wu et al. [19]. In addition, as torrefaction results in lowering the volatile content and the H content of the fuel, the slight increase in the H₂ content in Topell black and Torrcoal black experiments was not expected. This increase can be attributed to steam reforming reactions; due to the higher fixed carbon content of the torrefied material more char is available to react with steam under our process conditions. Lastly, the water content of the product gas is presented in the graphs. The water content in the product gas during Torrcoal black and Topell black experiments was lower than the parent materials. As the water measurement is not considered the most accurate, the

| Test | Biomass     | Date       | Fuel flow rate | ER  | SBR   | Temperature | Pressure | Steady state |
|------|-------------|------------|----------------|-----|-------|-------------|----------|--------------|
| 1    | Topell white| 19-02-15   | 12.0           | 0.31| 1.00  | 845         | 1.2      | 126          |
| 2    | Topell black| 28-05-15   | 12.0           | 0.30| 1.00  | 840         | 1.3      | 98           |
| 3    | Topell black| 10-12-14   | 12.0           | 0.20| 1.30  | 805         | 1.2      | 70           |
| 4    | Topell black| 29-05-15   | 12.0           | 0.36| 0.85  | 842         | 1.2      | 200          |
| 5    | Torrcoal white| 13-07-15  | 12.4           | 0.36| 0.85  | 843         | 1.1      | 120          |
| 6    | Torrcoal black| 10-07-15  | 12.1           | 0.36| 0.85  | 852         | 1.1      | 180          |

*a The SBR is calculated on an “as received” basis.
modified SBR* value was calculated, which consists of the total water (steam and biomass moisture) ratio to dry biomass input, to investigate whether the different moisture contents of untreated and torrefied material influence this observation. It is found that the SBR* is the same among the Topell feedstocks and slightly different between the Torrcoal feedstocks, 0.98 and 0.95 for Torrcoal white and Torrcoal black, respectively.

Both feedstocks’ results are mostly in agreement with literature. Several authors gasified wood that was torrefied at conditions relevant to Topell black [7,8,10,11]. However, even though the effect of torrefaction on the H2 and CH4 contents is the same, contradictions exist for the CO and CO2 contents. These differences for the CO
and CO₂ behaviors exist due to the different gasification conditions. For example, Berrueco et al. [8] who performed experiments with the most relevant conditions compared to this study (at 850 °C, 1 bar, with oxygen and steam), reported the same effect like us in CO, H₂ and CH₄ contents, but not for the CO₂ content. This reduction in the CO₂ content in our study may be due to a higher activity of the Boudouard reaction with torrefied feedstocks because of the higher availability of carbon in the torrefied feedstock. In addition, the lower volatile matter content of the torrefied biomass (7% and 10% less for Topell black and Torrcoal black, respectively) is expected to result in a lower primary tars formation. The latter would permit a lower steam demand for reforming of the hydrocarbons and, thus, a higher steam availability for the water-gas-shift (WGS) and char gasification reactions.

The variability in the ER and SBR values in the Topell black experiments resulted in changes in the H₂ and CO₂ contents, as expected. Increasing the SBR and decreasing the ER resulted in increasing the H₂ content in the product gas. On the other hand, the CO content remained the same. The latter may be attributed to the WGS reaction which worked as a stabilizing factor, if SBR increased and ER decreased, part of produced CO may react with the extra steam to produce H₂ and CO₂. In addition, Topell black (Tests 3) and Torrcoal black (Test 5) have been gasified using the same ER and SBR values. The limited differences in product gas composition are attributed to differences in wood origins and in torrefaction conditions (more severe for Torrcoal black than for Topell black).

Based on the μ-GC analysis of the product gas, torrefaction generally resulted in a reduced BTX content (see Figs. 7 and 8). According to Yu et al. [20], who studied tar formation of all three individual biomass components, i.e. cellulose, hemicellulose and lignin, BTX originates primarily from hemicellulose and cellulose, and secondly from lignin. As torrefaction leads to a decrease in the hemicellulose content, a reduction in the BTX was to be expected. Moreover, the reduction is larger for Torrcoal black which is
torrefied at higher temperature than Topell black, indicating a larger decrease in the hemicellulose content for the Torrcoal black. The most affected BTX species is the benzene for both feedstocks.

Torrefaction resulted in a reduction in the total tar content in the product gas for both feedstocks (Fig. 9). For each tar compound, Torrcoal white resulted in higher concentrations than Topell white, although under different gasification conditions (ER and SBR). Torrefaction resulted in a larger reduction of the total tar content for Torrcoal black. Moreover, all the tar compounds concentrations decreased in the Torrcoal black experiments, while for the Topell black all the tar compounds lighter than ethylbenzene decreased. This reduction in the total tar content during fluidized bed gasification due to torrefaction has been reported before in literature [7,8,10]. As torrefaction decreases the volatile matter content of the feedstock, a lower amount of primary tars is released in the devolatilization step in the gasifier. As a consequence, a lower amount of secondary and tertiary tars may be expected as well. Therefore, a more severe torrefaction, as in case of Torrcoal black, will lead to a larger reduction in volatile matter content and, therefore, in less tar formation.

Since the total tar content was affected by torrefaction, the individual tar classes were affected as well (see Fig. 10). For Topell black, Class 3 and Class 4 tars decreased by 37% (from 2.66 to 1.67 g.Nm⁻³) and 26% (from 2.0 to 1.5 g.Nm⁻³), respectively. Class 5 tars shows a slight, but not significant increase, from 0.14 to 0.18 g.Nm⁻³. The total tar concentration reductions and the total tar yield reduction were approximately 30% and 40%, respectively. Class 3 tars were decreased mainly due to a decrease in toluene. The decrease in Classes 3, 4 and 5 was much larger for Torrcoal black; it was approximately 50%, 61% and 82%, respectively. Class 3 tars decreased 3.0 to 1.5 g.Nm⁻³, Class 4 tars decreased from 3.2 to 1.2 g.Nm⁻³ and Class 5 tars decreased from 0.5 to 0.1 g.Nm⁻³. This large reduction in the total tar content and total tar yield derived from the reduction of toluene and naphthalene, which decreased more than 40%. Lastly, Class 2 tars (phenol in Fig. 9) totally converted. This decrease in phenol content was not expected as Torrcoal black is expected to contain more lignin than Torrcoal white. However, it can be explained, as it is reported before that the presence of H₂ in the product gas enhanced significantly the hydrodeoxygenation of the oxygenated tar compounds [21].

The simultaneous increase in ER and decrease in SBR resulted for Topell black in no significant changes in total tar concentration.
and yield (Figs. 9 and 10). However, small changes did occur in almost all the individual tar compounds. The combined increase in ER and decrease in SBR resulted in converting the phenol. The latter is among the reasons why, the relative fraction of Class 3 tars increased, whereas, the Class 4 tars relative fraction decreased.

Based on mass balance calculations, various process key performance indicators were calculated, such as CCE, CGE, molar ratio of H2/CO, gas yield, etc. (Table 3). For the Torrcoal samples, torrefaction resulted in a decrease in CCE, while the opposite was observed for the Topell samples. While the former was expected as a result of the lower volatile matter content [7,8,10], the latter was not. As described before [12] the feeding system consisted of a screw feeder which also grinds the biomass pellets during operation. By disconnecting the feeder from the gasifier and collecting and analysing the material downstream the feeding screw, it was found that the average particle size of Topell black was significantly smaller than the average particle size of Topell white (Fig. 11). Apparently, this was due a more severe grinding caused by the larger diameter of the Topell black pellets in combination with the increased brittleness resulting from the torrefaction. Siedlecki and de Jong [22] have reported that a smaller particle size will lead to a higher burnout rate (i.e. a higher CCE) and tar yield, which it did. Related to the increased CCE and CGE, the LHV of the gas increased as well. In addition, it was also checked if the deviating result for the Topell samples could be explained by the sub-optimal recirculation conditions or the high absolute pressure in the riser in the Topell white experiments. Therefore, the differential pressures measurements of the reactor were checked, but this was not the case.

For the Torrcoal samples, the particle size distribution after the feeder was not determined. Because of the more severe torrefaction conditions leading to an even further increased...
brittleness, one might expect even smaller particle sizes for Torrcoal black than for Topell black. However, due to the smaller pellet size, the grinding effect of the feeder was probably much smaller. For the Torrcoal samples, torrefaction led to a decrease in CCE, but the CGE remained the same. The latter can be attributed to the increase of the H₂ and CO contents. Finally, for both feedstocks torrefaction resulted in an increase in the gas yield as reported before [7,8,10].

4. Conclusions

Torrefaction, when combined with a densification step, offers benefits in logistics and handling operations. Therefore, in this study, steam-oxygen blown circulating fluidized bed gasification experiments at 850 °C have been performed with commercial torrefied woods and their parent materials in order to investigate the impact of torrefaction under our conditions. The examined operational conditions were relevant to typical operating conditions in practical applications.

It is concluded that torrefaction affected the gasification performance of both woody feedstocks the same way with respect to the permanent gas composition, gas yield and total tar content, but in different ways regarding the CCE and CGE. Torrefaction resulted in an increased gas quality, as it yielded higher H₂ and CO contents, a decrease of the CO₂ content, and a significant decrease of the total tar content. For Topell black, the decrease in the tar content concerned Class 3 and 4 tars, whereas, for Torrcoal black this decrease was larger and it concerned all tar classes. Moreover, in both cases torrefaction resulted in an increased gas yield in the gasifier. For the Torrcoal samples, torrefaction resulted in a decrease in CCE as expected based on the decrease in volatile matter content. The CCE remained approximately constant due to an increase in H₂ and CO content in the product gas. The Topell samples showed an increase in CCE and CGE upon torrefaction, which could be attributed to a significant grinding in the screw feeder. In addition to the benefits of torrefaction in logistics and handling, it is generally concluded that both torrefied fuels may offer benefits as a feedstock for steam-oxygen blown circulating fluidized bed gasification, in particular in terms of gas quality and yield.
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