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Improvement of gasification performance of *Eucalyptus globulus* stumps with torrefaction and densification pre-treatments

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HIGHLIGHTS

- Gasification of eucalypt stumps.
- Study of the biomass pre-treatments: densification, torrefaction and densification of torrefied feedstock.
- Effect of biomass pre-treatments on syngas yield and quality.
- Effect of biomass pre-treatments on the release of H_2S and NH_3.
- Selection of the best biomass pre-treatments to improve gasification performance and syngas composition.

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ABSTRACT

Eucalyptus globulus stumps has been used as biofuel for energy production. However, forest biomass presents some disadvantages (high moisture, volatiles and oxygen contents, hydrophilic nature, low calorific value, bulk density and energy density). To overcome this, torrefaction and pelletisation were tested. Based on the assays, 250 °C and 30 min were selected as the best conditions for the torrefaction. The summative chemical analysis has shown that torrefied stumps presented higher total extractives and lignin content and lower monosaccharides in relation to raw stumps. Eucalyptus stumps were also densified to produce pellets of raw and torrefied material. The materials obtained by the different pre-treatments studied were gasified in a fluidised bed reactor. Gasification gas obtained with raw stumps was compared to those obtained with pre-treated stumps by pelletisation (or densification), torrefaction and torrefaction followed by pelletisation. The results obtained showed that bio-syngas was improved when torrefied biomass was used, as there was an increase in gas yield and CGE (Cold Gas Efficiency) and lower tar were produced. The best pre-treatment for eucalyptus stumps was densification after torrefaction, as at 850 °C, gas yield was about 1.22 Nl/g daf and CGE was around 65%. This syngas contained around 38% of H_2, 27% of both CO and CO_2 and 5% of CH_4.

1. Introduction

Energy from biomass is one of the important pathways towards increasing the share of renewable energy. However, biomass has some disadvantages as a solid fuel: high moisture, volatiles and oxygen contents, hydrophilic nature, low calorific value, bulk density and energy density. Transportation costs are generally high, which affect the economic viability of the process [1].

To partially overcome these problems, biomass may be pre-treated *e.g.* by torrefaction. Torrefaction is a slow heating process at temperatures from 200 °C to 300 °C, usually in an inert atmosphere, by which biomass loses moisture and decreases the O/C ratio, thus increasing its energy content. Torrefied biomass is a uniform solid retaining around 70% of the initial mass and about 90% of the initial energy with increased hydrophobicity and microbial degradation resistance, and better grindability [2–10]. Therefore, torrefaction allows reduction in transport and storage costs and improvement in conversion efficiency [10].

Biomass torrefaction may be classified as [4]: *i*) light torrefaction (up to 230 °C), where light volatiles and moisture are removed and the heating value increases slightly; *ii*) mild torrefaction (at about 260 °C) where most moisture and volatiles are removed and cellulose is present in much higher content than lignin; and

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iii) severe torrefaction (up to 290 °C) at which an important fraction of cellulose is destroyed, and the heating value is mostly due to the lignin fraction. Temperature and degree of torrefaction are key parameters to balance the improvements in biomass fuel quality and the mass and energy losses [2].

Gasification of torrefied wood is more efficient than that of raw wood [10]. Efficiency is improved when torrefaction is made before entrained flow gasification [11], and carbon conversion is also enhanced when using torrefied wood [12]. For instance, the Cold Gas Efficiency (CGE) of sawdust torrefied at 250 °C increased in relation to that of raw sawdust [4] and syngas produced using torrefied pellets had a higher calorific value [13]. The gasification of torrefied bamboo also improved significantly and CGE increased around 88%, producing a syngas richer in H_2 and CO [14]. Kuo et al. [8] compared gasification performances of raw bamboo and of torrefied bamboo at 250 °C and 300 °C and concluded that 250 °C torrefied bamboos were the best for gasification, considering syngas yield, carbon conversion and CGE. The use of torrefied wood in relation to gasification of raw bamboo increased syngas production, the total power generation and the energy conversion efficiency [15].

Some authors analysed the integration of torrefaction with gasification [16–18] and reported that gasification of torrefied biomass was a good option, as total energy efficiency was improved. Chen et al. [18] stated that syngas quality and CGE improved when torrefied biomass was used instead of the raw material.

Torrefied biomass is easier to densify, because more fatty structures are developed during torrefaction that may act as binder [19,5]. When pellets are produced with torrefied biomass, the bulk density increases. The formation of pellets with torrefied biomass leads to less fines, higher uniformity, energy and bulk densities which improve the energetic conversion efficiency and reduce transportation and storage costs. Therefore, pellets using torrefied biomass are a better option to improve heating value, grindability, combustion performance, storage, transport and handling [11]. Sarkar et al. [19] also stated that the combination of torrefaction and densification was the best option for gasification at 900 °C, producing a syngas with a high lower heating value and high yields of H_2 and CO.

In this work the valorisation of biomass from *Eucalyptus globulus* stumps as a solid fuel was studied. *Eucalyptus globulus* is used intensively by the pulp and paper industry in the Iberian Peninsula; after 2 or 3 harvesting cycles the tree stumps are removed from the field and considered low value biomass wastes. An energetic valorisation of the stumps may be achieved by gasification, as this process is a technological proven concept for heat and electricity production. Besides, the solids obtained after gasification are rich in minerals and may be used for soil fertilization.

The main goal of the present work is to study the effect of the biomass pre-treatments of densification, torrefaction and densification of torrefied feedstock on gasification performance and gas yield and quality. The eucalypt stumps were converted into bio-syngas, whose yields, composition and energy properties were evaluated. The gasification results obtained with the different pre-treatments of *Eucalyptus globulus* stumps were compared with those obtained with the raw feedstock. This is the main innovation of the work presented. Another innovative aspect is related to gasification gas analysis to determine the effect of different pre-treatments on gasification gas composition, mainly tar, H_2S and NH_3 contents.

2. Experimental part

2.1. Material

Eucalyptus stumps were collected in the yard of one pulp mill (The Navigator Company) that uses them as feedstock for energy production. The stumps biomass was dried and ground to obtain particles with sizes from 2 to 4 mm. The pre-treatments that were applied were: *i*) densification through the production of pellets, ii) torrefaction and iii) densification of the torrefied feedstock.

The stumps biomass was characterized by elemental and proximate analysis according to international standards: moisture (CEN/TS 14774-2), ash (CEN/TS 14775), total carbon, hydrogen and nitrogen (CEN/TS 15104), sulphur (CEN/TS 15289) and calorific values (CEN/TS 14918). Table 1 shows the ultimate and proximate analyses of raw and torrefied eucalypt stumps as described in Pinto et al. [20].

Table 1
Ultimate and proximate analysis of raw and torrefied *Eucalyptus globulus* stumps.

Elemental analysis (% daf[a])	Raw stumps	Torrefied stumps at 250 °C
C	49.9	51.1
H	5.8	5.0
N	0.2	0.3
S	0.03	n.d.
Proximate analysis (% as received)		
Ash	1.5	1.9
Moisture	14.1	1.1
Volatile Matter	68.2	76.6
Fixed Carbon	16.2	20.4
HHV (MJ/kg daf[a])	22.3	22.7

n.d. – not detected.
[a] = (daf - dry and ash free basis).

2.2. Chemical composition and Py-GC/MS profile

The chemical analysis was performed according to Technical Association of the Pulp and Paper Industry (TAPPI) standard methods; ash (T 211 om-02), extractives (modified from T 12 os-75), lignin (Klason lignin according to T 222 om-02 and acid soluble lignin UM T 250) in 40–60 mesh granulometric fraction. The methodology and equipment used are detailed in [21].

The lignin was characterized by analytical pyrolysis. The extractive-free samples were powdered in a Retsch MM200 mixer ball and around 0.20 mg were pyrolysed in a 5150 CDS apparatus linked to an Agilent GC 7890B with a mass detector system 5977B. The capillary column used was a ZB-1701: 60 m × 0.25 mm *i.d.* × 0.25 μm film thickness. The chromatograph oven program was: 40 °C (held for 4 min), then increased to 70 °C at a rate of 10 °C min^{-1}, then 100 °C at a rate of 5 °C min^{-1}, 265 °C at 3 °C min^{-1} (held for 3 min), and increased to 270 °C at a rate of 5 °C min^{-1} (held for 9 min). The compounds were identified comparing their mass spectra with Wiley, NIST2014 and by mass fragmentation. The identified compounds results were presented as percentage of total area. The percentage of guaiacyl (G) and syringyl (S) derived products were separately summed and the S/G ratio calculated. The percentage of carbohydrates was also summed and the relation between carbohydrates and lignin was calculated and presented as C/L ratio.

2.3. Densification

Pellets obtained after densification of eucalypt stumps and of torrefied stumps had a diameter of 0.5 cm and average length of 2 cm. A Tenchini s.n.c. FT300 pelletiser was used. The stumps feedstock was compressed against the metallic shape of the pelletiser, without the addition of any additive. The pellets obtained for both the initial eucalypt stumps and the torrefied material, showed a good and stable shape.