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Biomass Gasification in Entrained Flow Reactor: Influence of Wood Particle Size

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The influence of beech-wood particle size on its gasification with O_2 was investigated in Entrained Flow Reactor (EFR) conditions. Experiments were performed in two facilities at very different scales: a drop tube reactor (DTR), and a pilot-scale EFR, the biomass feeding rates being set at 1 g/min and 90 kg/h respectively. Numerical simulation was used to bring a better understanding of the process.

In the DTR, the carbon conversion into gas sharply increased with temperature for the larger particle powder – D_{50}/D_{95} = 1230/1570 µm – from about 20% at 800 °C to 80 % at 1400 °C. For the smaller particle powder - D_{50}/D_{95} = 370/510 µm – the conversion was of 80 % or higher whatever the temperature. At 1400 °C, the same conversion of about 80 % was reached for both powders and the gas composition was quite similar. The difference of conversion between the two powders was attributed to heat transfer limitations in the solid, and to a shorter residence time of the larger particles in the reactor.

In the pilot-scale EFR, the different powder sizes – the smaller one being similar to the one used in the DTR, and up to D_{95} of 1040 µm - led to very close results in terms of temperature reached into the reactor, carbon conversion into gas and gas composition. For equivalent ratios (O_2/O_2 for stoichiometric combustion) of 0.36 and 0.45, the carbon conversion into gas was close to 100 %.

1. Introduction

Entrained Flow Reactor (EFR) is one of the most promising gasification technologies for biofuel production. Indeed, a high temperature (about 1500°C) and a high heat flux on the biomass particles ($>10^6$ $W.m^{-2}$) allow producing a syngas (H_2 – CO) almost free of tar and of gaseous hydrocarbons with a high conversion of biomass into gas. However, one of the drawbacks of the EFR fed with solids is the need to feed pulverized biomass to reach a good conversion into gas. The milling process is energy costly and feeding particles as large as possible is thus desirable.

The drop tube reactor (DTR) is a laboratory-scale equipment well adapted to study biomass gasification in conditions representative of EFR gasification. However, only a few studies performed at a temperature higher than 1000 °C can be found in literature. Moreover, the influence of biomass particle size on its pyrolysis or gasification in a DTR has only been scarcely studied, and most of the time under 1000 °C (Chen et al., 2013; Septien et al., 2012; Wei et al., 2006; Zanzi et al., 2002).

Our aim was to study the influence of beech-wood particle size on its gasification with O_2 in EFR conditions. Experiments were performed in two facilities at very different scales: a DTR, and a pilot-scale EFR, the biomass feeding rates being set at 1 g/min and 90 kg/h respectively. In parallel with the experiments, numerical simulation was performed to bring a better understanding of the process.

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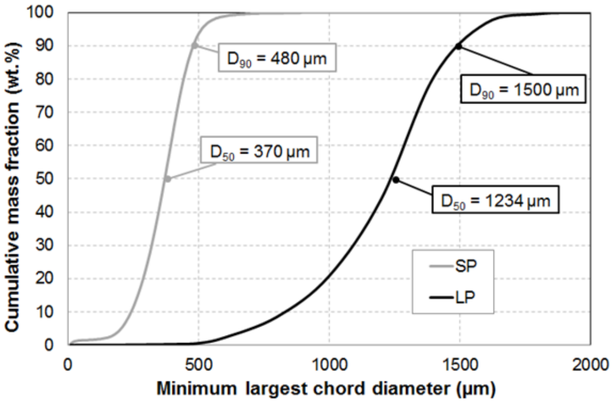

Figure 1: Particle size distribution of SP and LP beech powders

Table 1: Proximate and ultimate analyses of the SP and LP batches (^{b}: calculated by difference)

Proximate analysis			**Ultimate analysis (wt% dry ash free basis)**		
	SP	LP		SP	LP
Moisture (wt%)	8.79	9.2	**C**	49.1	49.2
Volatile matter (wt% dry basis)	84.3	84.6	**H**	5.7	5.7
Fixed carbon^b (wt% dry basis)	15.2	14.9	**N**	< 0.3	< 0.3
Ash (wt% dry basis)	0.5	0.5	**S**	0.014	0.015
			O^b	44.5	44.4

2. Investigation at laboratory scale

2.1 Description of the Drop Tube Reactor

The drop tube reactor (DTR) used in the present study was previously described in details by Billaud et al. (2014, 2016). It consists of an alumina tube inserted in a vertical electrical heater with three independent heating zones. The dimensions of the tube are 2.3 m in length and 0.075 m in internal diameter. The heated zone is 1.2 m long. The DTR works at atmospheric pressure and can reach a maximum temperature of 1400 °C. The wood particles are continuously fed into the reactor using a gravimetric feeding system. The main gas stream, which is a mixture of N_2 and air, is electrically pre-heated before entering the reactor. A sampling probe allows collecting the gas produced and the remaining solid.

Gasification products pass through a settling box where the remaining char is sampled. Soot is trapped downstream in a filter. A micro-gas chromatograph (µ-GC) allows analysing the main gaseous products (CO_2, CO, CH_4, N_2, H_2, C_2H_2, C_2H_4, C_2H_6, C_3H_8, C_6H_6). The H_2O content in the gas was measured by a psychrometer. The gas yields were calculated using N_2 as tracer gas.

2.2 Conditions of the experiments

Two batches of beech wood particles with different distribution sizes were used. The small particles (SP) and large particles (LP) batches were obtained by sieving between 315 and 450 µm and 1120 and 1250 µm respectively. The minimum largest chord diameter distribution of these two batches was measured by a "Camsizer" tool and is given in Figure 1. This quantity is close to the width for a wood particle with parallelepiped shape. There are no significant differences in the proximate and ultimate analyses of the two powders (Table 1).

The biomass feeding rate was set at 1 g.min^{-1} for all experiments. The temperature of the DTR – 800, 1000, 1200 and 1400 °C - was varied from one experiment to another. The total gas feeding rate was adjusted – 18.8, 15.9, 13.7, 12.1 NL.min^{-1} respectively at 800, 1000, 1200 and 1400°C - so that the gas residence time in the reactor was 4.3 s for all experiments. The air inlet flowrate was set to reach an equivalent ratio ER (O_2/O_2 for stoichiometric combustion) of 0.44. The O_2 content in the inlet gas was comprised between 2.0 and 3.1 %.

2.3 Modelling

The experiments were simulated using a model especially developed to represent biomass gasification in a DTR at temperatures up to 1400 °C. This model was presented in previous publications (Septien et al., 2013; Billaud et al., 2016). Briefly, the DTR is modelled as a plug flow reactor, in which gas and solid residence times are both calculated. Evolutions of the solid physical properties, going from wood to char, are taken into account. Biomass thermochemical conversion is decomposed into several phenomena: drying, pyrolysis modelled by a single global reaction associated to a kinetic law, char gasification by H_2O and CO_2 with adapted kinetics, gas phase reactions modelled by a detailed kinetic scheme (Ranzi et al., 2005) and soot formation. In the version of the model adapted to the smaller particles (Billaud et al., 2016), the temperature and concentrations were supposed to be uniform inside the solid particles. However, going towards larger particles of millimetre-size, heat transfer inside the particles can be limiting in the global conversion process, as shown with a characteristic time analysis (Septien, 2011). Thus the previous model was modified, so as to take into account internal heat and mass transfers in the wood particles. These particles are then represented by meshed spheres. In each mesh, the temperature, pressure, concentrations and density are uniform. Mass and energy balances are considered for each mesh. The results of this new version are presented in the next section.

2.4 Results and discussion for laboratory scale study

Carbon conversion into gas, considering the following gas species: CO, CO_2, CH_4, C_2H_2, C_2H_4, C_2H_6, C_3H_8 and C_6H_6, is represented as a function of temperature in Figure 2, for the SP and LP powders. For the SP powder, this conversion slightly decreases as temperature increases, which cannot be completely explained. The model shows a different trend with increasing conversion with temperature, linked to a better char gasification. According to the model, 1 to 7% of the carbon is in tar and soot depending on the temperature. However, the experiment and model results globally give similar carbon conversion levels which always remain higher than about 80%. For the LP powder, the carbon conversion into gas sharply increases with temperature. The experimental results clearly show that the carbon conversion into gas is lower for the SP than for the LP at temperatures of 800, 1000 and 1200 °C. Only at 1400 °C, the conversion is similar for the two powders.

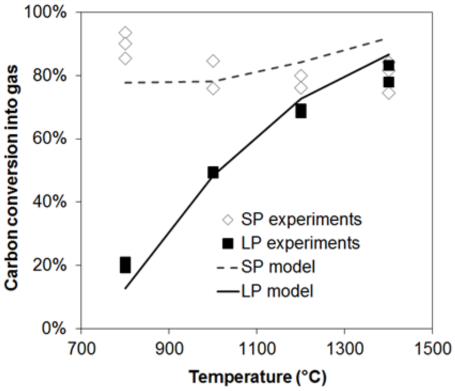

Figure 2: Carbon conversion into gas for SP and LP powders as a function of temperature in the DTR (experiments and model)

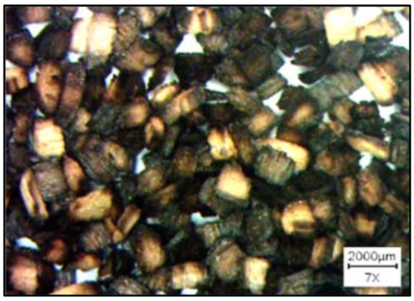

Figure 3: Picture of char recovered at the outlet of the DTR after LP gasification at 800° C

Observations of the chars recovered in the settling box show that, at least after the tests performed at 800 °C and 1000 °C, the solid particles are not completely pyrolysed but only partly brownish (Figure 3).

The simulation of the carbon conversion into gas is very satisfactory for the LP (Figure 2). Moreover, as shown in Figure 4 for the main gaseous species, the individual gas yields are also very well represented by the model in nearly all conditions, for the SP and LP powders. This was already shown and discussed in a previous publication (Billaud et al., 2016) for the SP powder. This previous study showed that the modelled phenomena (section 2.3) all together allowed well reproducing the experiments with SP, for which heat transfer limitations can be neglected. According to the model, O_2 reacts with pyrolysis gas species, and especially hydrocarbons, as soon as they are released. It was not necessary to include direct oxidation of solids (char and soot) by O_2 to well represent experimental figures. Water-gas shift reaction has a major influence on gas composition which is near to thermodynamic equilibrium from 1200 °C. Figure 4 shows that the H_2, CO and CH_4 yields are quite different for the two powders except at 1400 °C. On the contrary, the CO_2 yield is very close for LP and SP whatever the temperature. This is probably linked to CO_2 formation from combustion reactions, whose extent is linked to the O_2 quantity, similar in all cases.

The particle residence time calculated with the model is shown in Figure 5 as a function of the axial location in the DTR for SP and LP at two temperatures. Whatever the temperature, the calculated residence time is much lower for LP than for SP powder. This difference is due to the higher density of LP compared to SP, especially as long as they are not pyrolysed, which induces a higher falling velocity. Moreover, internal heat transfer limitations in the bigger particles induce a slower apparent pyrolysis velocity. Thus, considering heat transfer limitation in the particles and variations in the solid falling velocity, in combination with chemical transformation already validated for the SP (Billaud et al., 2016), allows explaining and reproducing the difference of conversion between the two powder batches within the new version of the model.

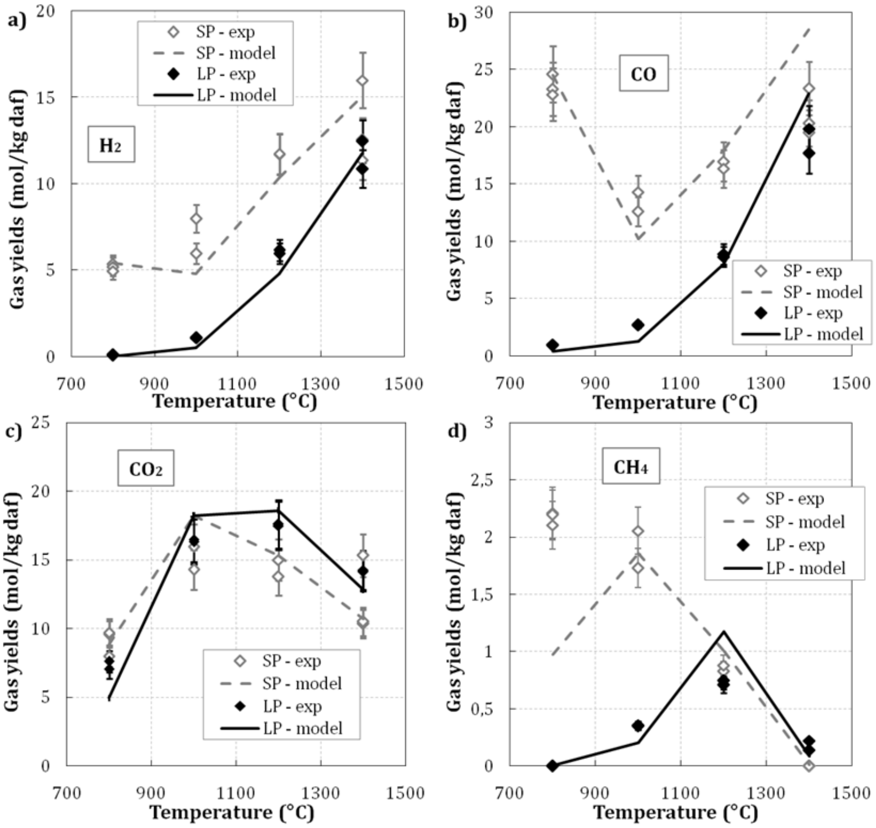

Figure 4: Main gaseous species yields for SP and LP powders as a function of temperature in the DTR (experiments and model)

3. Investigation at pilot scale

3.1 Description of the Pressurized Entrained Flow Biomass Gasifier (PEBG)

The PEBG is located at the SP Energy Technology Center (SP ETC) in Piteå (Sweden). It is an autothermal reactor designed to operate in slagging mode with process temperatures ranging between 1200 and 1500 °C for a maximum power of 1 MWth at pressures up to 10 bars. The gasifier consists of a ceramic lined reactor of 0.52 m in inner diameter and 1.67 m in vertical reactor wall length. Five thermocouples allow measuring the process temperatures at different locations inside the reactor. A gas stream is continuously sampled and sent to a micro-gas chromatograph (μGC) for online analysis of H_2, He, O_2, N_2, CO, CH_4, CO_2, C_2H_2, C_2H_4 and C_2H_6. Moreover, gas is periodically sampled with sampling bags to analyze benzene with a GC-FID. A stream of He is injected in the reactor (4 NL/min) and the product gas yields are calculated using it as a tracer. The carbon conversion into gas is calculated from the the CO, CH_4, CO_2, C_2H_2, C_2H_4, C_2H_6 and C_6H_6 yields. A more detailed description of this reactor is given by Weiland et al. (2013).

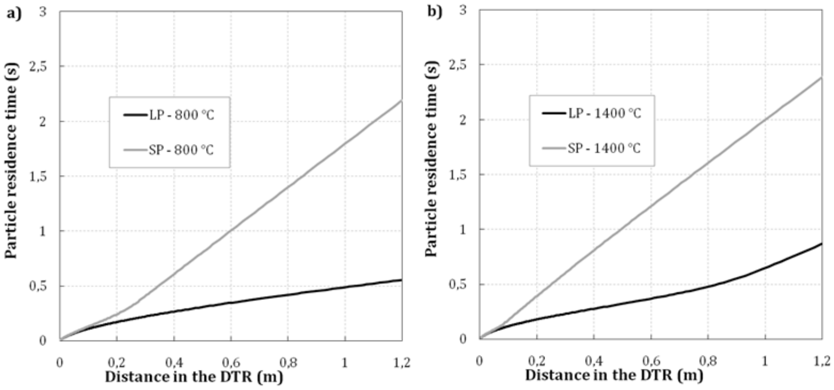

Figure 5: Solid particle residence time as a function of the axial location in the DTR at 800 °C (a) and 1400 °C (b) – model results

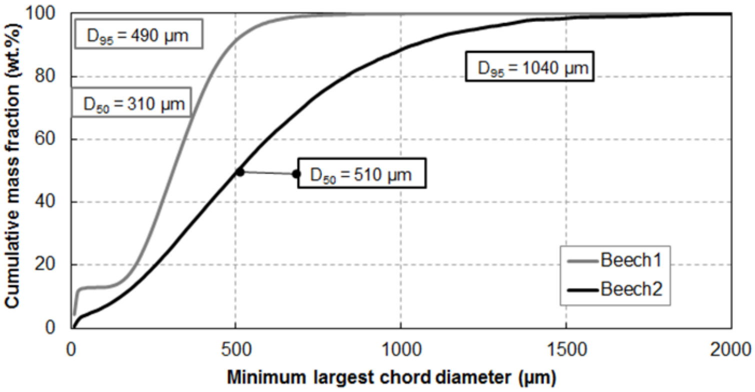

Figure 6: Particle size distribution of Beech1 and Beech2 powders

3.2 Conditions of the experiments

Two types of beech powder, with different particle size distributions, were used. They are referred to as Beech1 and Beech2. The proximate and ultimate analyses are similar to the ones of the beech powder used for the DTR study (Table 1). The minimum largest chord diameter distribution of these two batches is given in Figure 6. The Beech1 powder has a narrow size distribution with D_{50} = 310 µm and D_{90} = 490 µm. This batch is thus very similar to the SP one used in the DTR. The Beech2 powder has a wider size distribution with D_{50} = 510 µm and D_{90} = 1040 µm. The Beech2 particles look more heterogeneous than the Beech1 ones with parallelepiped particles but also needles. According to these size distributions we can estimate that about 40% in mass of Beech2 particles has a larger width than Beech1 particles.

Experiments were performed with each of the powder batches, varying the inlet O_2 flowrate and thus the equivalent ratio: 0.27 - 0.36 - 0.45. All the experiments were conducted with a biomass feeding rate of 90 kg/h (~415 kW) and a pressure of 2 bars. The plug flow gas residence time in the gasifier varied between 3.5 and 5.8 s, for an equivalent ratio going from 0.45 to 0.27.

3.3 Results and discussion for pilot scale study

Temperature measured at the middle of the reactor is represented in Figure 7 as a function of ER for each powder batch. As expected, temperature increases with ER, from 1050 °C for ER = 0.27 to 1300 °C for ER = 0.45. The type of wood powder has no influence on the temperature for the same ER value. Carbon conversion into gas increases from about 90% for ER = 0.27 to nearly 100% for ER = 0.36 and ER = 0.45. These results are consistent with previous ones (Weiland et al., 2015). Once again, the beech powder particle size has no significant influence on the carbon conversion into gas for a given equivalent ratio. It can be noted that, for a temperature of about 1180 °C (ER = 0.36), the conversion of biomass into gas is already almost total, even for the largest wood particles. The higher conversion of carbon into gas in the PEBG than in the DTR could be partly due to higher concentrations of oxidant gaseous species – O_2, H_2O, CO_2 – in the PEBG, which favours char gasification reactions, as well as hydrocarbon reforming reactions.

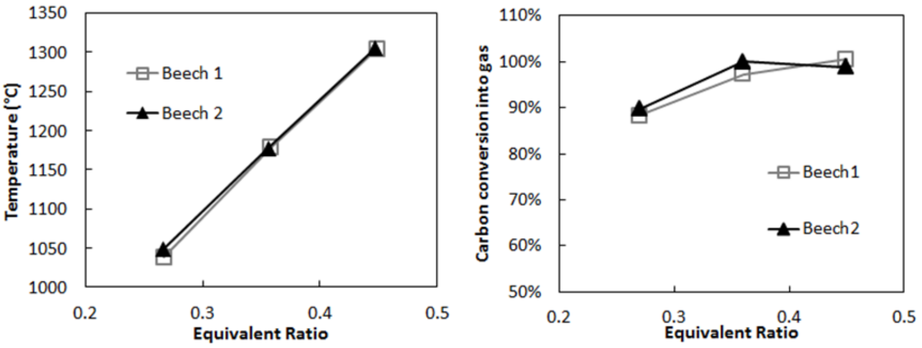

Figure 7: Temperature measured in the PEBG a) and carbon conversion into gas b) as a function of ER

4. Conclusion

The influence of beech-wood particle size on its gasification with O_2 was investigated in a DTR and a pilot-scale EFR. In the 1.2 m long DTR, the carbon conversion into gas sharply increased with temperature for the larger particle powder – D_{50}/D_{95} = 1230/1570 µm – from about 20% at 800 °C to 80 % at 1400 °C. For the smaller particle powder - D_{50}/D_{95} = 370/510 µm – the conversion was 80% or higher whatever the temperature. At 1400 °C, the same conversion of about 80 % was reached for both powders and the gas composition was quite similar. The difference of conversion between the two powders was attributed to heat transfer limitations in the solid, and to a shorter residence time of the larger particles in the reactor. By considering these phenomena in a model representing biomass gasification in the DTR, the simulation results of carbon conversion into gas and gaseous product yields were in very good agreement with the experimental ones.

In the pilot-scale EFR, the different powder sizes – up to D_{95} of 1040 µm - led to very close results in terms of temperature reached into the reactor, solid conversion into gas and gas composition. For equivalent ratios of 0.36 and 0.45, the carbon conversion into gas was close to 100 %. These results show that wood particles of about 1 mm in size could be well gasified in an EFR. The upper size limit should still be sought.

Acknowledgments

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