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Author post-print (accepted) deposited by Coventry University's Repository

Original citation & hyperlink:

Sher, F, Pans, MA, Sun, C, Snape, C & Liu, H 2018, 'Oxy-fuel combustion study of biomass fuels in a 20 kWth fluidized bed combustor' Fuel, vol. 215, pp. 778-786. https://dx.doi.org/10.1016/j.fuel.2017.11.039

DOI 10.1016/j.fuel.2017.11.039 ISSN 0016-2361 ESSN 1873-7153

Publisher: Elsevier

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Oxy-fuel combustion study of biomass fuels in a 20 kW_{th} fluidized bed combustor

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6 Abstract

7 Oxy-fuel combustion is one of the promising carbon capture technologies considered to be suitable for 8 future commercial applications with stationary combustion plants. Although more and more biomass 9 and waste are now being burned in stationary combustion plants, research on oxy-fuel combustion of 10 biomass has received much less attention in comparison to oxy-fuel combustion of coal. In this work, a 11 series of tests was carried out in a 20 kW_{th} fluidized bed combustor under oxy-fuel conditions firing 12 two non-woody fuels (miscanthus and straw pellets) and one woody fuel (domestic wood pellet). The 13 effects of the combustion atmosphere (air and oxy-fuel) and oxygen concentration in the oxidant of the 14 oxy-fuel combustion on gas emissions and temperature profiles were systematically studied with the 15 overall excess oxygen coefficient/oxygen level in the reactor being maintained roughly constant 16 throughout the tests. The experimental results showed that simply replacing the air with an oxy-fuel 17 oxidant of 21% O₂ and 79% CO₂ resulted in a significant decrease in combustion temperature and 18 ultimately led to the extinction of the biomass flame due to the larger specific heat of CO₂ compared to 19 N_2 . To keep a similar temperature profile to that achieved under the air combustion conditions, the 20 oxygen concentration in the oxidant of O_2/CO_2 mixture had to be increased to 30 vol.%. A drastic 21 decrease in CO emissions was observed for all the biomass fuels (up to 80% reduction when firing 22 straw) under oxy-fuel combustion conditions providing that the oxygen concentration in the oxidant of 23 O₂/CO₂ mixture was above 25 vol.%. NOx emissions were found to decrease with the oxygen 24 concentration in the oxy-fuel oxidant, due to i) the increase of bed temperature, which implies more 25 volatile-N released and converted in the highly-reducing dense bed zone and ii) the less dilution of the 26 gases inside the dense bed zone, which leads to a higher CO concentration in this region enhancing the

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27 reduction of NOx. Similar NOx emissions to those obtained with air combustion were found when the 28 oxygen concentration in the oxy-fuel oxidant was kept at 30%. Further analysis of the experimental 29 results showed that the gas emissions when firing the non-woody fuels were controlled mainly by the 30 freeboard temperature instead of the dense bed region temperature due to the characteristic high 31 contents of volatile matter and fines of this kind of biomass fuels.

32

Key words: Biomass combustion; Fluidized bed combustion; Oxy-fuel combustion; Carbon capture;
NOx and CO emissions

35

36 1. Introduction

37 Growing concerns on the greenhouse gas emissions and their potential impact on climate change 38 demand not only the application of CO_2 capture and storage (CCS) technologies to large point 39 anthropogenic CO₂ emission sources such as coal and natural gas fired power plants but also the 40 implementation of CO₂ negative combustion technologies such as Bio-energy with Carbon Capture and 41 Storage (BECCS) within the next decades. Although removing CO_2 from the atmosphere, i.e. direct 42 capture of CO_2 from air, may be necessary in the longer term, direct CO_2 capture is more technically 43 challenging and more expensive than CCS and BECCS applied to large scale combustion plants[1-3]. 44 Biomass is considered as a renewable fuel, a carbon-neutral energy source and hence its combustion 45 integrated with CCS can lead to negative CO₂ emissions. Biomass has already captured worldwide 46 attention in the context of greenhouse gas control even though fossil fuels are expected to retain their 47 dominant role in the world energy supply in the coming decades[4].

48

Oxy-fuel combustion is one of the most developed CCS technologies and considered as technically feasible and economically competitive for future commercial applications [5-7]. Oxy-fuel combustion refers to fuel being burned in a mixture of oxygen and recycled flue gas (RFG). Unlike conventional air combustion plants that use air as the oxidant, an oxy-fired plant employs an Air Separation Unit (ASU) to produce an almost pure oxygen stream. The oxygen stream is then combined with RFG to produce an oxygen enriched gas as the oxidant. The flue gas recycle is necessary to moderate the otherwise excessively high flame temperature that would result from fuel combustion in pure oxygen. After the removal of water and other impurities from the flue gas exhaust stream, high-purity CO_2 (up to 95%) is produced and almost ready for sequestration[8, 9]. As mentioned above, the combination of oxy-fuel combustion with biomass could effectively provide a method which would not only avoid further CO_2 emissions but also helps reduce the atmospheric CO_2 . Furthermore, the oxy-fuel process also offers other advantages such as improving the ignition and burnout performance.

61

62 Among all the available combustion technologies, fluidized bed combustion (FBC) is often considered 63 as the best choice for the combustion and/or co-combustion of biomass, waste and other low quality 64 solid fuels due to its fuel flexibility, long residence times, and uniform combustion temperatures. The 65 characteristics of FBC also offers several advantages for its application in oxy-fuel systems [10]. Firstly, 66 the difficulty of flue gas recirculation for temperature control in pulverized fuel (PF) applications could 67 be reduced in circulating fluidized bed (CFB) by means of the bed material recirculation, since the 68 specific heat of the solids is much higher than that of the recycled flue gas. Secondly, lower NOx 69 emissions and better sulphur removal are possible. Finally, it is easier to retrofit an FB boiler from air 70 to oxy-fuel combustion as there will be no need for a new burner.

71

72 The effects of oxy-fuel atmosphere and O₂ concentration in the oxy-fuel oxidant gas on pollutant 73 emissions (NOx and CO) in fluidized bed systems firing different kinds of coal have been thoroughly 74 investigated during the past years by a number of researchers. In general, the experimental results 75 showed that NOx emissions in oxy-fuel combustion with low O₂ concentrations are lower than those 76 obtained under air-firing atmosphere, because of the lower temperatures as well as higher char and CO 77 concentrations in the dense bed [10, 11]. Furthermore, NOx emissions were found to increase with the 78 increasing O₂ concentrations in the oxy-fuel oxidant, which is as a result of i) the increase of the 79 temperature in the furnace which elevates the concentrations of O and OH radicals and enhances NO 80 formation and ii) the lower gas velocity in the riser and longer residence time of fuel particles in the 81 combustor, which may promote the fuel-N conversion into NOx precursors [10-14]. In some works an opposite trend was found, i.e. an decrease of NOx with the increasing O₂ concentration in the oxy-fuel oxidant, as in the study of de las Obras-Loscertales et al. [15]. The authors explained this trend by means of the different operational procedure used, comparing with other works: in their work [15], an increase in the oxygen concentration was compensated with an increase of the coal flow rate fed to the reactor, keeping constant the total gas flow rate and excess oxygen coefficient in all tests. As a result, more unconverted char was present in the bed, favouring the NO reduction on the char surface.

88

89 Regarding CO emissions, Duan et al. [16] observed a much lower CO emission in air than those in an oxy-fuel atmosphere with the same O₂ concentration when firing two kind of coal in a 50 kW_{th} CFB 90 91 facility, due to the higher temperature achieved under the air combustion conditions. They also reported 92 a decrease in CO emissions when the O_2 concentration in the oxy-fuel oxidant gas increased, as the 93 oxidation of carbon was more complete and therefore less CO was formed. On the other hand, Hofbauer 94 et al. [14] observed similar CO emissions for air combustion and two oxy-fuel cases investigated (with 95 26 vol.% and 36 vol. % of O_2 in the oxidant, respectively), firing bituminous coal in a 150 kW_{th} CFB 96 reactor. Jia et al. [17] performed a series of oxy-fuel tests with flue gas recycle in a 100 kWth CFB 97 combustor firing bituminous coal. The CO emissions of oxy-fuel combustion were found to be equal or 98 slightly lower than those of air firing, due mainly to the higher cyclone temperature achieved with oxy-99 fuel combustion.

100

101 Biomass properties differ from those of coal in many important ways which results in different 102 combustion behaviours [18]. For example, biomass generally has less carbon, more oxygen, higher 103 hydrogen content and lower heating value. There are huge differences in volatile matter contents 104 between biomass and coal: biomass can lose up to 90% of their masses (as volatiles) in its first stage of 105 combustion, much higher than any ranks of coal (from less than 10% for anthracite to ca. 40% for high-106 volatile bituminous coals) [19, 20]. The effects of oxy-fuel combustion conditions on the combustion 107 performance and emissions of biomass fuels are expected to be differing from those of coal as a result 108 of the differences in properties between biomass and coal. So far, few have investigated oxy-fuel 109 combustion in fluidized bed reactors firing 100% biomass fuels and therefore further research is still

110 needed. Duan et al. [21] conducted a series of experiments firing three kinds of Chinese biomass, i.e. 111 rice husk, wood chips and dry wood flour, under air and oxy-fuel atmosphere in a 10 kWth CFB 112 combustor. The main objective of this study was to investigate the pollutant emissions of the co-firing 113 of biomass with coal under oxy-fuel combustion conditions although experiments firing only the 114 biomass fuels were also carried out for comparison purposes. They observed lower NO emissions in 115 the oxy-fuel atmosphere compared with those with air combustion. This behaviour was explained as the result of the reduced yield of NOx precursors like NH₃ during the devolatilization process and the 116 117 enhanced NO reductions via char/NO/CO reactions under the oxy-fuel combustion conditions. They 118 also concluded that the NO emission increased with the bed temperature, overall oxygen concentration 119 and the primary oxidant fraction when co-firing biomass and coal with a mixing ratio of 0.2 in oxy-fuel 120 combustion. However, the effects of the oxygen concentration in the oxy-fuel oxidant gas on the gas 121 emissions and temperature profiles firing 100% biomass fuels were not investigated in this study.

122

The objective of the present work is to continue delving into the barely-studied oxy-fuel combustion of biomass fuels firing three kind of biomass fuels, two non-woody (miscanthus and straw) and one woody (wood), in a 20 kW_{th} bubbling fluidized bed (BFB) combustor, studying the effects of the combustion atmosphere (air and oxy-fuel) and the oxygen concentration in the oxy-fuel oxidant on the gas emissions and temperature profiles.

128 **2** Experimental

129 2.1 Experimental setup

The experimental system, shown in Fig. 1, mainly includes a BFB combustor (20 kW_{th}) and the auxiliary systems for air supply, biomass feeding, and gas analysis. The combustor consists of a stainless steel reactor of 102 mm i.d. and 800 mm height, a freeboard of 154 mm i.d. and 1100 mm height, and a plenum of 102 mm i.d. and 300 mm height. A water cooled heat extraction probe located inside the bed allows the bed temperature to be controlled by means of the extraction of heat from the reactor. This probe can be moved vertically along the reactor to change the contact surface inside the reactor to prevent the bed from reaching very high temperature values and thus avoid agglomeration and

137 defluidisation of the bed particles. Furthermore, the cooling water flow rate of the probe can be adjusted to control the heat extraction and hence the combustion temperature inside the reactor. 138

139

140 As the real flue gas recirculation is not included with the experimental system, a premixed flow of CO_2 141 and O_2 which are supplied from gas cylinders and monitored by rota meters (calibrated for O_2 and CO_2 142 respectively) is used as the main oxidant for the oxy-fuel combustion and is also used as the fluidising 143 gas of the BFB reactor. For the conventional air combustion tests, compressed air is used as the main 144 oxidant and fluidising gas. The air flow rate is monitored by a rotameter that is calibrated for air. For 145 both oxy and air combustion tests, a small flow of compressed air is also fed through the biomass feeder 146 hopper to prevent backfire and to stop the sand particles coming into the feeding pipe. The oxidant gas 147 is fed into the reactor through a porous stainless gas distribution plate with 100 µm pore size and 12 148 mm thickness. An electric air pre-heater before the plenum and two electric half-cylindrical ceramic 149 radiant heaters surrounding the main bed area are used to preheat the combustion/fluidisation air during 150 the start-up of the combustor. The biomass pellets are fed to the reactor at the location just above the 151 distributor plate by means of a screw feeder. To ensure the fuel feed rate controllable and repeatable, 152 the feeder motor frequency is controlled by an inverter. The flue gas stream leaving the reactor passes 153 through a high efficiency cyclone to recover the elutriated solids and ash and then is exhausted through 154 the ventilation system. The gas composition at the exit of the reactor is continuously analysed by on-155 line gas analysers after going through the water-cooled sampling probe placed inside the gas exit pipe 156 of the cyclone, water condensation traps and particle filters. O₂, CO₂ and CO concentrations are 157 measured by an Easy line continuous gas analyser (ABB, EL3020), while the NOx concentration is 158 measured by a chemiluminescent NOx analyser (Horiba VA-3000). In order to minimise instrumental 159 errors and drifts, gas analysers are regularly calibrated with BOC/Linde Group calibration gases during 160 each test campaign. The reactor is equipped with pressure tapings and K-type thermocouples at different 161 heights. Both the pressure differential across the dense bed and the temperatures along the reactor are closely monitored during each test so that the signs of agglomeration, defluidisation or extremely high 162 163 temperature can be spotted at the earliest opportunity. A data taker and computer system are used to

164 continuously record all of the measured process data (pressure differentials, temperatures, gas165 composition, etc.).





169 2.2 Biomass fuels

170 Three different types of biomass fuels, two non-woody (miscanthus and wheat straw) and one woody (domestic wood), were tested with the 20kWth BFB combustor under both conventional air combustion 171 172 and oxy-fuel combustion conditions. All fuels were purchased from the UK suppliers Brites, in the case 173 of the wood, and Agripellets Ltd, in the case of miscanthus and straw, and used in pellet form. 174 Miscanthus pellets have an average diameter of 6.30 mm and an average length of 18.70 mm, with a 175 bulk density of 603 kg/m³. Straw pellets have an average diameter of 6.15 mm and an average length 176 of 15.60 mm, with a bulk density of 628 kg/m^3 . Wood pellets have an average diameter of 6.00 mm and 177 an average length of 23.10 mm, with a bulk density of 677 kg/m^3 . The proximate and ultimate analyses of the fuels are shown in Table 1. 178

- 179
- 180

Table 1. Ultimate analysis, proximate analysis and calorific values of biomass fuels*.

	Miscanthus	Straw	Wood
Proximate (wt%, dry basis, except moisture)			
Moisture (wt%, as received)	3.34	5.22	3.94
Ash	1.80	6.23	0.70
Volatile matter	82.85	76.31	85.11
Fixed carbon	15.37	17.54	14.19
Ultimate (wt%, dry basis)			
Carbon	45.87	43.80	47.18
Hydrogen	6.74	6.78	6.84
Nitrogen	0.38	0.55	0.17
Sulphur	0.19	0.58	0.17
Oxygen (by difference)	46.82	48.29	45.65
Calorific value HHV, MJ/kg [22]	19.10	18.21	19.81

181 *Fuels are characterised according to the UK/European solid biofuels' standards. In particular, volatile matter is 182 determined at 900°C

183

184 **2.3 Procedure and operating conditions**

The operating conditions for all of the runs and the average flue gas compositions at steady state at every condition are summarized in Table 2. Garside 14/25 sand with a Sauter mean diameter (d_{32}) of

187 0.78 mm and a density of 2655 kg/m³ was used as the inert bed material [23]. The sand was added to

188	the reactor from the top flange up to a height of 25 cm (3.2 kg). The excess oxygen coefficient levels
189	were maintained roughly constant in all experiments (30%). To investigate the effect of the O_2
190	concentration in the oxy-fuel oxidant gas on temperature profiles and gas emissions, a mixture of O ₂
191	and CO_2 gases with various O_2/CO_2 ratios was produced and used as the main oxidant of each oxy-fuel
192	combustion test. As the overall excess oxygen coefficient/oxygen level must remain constant, variations
193	in the O ₂ concentration in the oxy-fuel oxidant were hence achieved through variations in the CO ₂ flow
194	rate and at the same time keeping the O_2 flow rate and fuel feeding rate constant in all the experiments
195	(Table 2). The names given to the different oxy-fuel conditions (oxy-21, oxy-25 and oxy-30, as shown
196	in Table 2) are related with the O ₂ concentration in the oxy-fuel oxidant fed to the reactor under each
197	oxy-fuel combustion test condition (i.e. 21, 25 and 30 vol. %, respectively, as indicated in the 3 rd row
198	at Table 2). However, it is worth mentioning that the overall oxygen concentration for the total amount
199	of oxidants fed to the reactor was somewhat different from the indicated O ₂ concentration in the oxy-
200	fuel oxidant as pure air was fed always to the fuel hopper in order to avoid backfiring. The use of air as
201	the feeder gas instead of an oxy-fuel mixture was resulted from safety concerns, i.e. the failure of the
202	interlock between CO_2 flow and O_2 flow could lead to pure O_2 being fed to the fuel hopper. The 4 th row
203	of Table 2 indicates the average $O_2/N_2/CO_2$ concentration of the all gases fed to the reactor for every
204	condition studied. To investigate the effect of the combustion atmosphere, conventional air combustion
205	experiments were also carried out with all the biomass fuels. In addition, at least three runs were
206	performed with each biomass for each condition studied, in order to verify the results achieved.

Table 2. Operating conditions for oxy-fuel and air combustion experiments and average flue gas compositions at
 steady state for the different conditions studied.

		Air	Oxy-21	Oxy-25	Oxy-30
Excess oxygen coefficient (%)		30	30	30	30
O2 in Oxy-fuel mixture (vol %)		-	21	25	30
Average	$O_2\!/N_2\!/CO_2$	21/79/0	21/22/57	24/25/52	27/28/45
concentration of the all gases fed					
to the reactor (vol. %)					
Feeder air (L/min) ¹		95	95	95	95
Primary air (L/min) ¹		250	-	-	-

Total O ₂ flow (L/min) ¹	72.5	72.5	72.5	72.5		
CO ₂ flow (L/min) ¹	-	197.5	157.5	122.5		
Total gas flow (L/min) ¹	345	345	305	270		
Superficial gas velocity (U)	2.56	2.56	2.27	2.01		
(m/s) ¹						
Fluidization number $(U/U_{mf})^2$	8.94	8.90	7.90	7.02		
Biomass feed rate (kg/h)	3–5	3–5	3–5	3–5		
Static height of bed materials	250	250	250	250		
(mm)						
Average diameter of bed	0.78	0.78	0.78	0.78		
materials (mm)						
Cooling water flow rate (L/min)	1.2 ± 0.2	1.2 ± 0.2	1.2 ± 0.2	1.2 ± 0.2		
$O_2 (vol. \%)$	4.72 ± 0.33	NSV ³	5.27 ± 0.79	5.49 ± 0.37		
<i>CO</i> ₂ (<i>vol.</i> %)	15.65 ± 0.31	NSV	72.20 ± 2.73	69.14 ± 2.09		
POC NOx (ppm)	69.4 ± 8.3	NSV	86.9 ± 5.9	95.8 ± 3.3		
CO (vol. %)	0.47 ± 0.11	NSV	0.52 ± 0.07	0.33 ± 0.02		
\square O_2 (vol. %)	4.70 ± 0.34	NSV	$5.03\ \pm 0.31$	5.63 ± 0.68		
CO_2 (vol. %)	15.74 ± 0.27	NSV	70.23 ± 2.60	68.44 ± 2.20		
NOx (ppm)	111.3 ± 13.6	NSV	156.3 ± 19.2	116.9 ± 7.2		
E CO (vol. %)	0.57 ± 0.20	NSV	0.31 ± 0.09	0.46 ± 0.13		
O_2 (vol. %)	5.03 ± 0.30	NSV	4.97 ± 0.21	5.95 ± 0.61		
<i>CO</i> ₂ (<i>vol.</i> %)	15.35 ± 0.65	NSV	70.61 ± 2.05	68.49 ± 0.66		
NOx (ppm)	183.7 ± 15.4	NSV	340.1 ± 0.1	243.6 ± 34.2		
5 CO (vol. %)	0.45 ± 0.10	NSV	0.17 ± 0.06	0.16 ± 0.08		

210 ¹ Calculated at 20°C and 1 atm.

211 ² Both U and U_{mf} calculated at 800°C.

³Not Stable Value.

213

Fig. 2 shows the temperature profiles and the gas product distribution at the outlet of the reactor in a typical experiment (in this case firing miscanthus). Each series of tests was always started with conventional biomass air combustion. After stabilisation, which took about 2 hours, the combustion environment was changed from air to oxy-fuel with continuous biomass feeding. After finishing the oxy-fuel tests, the combustion oxidant was switched back to air with continuous biomass feeding operation, minimising any errors resulted from biomass feeding rate fluctuations between the two combustion environments.



221

Fig. 2. The temperature profiles, and gas concentrations at the outlet of the reactor during miscanthus pellets combustion under air and oxy-fuel conditions.

225 **3 Results and discussion**

226 **3.1** Effect of combustion atmosphere on fuel combustion and temperature profiles

Fig. 3 shows the temperature profiles along with the height of the reactor under different combustion atmospheres for all fuels. As it can be seen, when using air as oxidant the temperature profile for the non-woody fuels shows a clear maximum located in the splash zone/beginning of the freeboard (T4), above the dense bed, when burned in air. This behaviour happens as a result of the characteristic high volatile matter content of biomass fuels and its release and combustion mostly in the splash zone and freeboard, instead of inside the dense bed as being observed in the case of coal combustion [24]. Other 233 authors reported similar results [25]. The maximum is not that clear in the case of wood as T2 and T4 234 are quite similar. As the volatile matter content is similar for all the fuels (Table 1) the differences in 235 the temperature profiles between the woody and the non-woody fuels could be partly due to the 236 difference in the content of fines in the fuels. To confirm this theory, the fines content of the three 237 biomass fuels used in this work after passing through the screw-feeder was determined. Fines were 238 assumed to be those particles passing through the sieve with apertures of 3.18 mm. The results obtained 239 showed that wood has significantly less fines content than the non-woody fuels (ca. 4 wt. % for wood 240 and between ca. 8.5-9.5 wt.% for the non-woody biomasses), and this implies that a significantly larger fraction of the woody biomass fuel is expected to be burned in the dense bed of the reactor and hence 241 lead to a higher temperatures in that region in comparison to the non-woody biomass fuels. On the other 242 hand, it can also be seen that usually after the temperature peak, a remarkable temperature decrease in 243 244 the freeboard is observed in all three biomass fuels, due to the fact that the heat extracted by the water 245 cooling probe from the upper part of the freeboard is much higher than the heat released from the 246 combustion of any unburned fuels within the freeboard.



248

Fig. 3. Temperature profiles for all fuels under air and different oxy-fuel mixtures. The green arrows placed on the left in all graphs indicate continuous drop of temperatures.

252 As it can observed from Fig. 2 and Fig. 3, replacing the main combustion air with the oxy-fuel mixture 253 with the same oxygen concentration at 21 vol.% caused a significant drop of all temperatures along the 254 reactor and freeboard, with no steady temperature being reached even after one hour of continuous 255 feeding under the same conditions (the arrows in Fig. 3 indicate continuous drop of temperatures). Due 256 to this constant drop in temperatures the steady biomass combustion could no longer be sustained within 257 the reactor and CO and O₂ concentrations increased abruptly, meanwhile NOx concentration showed a 258 sharp decline (Fig. 2). The main reason for the observed temperature decreases is due to the higher 259 specific heat of CO₂ compared with that of nitrogen. It is also mentioned in the literature that CO₂ plays 260 a significant role in reducing the fuel burning rates by limiting the concentration of O and H radicals during oxy-fuel combustion [26]. In order to continue the experiment, the oxygen concentration in the 261 oxy-fuel oxidant gas was increased before the main combustion temperature decreased to about 500 -262

263 600°C. By increasing the O₂ concentration from 21 vol.% to 25 vol.% in the oxy-fuel oxidant gas, a significant increase in the freeboard temperatures (T5-T7) was noticed for all the fuels, as a consequence 264 of both the increase of the O_2 concentration (which resulted in a higher oxidation rate of the fuel) and 265 the reduction of the CO_2 flow rate, almost matching those temperature values achieved with air 266 267 combustion and only the temperature values at the beginning of the freeboard (T4) slightly lower than those observed during air combustion (between 30 and 65 °C lower) (Fig. 2 and Fig. 3). However, the 268 269 main bed temperatures (T2 and T3) remained quite low and were about 100 - 200 °C lower than those 270 observed with air combustion. To almost fully match the temperature profile of oxy-fuel combustion to 271 that of air combustion, the oxygen concentration in the oxy-fuel mixture had to be increased to about 272 30 vol.% as it can be seen from Fig. 2 and Fig. 3. With 30 vol.% O_2 in the oxy-fuel oxidant, the 273 temperatures were seen to be slightly higher than those achieved with air, especially for wood and 274 miscanthus. These results agree with literature which suggest the required O_2 concentration in the oxy-275 fuel oxidant lies in the range 27 - 30 vol.% [16, 27-29].

276

277 **3.2 Effect of combustion environment on CO emissions**

278 The results of CO emissions from different types of biomass fuels tested under air and oxy-fuel 279 combustion are plotted in Fig. 4. The results are expressed in the amount of emission per energy unit 280 (ng/J). This kind of emission unit is commonly used in oxy-fuel literature as the volumetric flow rate 281 of the flue gas of an oxy-fuel combustion plant with real flue gas recirculation is much lower (up to 282 80%) than that of conventional air combustion, and thus the conventional emission unit of volumetric 283 concentration (ppm or vol.%) cannot be used for direct comparison of emissions between two 284 combustion environments. As no steady conditions were reached at oxy-21 conditions, CO and NOx 285 emissions of oxy-21 combustion tests were not going to be compared with those of air combustion and 286 oxy-30 combustion tests both of which had achieved steady-state conditions (Fig. 2). The results of the oxy-25 combustion tests were included for the comparison but it should be noted that these tests were 287 288 only reached quasi-steady state conditions (Fig. 2). Fig. 4 shows that the switch from air to oxy-25 led 289 a decrease in the CO emission for all the fuels but the reductions in CO emissions were more

290 pronounced with the non-woody biomass fuels. As commented in Section 2.3, the increase of the 291 oxygen concentration from 21 vol.% to 25 vol.% in the oxy-fuel atmosphere was achieved through 292 reducing the CO_2 flow rate while maintaining the O_2 flow rate (in order to maintain constant the excess 293 oxygen coefficient/O₂ value), and this resulted in a lower overall fluidizing gas flow rate than that used 294 with air combustion and hence a longer residence time of the combustible gas in the reactor, benefiting 295 the oxidation of CO with O₂. However, as it can be seen from Fig. 2 and Fig. 3, the main bed 296 temperatures (T2 and T3) achieved under the oxy-25 combustion conditions were significantly lower 297 than those obtained under the air combustion conditions (between 100 and 200 °C lower as indicated 298 in Section 3.1), due to the aforementioned higher specific heat of CO_2 compared to N_2 . The lower bed temperatures meant more incomplete carbon combustion and as a consequence higher CO emissions in 299 300 the bed zone, agreeing with the trend observed by Duan et al. [16]. The observation of lower CO 301 emissions with oxy-25 than those of air combustion shown in Fig. 4 further proves that when firing 302 biomass fuels in a FB reactor the main combustion reaction takes place mainly above the dense bed in 303 the splash region and/or at the beginning of the freeboard, while the main bed zone plays a minor role 304 for the combustion of biomass fuels. As shown in Fig. 2 and Fig. 3, for each biomass fuel, the gas 305 temperatures in the freeboard with oxy-25 were slightly higher than those achieved with air combustion 306 and this, together with the longer residence time mentioned above, could have led to the lower CO 307 emissions measured at the exit of the cyclone. The reduction in CO emissions when switching from air 308 to oxy-25 was significant in the case of wood (from 2600 to 1900 ng/J, a 26% decrease) and even more 309 pronounced for the non-woody fuels (from 3250 to 1500 ng/J firing miscanthus, a 54% decrease, and 310 from 2900 to 600 ng/J firing straw, a 80% decrease). The difference observed between the woody 311 biomass and the non-woody fuels could partly be due to the difference in the content of fines in the 312 fuels: as the wood has significantly less fines, an important part of the combustion of this fuel should take place in the low-temperature dense bed zone, and hence resulting in higher CO concentrations in 313 314 the dense bed zone and an overall less reduction in CO emissions when oxy-25 replaces the air 315 combustion.

317 As it can also be seen from Fig. 4, increasing the O₂ concentration from 25 to 30 vol.% resulted in a further decrease in the CO emissions in the case of wood, while CO emissions slightly increased for 318 319 miscanthus and slightly decreased for straw. When the O₂ concentration in the oxy-fuel oxidant gas 320 reached 30 vol.%, the gas temperature profiles in the whole reactor almost completely matched the 321 temperature profiles achieved with air combustion (as seen in Fig. 2 and Fig. 3), and hence CO 322 emissions were expected to be lower than those of air combustion and oxy-25 combustion as observed 323 with the oxy-combustion of wood. The trends observed with different fuels could be further complicated by the aforementioned differences in the content of fines in the fuels and by the fact that the oxy-25 324 325 combustion tests had only reached quasi-steady state conditions. Wood has a much lower content of 326 fines comparing with the non-woody fuels, which leads to an increase in the importance of the dense 327 bed on the CO chemistry.

328



Fig. 4. CO emissions from different types of biomass fuels under air and different oxy-fuel combustionenvironment.

332 3.3 Effect of combustion environment on NOx emissions

333 The NOx emissions from different types of biomass fuels studied under various combustion 334 environments are presented in Fig. 5. Fig. 5 clearly shows that the NOx emissions' level depends 335 directly on the nitrogen content of the fuel, indicating that NOx are mainly originated from fuel-N 336 conversions [30]. Prompt-NOx and thermal-NOx weren't expected to be of importance due to the 337 relatively low combustion temperatures reached at the reactor. It can be seen from Fig. 5 that the NOx 338 emissions increased when the combustion environment was switched from air to oxy-25. In order to 339 explain this behaviour it is necessary to briefly review the fuel-NOx formation mechanism, which can 340 be summarized as follows. As the fuel is fed into the furnace, volatile release takes place. The fuel nitrogen released with the volatiles (volatile-N) further decomposes into NOx precursors such as 341 342 cyanide (HCN) and ammonia (NH₃), which are produced mostly from the amino acid containing fuel-N in the pyrolysis process [31]. Furthermore, NH₃ can be regarded as the primary product of fuel-N, 343 344 directly from pyrolysis or the secondary conversion from HCN through reaction R1 [32-35].

$$345 \qquad HCN + 2[H] \rightarrow NH_3 + [C] \tag{R1}$$

346 Depending on temperature and oxygen concentration, NH_3 could be partly oxidized to NO (reaction 347 R2) or acts as a reducing agent to reduce NO into N₂ (reaction R3).

348
$$NH_3 + \frac{5}{4}O_2 \to NO + \frac{3}{2}H_2O$$
 (R2)

349
$$\frac{2}{3}NH_3 + NO \rightarrow \frac{5}{6}N_2 + H_2O$$
 (R3)

In this study, biomass was fed at the bottom of the dense bed zone, right above the distributor plate. Therefore, there were various fuel-rich (or oxygen-depleted) pockets existing in the dense bed zone. Within these fuel-rich pockets, a higher NH₃ concentration would favour NO reduction (reaction R3). At the same time, a large amount of solid fuel particles existed in the dense bed zone, thus enhancing the NO reduction by CO and char via reaction R4 [36]:

$$355 \qquad NO + CO \xrightarrow{char} \frac{1}{2}N_2 + CO_2 \tag{R4}$$

356 As shown in Fig. 2 and Fig. 3, the bed temperatures under the oxy-25 combustion conditions were considerably lower than those achieved under the air combustion conditions, whereas the temperatures 357 358 of the freeboard region, especially the top part of the freeboard, under the oxy-25 combustion conditions 359 were similar to or slightly higher than those under the air combustion conditions. Therefore, more 360 volatiles were combusted in the oxygen-rich freeboard zone under the oxy-25 combustion conditions 361 than under the air combustion conditions, hence favouring the oxidation of volatile-N such as NH_3 and 362 enhancing the formation of NOx via reaction R2. This explains the higher NOx emissions found under 363 the oxy-25 combustion conditions. The results obtained here with the biomass fuels disagree with the common trend found in the literature firing coal. NOx emissions in oxy-coal combustion with low O_2 364 365 concentrations (between 21-25 vol.%) in the oxidant have been found to be usually lower than those 366 observed with air-firing atmospheres, mainly because of the lower temperatures achieved in the bed 367 region at the oxy-coal conditions [11, 37-39]. In the present work, the main bed temperature reached 368 when burning the biomass fuels under the oxy-25 conditions was also lower than that obtained under 369 the air combustion conditions. However, the biomass fuels tested in the present study contain much 370 higher amounts of volatiles (76 - 85 wt. %) than the coals investigated with the previous studies of oxy-371 coal combustion (5 and 36 wt. %) [11, 37-39]. The much higher volatile matter contents with the 372 biomass fuels would mean a larger part of each biomass fuel being burned in the high-temperature 373 freeboard zone in comparison to coal, and accentuates again the decrease in the importance of the dense 374 bed on pollutant emissions when firing biomass fuels.



Fig. 5. NOx emissions from different types of biomass fuels under air and different oxy-fuel combustion
 environment.

378

379 As shown in Fig. 5, the increase in NOx emissions is more pronounced for fuels with higher fuel-N 380 contents. The increase was seen to be largest for the wheat straw pellet, almost doubling the NOx 381 emissions value obtained under the air combustion conditions (from 160 ng/J to 300 ng/J approx.). On 382 the other hand, when firing wood the NOx emissions remained at almost the same value when switching from air to oxy-25. This is not unexpected as the wood pellet fuel contains a very low level of fuel-N 383 384 which means other NOx formation routes such as thermal-NOx and prompt NOx can also contribute to 385 the observed NOx emissions. In addition, the wood pellet fuel has a lower fines content and therefore 386 more fuel combustion and NOx formation happen in the dense bed region, weakening the difference in 387 NOx emissions between air combustion and oxy-25 combustion seen with the non-woody biomass 388 fuels.

Switching from oxy-25 to oxy-30 resulted in a significant decrease of the NOx emissions for the non-390 391 woody fuels, leading to similar NOx emission levels in comparison to the air-fired tests. As it was seen in Section 3.1, switching from oxy-25 to oxy-30 led to an increase of the dense bed zone temperature, 392 393 matching that achieved under the air combustion conditions. As a result, more volatile-N was released 394 and converted in the dense bed zone which contains many fuel-rich pockets that favour the reduction 395 of NO to N₂ via reaction R3. Furthermore, as explained in Section 2.3, a higher O₂ concentration in the 396 oxidant implies a lower CO₂ flow rate, in order to keep constant overall excess oxygen 397 coefficient/oxygen in the system. The lower CO₂ amount fed to the reactor meant less total oxidant flow 398 to the reactor and therefore less dilution of the gases inside the reactor. Due to this, higher concentrations 399 of NOx reducing species, especially CO, were expected in the dense bed zone which now has similar 400 temperatures to those of air combustion (Fig.2 and Fig.3), promoting the heterogeneous reduction of 401 NOx on the char surfaces, according to reaction R2. Higher O_2 concentrations in the bed zone with oxy-402 30 could also promote the oxidation of volatile-N to NOx, hence offsetting the reductions of NOx by 403 CO, char and other NOx reducing species. This may partly explain why the NOx emissions of straw 404 combustion under the oxy-30 condition conditions were found to be slightly higher than those under 405 the air combustion conditions, whereas the opposite trend (i.e. slightly lower) was observed for the 406 miscanthus combustion. A significant portion of the formed CO in the dense bed is burned afterwards 407 in the high-temperature freeboard, leading to lower CO emissions at the outlet of the reactor under the 408 oxy-30 combustion conditions than under the air combustion conditions (Fig. 4). Liu et al. [40] found 409 that the conversion of fuel-N to NOx for coal combustion in 30% O₂/70% CO₂ was smaller than that 410 for coal combustion in air. They explained this trend in terms of the lower dilution of the gases 411 accomplished when working under the oxy-fuel condition, which increase the concentration of reducing 412 species inside the reactor. Andersson et al. [41] found that the formation of NO from fuel-N in oxy-fuel 413 combustion was the same, or slightly higher, than that under air-firing conditions, whereas the reduction 414 of NO could be up to 50% greater under oxy-combustion conditions. Lupiañez et al. [42] found similar NOx emissions in both air and oxy-fuel conditions (with O2/CO2 ratios of 25/75 and 40/60) in the 415 416 combustion of lignite in a 90 kW fluidized bed combustor. Díez et al. [43] observed a strong diminution 417 in NOx emissions when the O₂ concentration in the oxy-fuel oxidant was increased to 40 and 60%,

when firing anthracite in a 90 kW fluidized bed combustor. The authors explained this behaviour in
terms of the heterogeneous (char, CaCO₃, CaO) and homogeneous (CO) interactions of the NOx
formed, which can significantly contribute to NOx depletion in the dense bed zone.

421

422 **4** Conclusions

423 Three biomass fuels, one woody and two non-woody, have been tested in a 20 kW_{th} fluidized bed 424 combustor. The effect of combustion atmosphere (air or oxy-fuel) and oxygen concentration in the 425 oxidant under the oxy-fuel combustion conditions on temperature profiles and gas emissions (NOx and 426 CO) were systematically investigated. The following conclusions can be drawn from the obtained 427 experimental results:

428 (1) Simply replacing the air with an oxy-fuel oxidant comprised of 21 vol.% O_2 and 79 vol.% CO_2 429 results in a significant decrease in gas temperatures and ultimately leads to the extinction of the 430 biomass flame due to the larger specific heat of CO_2 compared to N_2 . To keep a similar 431 temperature profile to that achieved under the air combustion conditions, the oxygen 432 concentration in the oxy-fuel oxidant of O_2/CO_2 mixture has to be increased to ca. 30 vol.%.

433 (2) A drastic decrease in CO emissions can be achieved for all the biomass fuels (up to 80%
434 reduction when firing straw) under oxy-fuel conditions when the oxygen concentration in the
435 oxy-fuel oxidant is 25 vol.% or more as a result of the higher residence time of the gas inside
436 the reactor and the freeboard/reactor temperature profile matching that of air combustion.

(3) NOx emissions decrease with the oxygen concentration in the oxy-fuel oxidant due to i) the
increase of bed temperature, which implies more volatile-N released and converted in the
highly-reducing dense bed zone and ii) the less dilution of the gases inside the dense bed zone,
which leads to a higher CO concentration in this region enhancing the reduction of NOx.
Similar NOx emissions to those obtained with air combustion were found when the biomass
fuels were burned in the oxy-fuel oxidant gas containing 30 vol.% oxygen.

(4) The freeboard temperature is the dominant variable influencing both CO and NOx emissionscompared with the influence of bed temperature, especially when firing the non-woody fuels.

445 Acknowledgment

- 446 This work was supported by the UK Engineering and Physical Sciences Research Council [grant
- 447 number EP/M01536X/1].

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