

Experimental Study of a Single Particle Reactor at Combustion and Pyrolysis Conditions

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Emission formation during thermal conversion of biomass fuels at different conditions has been the scope of this study. The experiments were conducted in a quartz glass reactor where the temperature and atmosphere can be adjusted. The fuels were selected accurately in order to cover a wide range of biomass/waste compositions. They are torrefied softwood, spruce bark, waste wood, miscanthus, and wheat straw. The fuels were first grinded and then pressed into pellets of the same size and weight with a pellet maker. Each pellet was about 200 mg.

The experiments were performed under combustion and pyrolysis condition, with atmosphere of 3 % O₂ and 100 % N₂, respectively. Each fuel was combusted under temperatures of 800, 900, and 1,050 °C. O₂ and the formed SO₂, NO, CO and CO₂ were monitored by three analysers. The fuel pellet was kept under combustion and pyrolysis at four residence times. The residue weight was measured after each process and the comparison with the ash content of the original fuel is made. Time dependent formation of NO and SO₂ and other emissions is presented and discussed in this paper. Effect of temperature and combustion condition is also considered for the conclusion.

1. Introduction

Currently, the contribution of bioenergy resources to the global energy production is substantial and it is also increasing with a high rate. It is the only renewable source that can replace fossil fuels in all energy markets – in the production of heat, electricity, and fuels for transport. With the current rate, it is expected that energy from biomass sources can be expanded even more in the future, which could result in a higher potential for greenhouse gas savings and reduction of environmental emissions. On the other hand, additional social-economic opportunities besides the bioenergy conversion process and energy security for rural areas are important factors which may enhance the bioenergy share. According to an IEA report, bioenergy could sustainably contribute between a quarter and a third of global primary energy supply in 2050 (Bauen et al., 2009).

However, the formation of emissions such as CO, NO_x, particulates, and SO_x are the main concerns during the conversion of biomass to energy, e.g. combustion. State of the art studies focus on the efficiency increase of the emission reduction techniques (Dvořák et al., 2011). The formation of these pollutants depends on many operating parameters and inherent factors. Temperatures, mixing condition, excess air ratio (Houshfar et al., 2011), residence time, reactor design, fuel elemental composition (Glarborg et al., 2003), ash content etc. are among the main affecting factors. Effect of blending various types of fuels with different composition is also investigated to enable maximum use of herbaceous fuels (Houshfar et al., 2012). The formation of one emission have also effects on the release of others (Wei et al., 2004).

Release of chlorine and potassium will also result in increased corrosion to the super-heater tubes and boiler section in general (Khalil et al., 2011). On the other hand, to have a pure study on the release rate of e.g. fuel nitrogen or fuel sulphur, pyrolysis condition can be used to investigate those effects. Fate of fuel nitrogen is investigated in a recent study using the single particle reactor (Vähä-Savo et al., 2011). Emission formation (SO₂, NO) during thermal conversion of biomass fuels at different conditions has been the scope of this study. CO and CO₂ were also measured and compared at different temperature ranges. The fuels are selected in a way to cover a wide range of solid biomass fuels.

2. Experimental setup and test conditions

The experimental system, measurement devices, fuel properties, and test conditions are described in this section.

2.1 Fuels

The fuels were selected accurately in order to cover a wide range of biomass/waste compositions. They are torrefied softwood, spruce bark, waste wood, miscanthus, and wheat straw. The fuels were first grinded and then pressed into pellets of the same size and weight with a pellet maker. Each pellet was about 200 mg. Fuel composition is presented below. Proximate and ultimate analyses of the fuels are presented in Table 1 and Table 2.

Table 1: Proximate analysis of fuels

Fuel	Origin	Moisture content	Ash content	Volatile matter + Fixed Carbon
		H ₂ O	Ash	VM+FC
		wt % w.b.	wt % d.b.	wt % w.b.
Torrefied softwood	Austria	2.1	0.48	97.4
Spruce bark	Austria	12.8	5	82.8
Waste wood	Austria	13.2	1.7	85.3
Straw	Denmark	10	5.6	85.0
Miscanthus	Austria	9.7	2.3	88.2

Table 2: Ultimate analysis of fuels (as dry basis)

Fuel	Origin	Carbon	Hydrogen	Nitrogen	Oxygen	Sulphur	Chlorine
		C	H	N	O*	S	Cl
		wt % d.b.	wt % d.b.	wt % d.b.	wt % d.b.	mg/kg d.b.	mg/kg d.b.
Torrefied softwood	Austria	56.8	5.7	0.03	36.99	38.7	11
Spruce bark	Austria	49.7	5.6	0.34	39.32	301	98.8
Waste wood	Austria	49	6.1	1.33	41.52	370	3,100
Straw	Denmark	45.9	5.9	0.64	41.66	1,110	1,900
Miscanthus	Austria	48	5.9	0.28	43.30	392	1,810

* Calculated by difference

The standard methods are used to characterize the fuels. Determination of the C-, H-, and N-contents was with ÖNORM CEN/TS 15104. ÖNORM CEN/TS 12290/15297 and ÖNORM CEN/TS 15289 were implemented for sulphur and chlorine detection. Moisture and ash content were also determined with ÖNORM CEN/TS 14774 and CEN/TS 14775, accordingly.

2.2 Single particle reactor

The experiments were conducted in a quartz glass reactor called the single particle reactor, where the temperature and atmosphere can be adjusted. The reactor is shown in Figure 1.

The experiments are performed under combustion and pyrolysis condition, with atmosphere of 3 % O₂ and 100 % N₂, respectively. Each fuel was combusted under temperatures of 800, 900, and 1,050 °C, respectively. The fuel pellet was kept under combustion and pyrolysis at four residence times: 1- Flameout, 2- 50 % Char burnout, 3- Char burnout, and 4- Char burnout+5min. Flameout is the time where the devolatilization stage is finished, i.e. flame is disappeared. The "char burnout" is the time when the CO₂ signal is zero on the gas analyser, and consequently "50 % char burnout" is half of this time. The residue

weight was measured after each process and the comparison with the ash content of the original fuel is made.

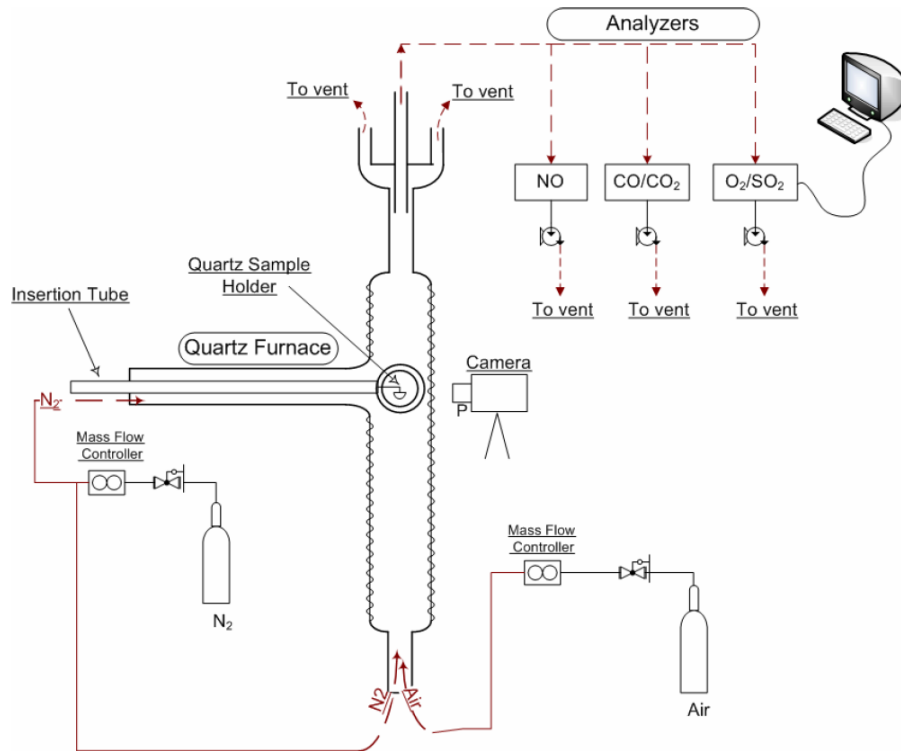


Figure 1: Schematic of the single particle reactor setup and emission monitoring devices

Table 3: Experimental matrix

Fuel	Temperature (°C)	Atmosphere	Fuel	Temperature (°C)	Atmosphere
Bark	800	3 % O ₂	Bark	800	100 % N ₂
Bark	900	3 % O ₂	Bark	900	100 % N ₂
Bark	1,050	3 % O ₂	Bark	1,050	100 % N ₂
Miscanthus	800	3 % O ₂	Miscanthus	800	100 % N ₂
Miscanthus	900	3 % O ₂	Miscanthus	900	100 % N ₂
Miscanthus	1,050	3 % O ₂	Miscanthus	1,050	100 % N ₂
Torrefied Wood	800	3 % O ₂	Torrefied Wood	800	100 % N ₂
Torrefied Wood	900	3 % O ₂	Torrefied Wood	900	100 % N ₂
Torrefied Wood	1,050	3 % O ₂	Torrefied Wood	1,050	100 % N ₂
Waste Wood	800	3 % O ₂	Waste Wood	800	100 % N ₂
Waste Wood	900	3 % O ₂	Waste Wood	900	100 % N ₂
Waste Wood	1,050	3 % O ₂	Waste Wood	1,050	100 % N ₂
Straw	800	3 % O ₂	Straw	800	100 % N ₂
Straw	900	3 % O ₂	Straw	900	100 % N ₂
Straw	1,050	3 % O ₂	Straw	1,050	100 % N ₂

2.3 Measurement system

O₂ and the formed SO₂, NO, CO and CO₂ were monitored by three analysers. NO were measured with two analysers, Servomex and Teledyne model 200EM, where Servomex data was used for NO level above 50 – 60 ppm and Teledyne was more accurate for NO concentration below 60 ppm. The flow was dominated by the feed, implying that the mass flow rate of a species is proportional to the measured concentration.

3. Results and discussion

Time dependent formation of NO and other emissions is presented and discussed in this paper. Effect of temperature and combustion condition is also considered for the conclusion. This study extends current understanding of the nature of biomass particle combustion and char formation with regard to various emissions in a same reactor (Karlström et al., 2013) and fluidized bed reactors (Kilpinen et al., 2002). Because of the high temperatures, the drying process was fast and had overlap with devolatilization, so the drying phase was not determined. To determine devolatilization process and char burnout time, we have observed the flame envelope through a viewing port as well as monitored CO₂. The devolatilization and char burnout time for different fuels and at various temperature ranges is as below:

Table 4: Required time (seconds) for the devolatilization and char burnout

Fuel	Flameout			Char burnout		
	800 °C	900 °C	1,050 °C	800 °C	900 °C	1,050 °C
Torrefied softwood	60	50	40	2,850	1,790	1,200
Spruce bark	60	50	60	2,240	1,540	1,140
Waste wood	80	45	55	1,980	1,020	870
Straw	60	50	45	1,090	760	600
Miscanthus	60	50	50	1,500	980	660

The typical time-dependent concentration diagram for one of the experiments is shown in Figure 2. In this case (combustion of straw with 3 % O₂ at 900 °C), NO release reaches a peak after about half a minute and then drops rapidly. The figure shows that most NO is released during the devolatilization stage. The char nitrogen conversion, however, takes a long time (around 12 min).

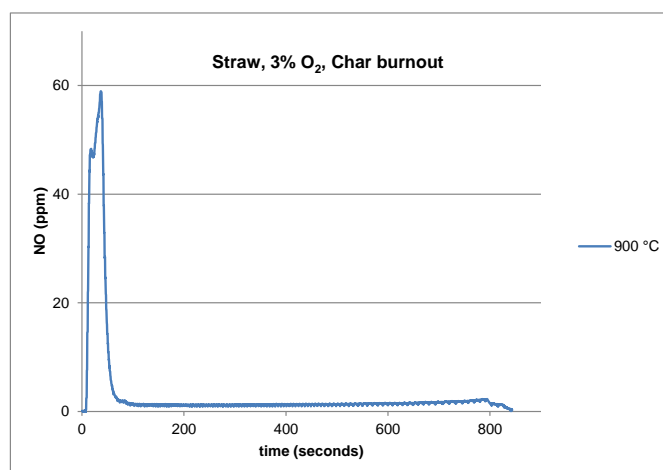


Figure 2: Nitrogen oxide concentration (ppm) measured from start of combustion up to complete char burnout for straw at 900 °C

Figure 3 shows molar concentration of four measured gases at 800, 900, and 1,050 °C for bark combustion with 3 % O₂. The other four fuels follow the same trends, but the range is different since the nitrogen and sulphur content is different in the fuels. The peaks in the graphs correspond to the devolatilization process, where the char burnout starts afterwards. After the NO peak there is a minimum point in the NO curve, after this minimum the NO formation slowly again increases to until all fuel nitrogen is oxidized. This minimum point is considered as the start of char oxidation, i.e. char-N conversion. The char-N is converted mainly to NO since the temperature is relatively low and there is unlikely to form nitrous oxide, N₂O (Winter et al., 1999).

Increased temperature from 800 to 900 °C has little effect on the NO formation, even though a little decrease is shown in the graph. Temperature increase to 1050 °C, however, decreased the NO formation considerably by 90 %. Higher temperature, increase the level of CO and SO₂ formation. The NO release from biomass fuels during char oxidation is discussed in detail in a recent study (Karlström et al., 2013).

Figure 4 shows the molar concentration for the pyrolysis process of bark samples. The NO release is reduced by a factor of 5 – 6 at 800 and 900 °C while at 1,050 °C the NO remains almost the same.

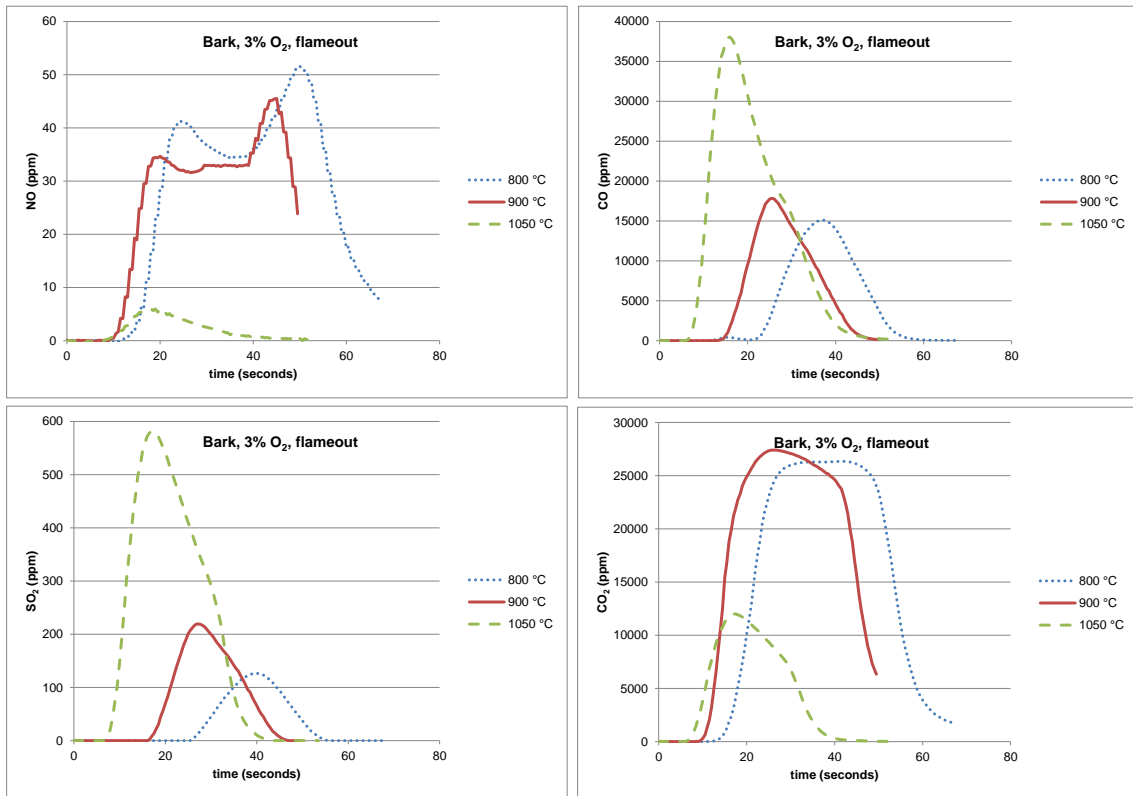


Figure 3: Gas concentration at different temperatures in the flue gas for combustion of bark with 3 % O₂

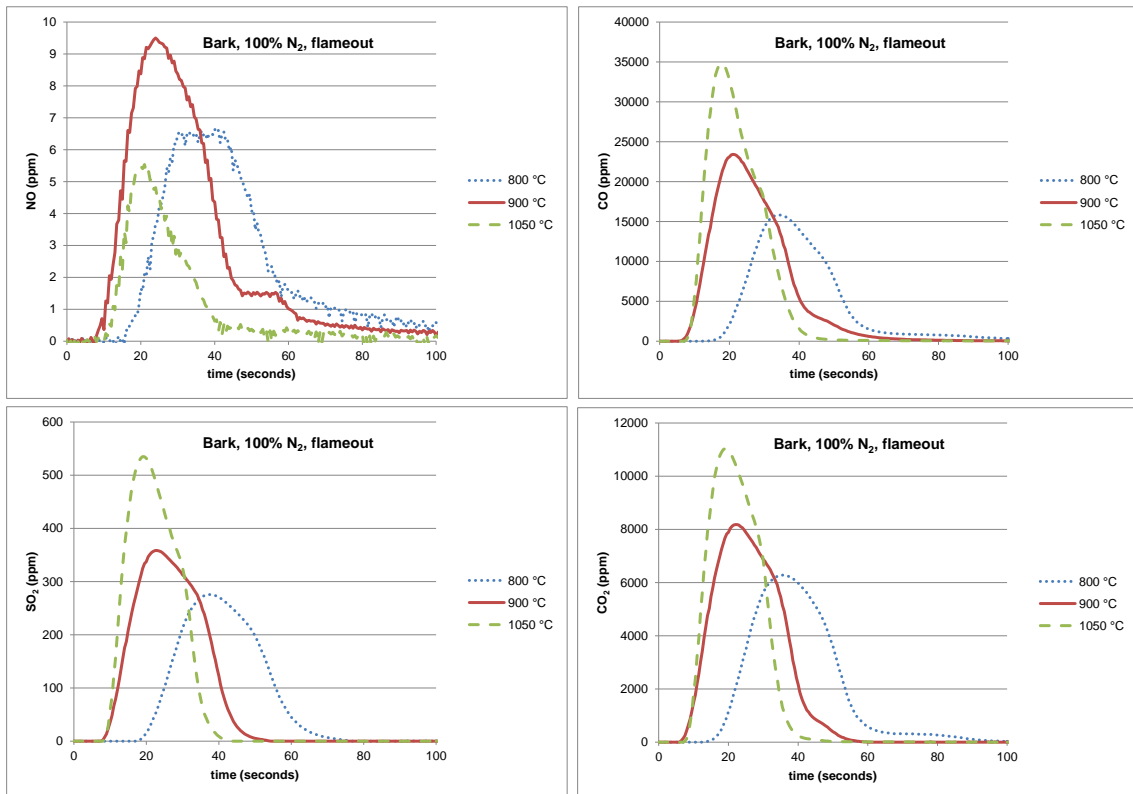


Figure 4: Gas concentration at different temperatures in the flue gas for pyrolysis of bark with 100 % N₂

The observed NO behaviour is very interesting. One possibility is that due to the high temperature at 1,050 °C, the devolatilization rate is such high that all O₂ is consumed for oxidizing them. On the other hand, there is a significant increase in the CO level. The reduction in NO could also be due to secondary reactions in the flame envelope, like de-NO_x reactions

In general, the SO₂ release with and without O₂ looks rather similar. SO₂ was, however, increased at 100 % N₂ condition for the lower temperature range, which may be explained from the interaction of other formed emissions, e.g. CO, with sulphur oxides.

4. Conclusions

In this study, the release of nitrogen oxide (NO) and other emissions (CO, CO₂, and SO₂) was investigated during combustion and pyrolysis of biomass particles from bark, miscanthus, torrefied wood, waste wood, and straw. Experiments were performed in a single particle reactor at 900 – 1,050 °C and with 3 or 0 vol % O₂. Continuous gas analysing systems were used to determine the flue gas composition in the reactor exit. NO and other emissions are correlated with temperature at combustion condition. The formation of NO at 1,050 °C is very low while in this temperature the effect of other NO_x formation mechanism, e.g. thermal NO, is also negligible. It seems that especially at 1,050 °C the devolatilization is so strong that there is a shortage of O₂ also for the 3 % O₂ case. NO formation with 100 % N₂ atmosphere was suppressed by a factor of 5–6. The lower fuel-N content results in a lower NO concentration in the flue gas; torrefied wood with the lowest nitrogen content gives about 12 ppm as peak in the combustion condition. SO₂ concentration increased at pyrolysis condition compared to combustion experiments.

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