

## RELEASE AND CONVERSION OF FUEL-BOUND NITROGEN DURING FIXED-BED GASIFICATION AND SUBSEQUENT STAGED COMBUSTION

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**ABSTRACT:** This paper presents experimental results derived from test runs performed with a laboratory-scale updraft fixed-bed gasifier coupled to a combustion chamber to produce data for the investigation of the release behaviour and the conversion of fuel-bound nitrogen during gasification and subsequent staged combustion of the producer gas using softwood pellets. The concentrations of relevant nitrogenous gas species including tars have been measured in the producer gas and at different positions in the combustion chamber. Based on the experimental measurements and results derived from the test runs, the pathway of the fuel-bound nitrogen could be described. Results show that during updraft fixed-bed gasification the fuel-bound nitrogen is mainly released as N bound in tars from the packed bed and is then subsequently released as HCN, NO, NH<sub>3</sub> and N<sub>2</sub> as a result of tar cracking during combustion. This strong N-fixation in the tars was not expected. It is of great relevance for the understanding of the behaviour of the fuel-bound nitrogen as a basis for a low-NO<sub>x</sub> combustion of the producer gas.

**Keywords:** fixed-bed gasification, nitrogen release, staged combustion

### 1 INTRODUCTION

Amongst the thermo-chemical conversion technologies, biomass gasification has attracted a high interest as it may offer higher efficiencies in relation to combustion. As the market for small-scale residential combustion units is expected to further increase within the next years, small-scale fixed-bed gasifiers may also be used for heating purposes as well as for micro-CHP applications (e.g. Stirling engine or micro-turbine) in the near future.

The scope of this work is the experimental investigation of a laboratory scale fixed-bed updraft gasifier coupled to a combustion chamber for heat production with respect to the release behaviour and the conversion of fuel-bound nitrogen during gasification and subsequent staged combustion of the producer gas accurately considering the influence of tars as it is well known that updraft gasifiers produce high amounts of tars. During gasification of biomass fuels, nitrogen compounds, such as HCN, NH<sub>3</sub>, and nitrogen oxides (NO, NO<sub>2</sub> and N<sub>2</sub>O) are released from the fuel to the gas phase [3]. Several authors identified HCN, NH<sub>3</sub> and N<sub>2</sub> to be the major nitrogenous components of the producer gas of gasifiers [3, 4 and 5]. However, these results have been derived from experiments performed with fluidised bed gasifiers. For fixed-bed updraft gasifiers no data are available. In the combustion chamber these nitrogenous species further react and partly form NO<sub>x</sub>, which is a primary contributor to photochemical smog. Based on the experimental measurements and results derived from the test runs, the pathway of the fuel-bound nitrogen could be described. The understanding of the behaviour of the fuel-bound nitrogen is of great relevance for a low-NO<sub>x</sub> combustion of the producer gas.

This paper presents the experimental results from a laboratory scale fixed-bed updraft gasifier operated with softwood pellets coupled to a staged combustion chamber for heat production.

### 2 THEORETICAL BACKGROUND

#### 2.1 Gasification

The updraft gasifier is the oldest and simplest form of fixed-bed gasifiers. It can handle biomass fuels with high ash (up to 15 %) and high moisture content (up to 50 %) [1]. It is more robust than other fixed bed gasifiers because it is less sensitive to variations in size and quality of biomass. In an updraft gasifier, the biomass fuel is fed continuously from the top of the gasifier, which is usually a cylindrical reactor, and forms a packed bed on the grate. The gasification agent, dry or humidified air, is injected from the bottom below the grate, passes through the fuel bed and hot product gases exit the gasifier at the top at around 420-570 K. The overall gasification process can be separated into four different reaction zones, stratified along the reactor height. At the top of the bed the fuel is heated and dried by up-streaming hot gases. Above temperatures of about 500 K pyrolysis of the fuel takes place and solid char particles and volatiles are formed. The char particles move downwards, heat up and get reduced by hot gases as gasification processes start above approximately 1000 K.

Finally the remaining char is burnt by the supplied air (primary air) at the bottom of the gasifier, supplying heat for the overlying processes. The fuel ash falls through the grate. Due to char combustion, the solid temperature attains high values slightly below 1500 K above the grate [4]. Thus, updraft gasifiers are susceptible to blocking due to ash fusion. The producer gas consists of CO, H<sub>2</sub>, CO<sub>2</sub>, CH<sub>4</sub>, water vapour, nitrogen - if air is used as gasification agent - high amounts of tars and traces of higher hydrocarbons, such as ethane and propane, as well as organic acids.

In biomass grate furnaces the fuel-bound nitrogen is mainly released as HCN, NH<sub>3</sub> and nitrogen oxides (NO, NO<sub>2</sub> and N<sub>2</sub>O) from the bed [6]. Several authors identified HCN, NH<sub>3</sub> and N<sub>2</sub> as the major nitrogenous components of the producer gas of gasifiers [3, 4 and 5]. With respect to updraft gasification this is not correct as it will be shown in this paper.

#### 2.2 Combustion

In the combustion chamber the producer gas is mixed with combustion air and burnt almost completely. A basic concept of air staging was developed for the combustion

chamber based on CFD modelling in order to reduce the NO<sub>x</sub> emissions in the flue gas. Air staging is based on the introduction of the combustion air by two separate streams, secondary and tertiary air (see Fig. 1). Thus, the combustion chamber consists of two different reaction zones, whereas the first combustion zone is realised as oxygen-deficient, fuel-rich zone. Through the reducing conditions in this zone the formation of fuel NO is inhibited [7]. In the second combustion chamber (oxidizing zone) the remaining combustion air is supplied in order to provide complete burnout of the flue gas.

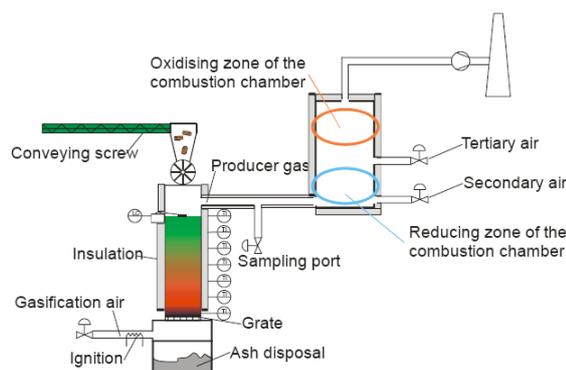
### 3 EXPERIMENTAL PROCEDURE

Several test runs have been performed with a laboratory-scale experimental facility in order to investigate the release and conversion of the fuel-bound nitrogen.

#### 3.1 Experimental facility

Fig. 1 shows the basic geometry of the experimental facility used. The core of the facility is the updraft gasifier, which consists of a cylindrical shaft, covered with an insulation made of ceramic fibres, and with an inner diameter of 0.125 m and a length of 0.6 m. The pellets are fed from the top of the gasifier by means of an air-tight rotary feeder. The height of the fuel bed inside the gasifier is held constant at about 0.42 m by means of a mechanical level control. The gasification air is injected from the bottom below the grate, which is used as air distributor. The ash produced in the process falls through the grate and is discharged by means of a conveying screw.

The gasifier is coupled to a combustion chamber through a well isolated tube. The cylindrical combustion chamber with a length of approx. 0.5 m, insulated with firebricks, is surrounded by a water boiler, which is coupled to an external heat exchanger. Air staging is applied in order to reduce NO<sub>x</sub> emissions in the flue gas. Thus, the combustion chamber is separated into two different reaction zones, a reducing and an oxidising zone. The distance between secondary and tertiary air nozzles is 0.25 m. Secondary (reducing zone) and tertiary (oxidising zone) air are injected through radial air nozzles in order to provide efficient mixing of unburned flue gas with the combustion air for a complete burnout. The design of the combustion chamber was optimised by CFD simulations. A flue gas fan placed at the outlet of the combustion chamber covers pressure losses and controls the pressure inside the gasifier.



**Figure 1:** Experimental facility

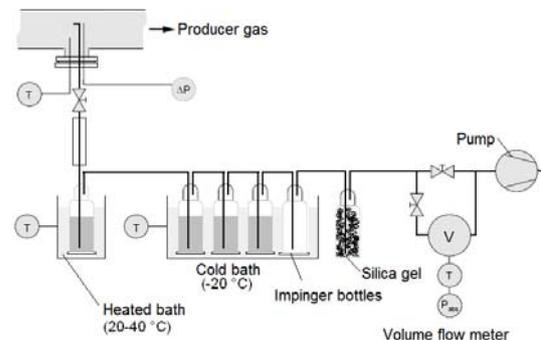
The control system of the experimental facility is based on combined load and combustion control. Load control is provided by regulation of the gasification air as the height of the fuel bed is held at a constant level. Combustion control is achieved by the regulation of secondary and tertiary air feeds guided by the O<sub>2</sub> concentration in the flue gas (lambda sensor). Therefore, all air flow rates are controlled by means of automatic valves.

#### 3.2 Measurement and analyses methods applied

A significant number of different measurement and sampling technologies was applied in order to gain detailed information about the gasification process and the nitrogen conversion. To define the performance of the facility, characteristic process data such as air flow rates, important temperatures and boiler load were recorded continuously.

The flow rates of the three air streams (primary, secondary and tertiary air) have been measured by means of flow sensors in order to calculate the air to fuel ratio of the gasifier, the stoichiometric air ratio in the reducing zone ( $\lambda_{red}$ ) of the combustion chamber and the overall stoichiometric air ratio ( $\lambda_{tot}$ ) of the entire experimental facility

The tar content of the producer gas was measured using a gravimetric method (see Fig. 2) according to [8]. Samples of the producer gas are drawn through the impinger bottles that are filled with isopropanol as solvent and cooled down to 253 K. Heavy tars are collected in the bottles while light gases pass through. The overall volume flow during each sampling campaign was measured by means of a volume flow meter. All samples have been dried in a vacuum drier till all solvent is evaporated.

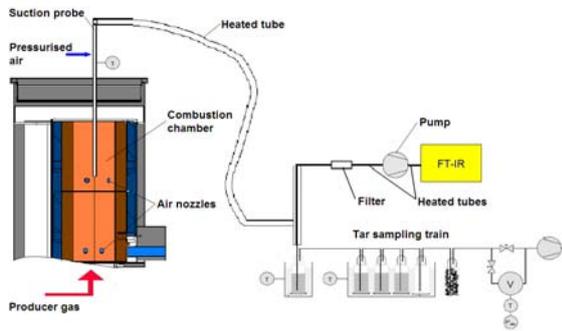


**Figure 2:** Tar sampling train

The remaining tars were weighted and by means of the recorded volume flow the tar content of the flue gas could be estimated. The composition of the tars (C, H and N) was measured using an elemental analyser. The O content was calculated by difference.

The composition of the flue gas in the reducing zone of the combustion chamber was determined regarding HCN, NH<sub>3</sub>, NO, NO<sub>2</sub> and tar content. Therefore, samples of the flue gas have been taken at different heights in the middle axis of the combustion zone by means of a cooled suction probe, which was injected from the top of the combustion chamber and coupled to the tar sampling train for measuring the tar content and a FT-IR in order to quantify NO, HCN, NH<sub>3</sub> and NO<sub>2</sub> (see Fig. 3). The suction probe has been cooled by means of pressurized air in order to

inhibit chemical reactions of the flue gas species as well as the thermal decomposition of the tars inside the probe.



**Figure 3:** Scheme of the combustion chamber including measurement set-up for tar sampling and flue gas measurements in the reducing zone of combustion chamber

Explanations: suction tube cooled with pressurised air to approx. 673 – 873 K

Moreover, the composition of the flue gas at boiler outlet was continuously measured by using standard flue gas analysers for CO (NDIR), CO<sub>2</sub> (NDIR) and NO<sub>x</sub> (CLD).

Standardized softwood pellets (EN 14961-2 A1) were used for the test runs (see Table I). The pellets have been analysed concerning the following parameters:

- moisture content: determination of weight loss at 105 °C (ÖNORM EN 14774-1:2009 12 01)
- ash content: method according to ÖNORM EN 14775:2009 12 15
- C, H, N: elemental analyser (ÖNORM EN 15104:2001 04 01)

**Table I:** Chemical characterisation of wood pellets

Explanations: GCV ... gross calorific value; NCV ... net calorific value

	Units	Pellets (softwood)
moisture content	g kg <sup>-1</sup> w.b.	69.0
C	g kg <sup>-1</sup> w.b.	493.0
H	g kg <sup>-1</sup> w.b.	62.0
N	g kg <sup>-1</sup> w.b.	0.5-0.63
Ash content	g kg <sup>-1</sup> w.b.	3.7
GCV	kJ kg <sup>-1</sup> w.b.	19,920
NCV	kJ kg <sup>-1</sup> w.b.	17,110

#### 4 RESULTS

In this section the results of the measurements performed with the experimental facility are summarised. All experiments and measurements have been performed under stationary conditions of the experimental facility. Experiments have been performed for different loads by changing the primary air flow rate.

Several test runs have been performed in order to investigate the release and the conversion of the fuel-bound nitrogen. As already mentioned before, an air staging concept was developed for the combustion chamber based on CFD-modelling in order to reduce the NO<sub>x</sub> emissions in the flue gas. Therefore, the concentrations of relevant nitrogenous gas species including tars have been measured in the producer gas and at different positions in the combustion chamber in

order to provide an insight into the single stages of the conversion process of the fuel-bound nitrogen.

However, due to the high tar content of the producer gas it was not possible to use the FT-IR for measurements in the producer gas. Therefore, based on the elemental analyses of the fuel and the tars mass balances for different operating conditions of the gasifier have been performed. The results, summarised in Tables II and III, show clearly that 58 to 68 wt% of the fuel-bound nitrogen is released as N bound in the tars from the packed bed. The remaining fuel-bound nitrogen is probably released as HCN and N<sub>2</sub> [10]. Moreover, the experiments showed that the concentrations of NH<sub>3</sub> in the reducing zone are significantly lower than the HCN concentrations (See Fig.4 and Fig.5).

The ratio of N in tars is the same range for 100% load and 33% load, although the tar content of the producer gas is higher for 100% load. However, the concentration of nitrogen in the producer gas increases with higher air to fuel ratios, corresponding to lower loads, which results in a dilution of the producer gas components.

These findings seem to be contradictory to other studies as HCN, NH<sub>3</sub> and N<sub>2</sub> have been identified to be the major nitrogenous components of the producer gas of gasifiers [3, 4 and 5]. However, these results have been derived from experiments performed with fluidised bed gasifiers.

**Table II:** Release of fuel-bound nitrogen during fixed-bed gasification (100% load)

Explanations: air to fuel ratio: 1.1; volume related to standard conditions (273K, 1013.25 mbar)

Test run	Units	1	2	3
Fuel rate	kg h <sup>-1</sup>	3.8	3.9	3.8
Nitrogen in fuel	g kg <sup>-1</sup> d.b.	0.5	0.6	0.6
Gasification air rate	m <sup>3</sup> h <sup>-1</sup>	3.9	3.9	3.7
Producer gas	m <sup>3</sup> h <sup>-1</sup> w.b.	6.9	6.6	6.7
Tar content	g m <sup>-3</sup> w.b.	136	165	159
Nitrogen content of tar	g kg <sup>-1</sup> d.b.	1.1	1.2	1.2
Nitrogen in fuel	g h <sup>-1</sup>	1.8	2.3	1.9
Nitrogen bound in tar	g h <sup>-1</sup>	1.0	1.3	1.3
Nitrogen ratio (tar/fuel)	%	58.3	58.4	66.4

As it is well known the tar content of the producer gas of fluidized bed gasifiers is significantly lower compared to updraft fixed-bed gasifiers. Therefore, the results of this study can not be compared with the reported results. The release of fuel-bound nitrogen during updraft fixed-bed gasification has not been investigated before.

**Table III:** Release of fuel-bound nitrogen during fixed-bed gasification (33% load)

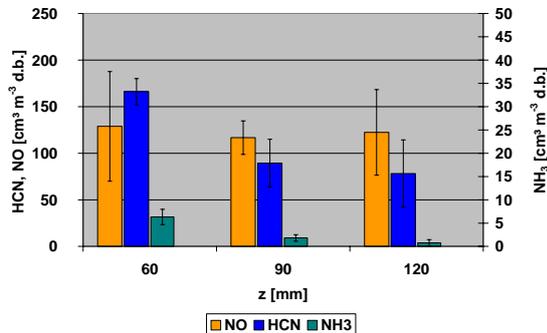
Explanations: air to fuel ratio: 1.6; volume related to standard conditions (273K, 1013.25 mbar)

Test run	Units	4	5	6
Fuel rate	kg h <sup>-1</sup>	1.2	1.3	1.3
Nitrogen in fuel	g kg <sup>-1</sup> d.b.	0.5	0.6	0.6
Gasification air rate	m <sup>3</sup> h <sup>-1</sup>	2.0	2.0	1.8
Producer gas	m <sup>3</sup> h <sup>-1</sup> w.b.	2.8	3.0	2.9
Tar content	g m <sup>-3</sup> w.b.	129	113	120
Nitrogen content of tar	g kg <sup>-1</sup> d.b.	1.2	1.3	1.2
Nitrogen in fuel	g h <sup>-1</sup>	0.6	0.7	0.6
Nitrogen bound in tar	g h <sup>-1</sup>	0.4	0.4	0.4
Nitrogen ratio (tar/fuel)	%	68.3	58.1	64.0

Fig. 4 and Fig. 5 show the results of measurements derived from the reducing zone of the combustion chamber. The oscillations of the calculated values result from the swirl flow of the flue gas in the combustion chamber as the presented results are the horizontal profiles of the species at the considered measuring point. The tar content of the flue gas, measured in the reducing zone of the combustion chamber, varied between 0.1 and 1 g Nm<sup>-3</sup> (d.b.) for the investigated test runs and is almost negligible compared to the high tar content of the producer gas (see Table II and Table III). The tars are cracked into lighter gases due to sufficiently high combustion temperatures (more than 1500 K) and the availability of oxygen introduced with the secondary combustion air. The results show that the tars exhibit high reactivity and that, due to the tar decomposition, the tar-bound nitrogen is released as HCN, NH<sub>3</sub>, NO and N<sub>2</sub> (see Fig. 4 and Fig. 5). The dominating N species are NO and HCN.

However, elemental analyses of the tars, sampled from the flue gas in the reducing zone of the combustion chamber, show that the nitrogen content of these tars is significantly higher than the N-content of the tars in the producer gas. This may indicate that the N containing tars exhibit a lower reactivity than N free tars.

In the oxidising zone of the combustion chamber the remaining combustion air is supplied and existing NH<sub>3</sub> and HCN get partly oxidised to NO and NO<sub>2</sub>. Some NH<sub>3</sub> and HCN is reduced to N<sub>2</sub>. The influence of different air to fuel ratios on the measured concentrations of NH<sub>3</sub> and HCN in the reducing zone of the combustion chamber is marginal for the investigated air to fuel ratios.

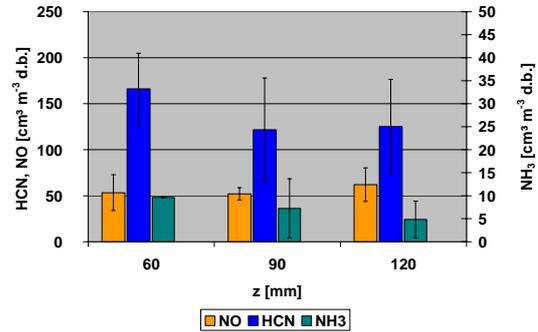


**Figure 4:** Measured NH<sub>3</sub>, HCN and NO concentrations at different positions in the reducing zone of the combustion chamber at 100% load

**Explanations:** measurement results from 3 test runs at stable load; z ... distance from secondary air nozzles; total height of reducing zone: 240 mm; air to fuel ratio: 1.1; N-content in the fuel: 0.5-0.63 [g kg<sup>-1</sup> d.b.];  $\lambda_{red}$ : 0.6;  $\lambda_{tot}$ : 1.2

There is a slight decrease of the NH<sub>3</sub>- and HCN-concentrations along the investigated positions as result of chemical conversion due to the high CO-content of the producer gas and the oxygen-deficient, fuel rich conditions in the reducing zone of the combustion chamber. On the contrary, Fig. 4 and Fig. 5 show that the NO-concentrations do not change significantly along the investigated positions. This can probably be attributed to two effects. On the one hand NO is reduced to N<sub>2</sub> due to the high CO-content of the producer gas and the oxygen-deficient, fuel rich conditions in the reducing zone of the combustion chamber. On the other hand NO is formed during conversion of HCN and NH<sub>3</sub> [7]. However, the exact pathway of the reactions involved is not yet really

understood. Further investigations are necessary in order to gain detailed information regarding this phenomenon.



**Figure 5:** Measured NH<sub>3</sub>, HCN and NO concentrations at different positions in the reducing zone of the combustion chamber at 33% load

**Explanations:** measurement results from 3 test runs at stable load; z ... distance from secondary air nozzles; total height of reducing zone: 250 mm; air to fuel ratio: 1.6; N-content in the fuel: 0.5-0.63 [g kg<sup>-1</sup> d.b.];  $\lambda_{red}$ : 0.6;  $\lambda_{tot}$ : 1.4

However, the NO concentration in the reducing zone of the combustion chamber and the NO<sub>x</sub> concentration in the flue gas at boiler outlet (see Table IV and Table V) are significantly smaller for 33% load. This can probably be attributed to different residence times and temperatures of the flue gas in the reducing zone of the combustion chamber as the reduction of the nitrogenous species to N<sub>2</sub> is a function of residence time and temperature [7].

The contribution of thermal NO<sub>x</sub> to the overall NO<sub>x</sub> emissions is supposed to be marginal as the flue gas temperature, repeatedly measured in the axial middle of the oxidising zone of the combustion chamber, did not exceed 1400 K. As it is well known NO<sub>x</sub> formation over the thermal NO<sub>x</sub> path becomes important at temperatures above 1800 K [7]. Almost complete burnout of the flue gas is achieved at the end of the combustion chamber which is indicated by very low CO emissions in the flue gas (see Table IV and Table V).

**Table IV:** Measured NO<sub>x</sub>, CO and O<sub>2</sub> concentrations in the flue gas at boiler outlet (100% load)

**Explanations:** N-content in the fuel: 0.5-0.63 [g kg<sup>-1</sup> d.b.]; mv ... measured mean values; s ... standard deviation; NO<sub>x</sub> and CO related to 13% O<sub>2</sub> and dry flue gas; NO<sub>x</sub> calculated as NO<sub>2</sub>

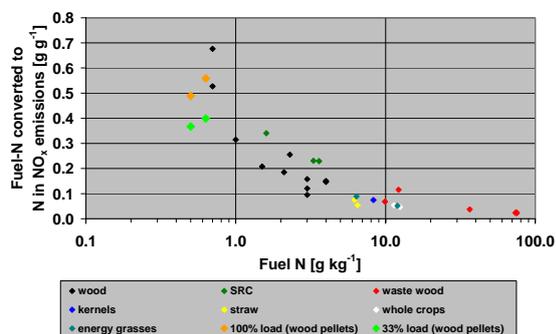
	Units	100% load	
		mv	s
NO <sub>x</sub>	mg m <sup>-3</sup>	76.9	8.8
CO	mg m <sup>-3</sup>	1.9	0.7
O <sub>2</sub>	volume fraction in % d.b.	3.9	-

**Table V:** Measured NO<sub>x</sub>, CO and O<sub>2</sub> concentrations in the flue gas at boiler outlet (33% load)

**Explanations:** N-content in the fuel: 0.5-0.63 [g kg<sup>-1</sup> d.b.]; mv ... measured mean values; s ... standard deviation; NO<sub>x</sub> and CO related to 13% O<sub>2</sub> and dry flue gas; NO<sub>x</sub> calculated as NO<sub>2</sub>

	Units	33% load	
		mv	s
NO <sub>x</sub>	mg m <sup>-3</sup>	55.1	13.0
CO	mg m <sup>-3</sup>	15.7	14.4
O <sub>2</sub>	volume fraction in % d.b.	5.7	-

In Fig. 6 the amount of fuel N converted to NO<sub>x</sub> versus the N content in the fuel is shown for different biomass fuels and for different loads of the experimental facility.



**Figure 6:** Fuel N converted to NO<sub>x</sub> in dependence of N content in the fuel for different biomass fuels and different loads of the experimental facility in comparison to conventional combustion plants equipped with air-staging

Explanations: data source: [9]

The figure clearly indicates that the N conversion rate increases with decreasing N content in the fuel. The rate of fuel-bound nitrogen converted to NO<sub>x</sub> during fixed-bed gasification and subsequent staged combustion varies between 49 and 56 wt% for 100% load as well as between 37 and 40 wt% for 33% load. Compared to conventional wood combustion systems equipped with air-staging the N conversion rate is rather similar at full load and smaller for 33% load during fixed-bed gasification and subsequent staged combustion.

## 5 CONCLUSIONS

Numerous experiments have been performed with a laboratory-scale updraft fixed-bed gasifier coupled to a combustion chamber in order to investigate the release behaviour and the conversion of the fuel-bound nitrogen. Test runs have been performed at different operating conditions.

The experimental results clearly show, that during updraft fixed-bed gasification 58 to 68 wt% of the fuel-bound nitrogen is released as N bound in the tars from the packed bed and is then subsequently released as HCN, NO, NH<sub>3</sub> and N<sub>2</sub> as a result of tar cracking in the reducing zone of the combustion chamber under lean-air (fuel-rich) conditions.

In the oxidising zone of the combustion chamber the remaining combustion air is supplied and existing NH<sub>3</sub> and HCN are partly oxidised to NO and NO<sub>2</sub>. Almost complete burnout of the flue gas is achieved which is indicated by very low CO emissions in the flue gas. NO-concentrations do not change significantly along the investigated positions in the reducing zone of the combustion chamber. This can probably be attributed to two effects. On the one hand NO is reduced to N<sub>2</sub> due to the high CO-content of the producer gas and the oxygen-deficient, fuel rich conditions in the reducing zone of the combustion chamber. On the other hand NO is formed during conversion of HCN and NH<sub>3</sub>. Further investigations in order to gain detailed information about the reaction mechanisms involved are needed. The exact conversion pathway of the fuel-bound nitrogen via N

bound in tars into nitrogenous gas species is still unknown, but the accumulation of fuel-bound N in the tars contained in the producer gas is of great interest. The findings concerning nitrogen release and subsequent conversion during updraft fixed-bed gasification and subsequent staged combustion of the producer gas are new and of great relevance for the design of low-NO<sub>x</sub> combustion chambers and their accurate modelling. In the next step, the experimental results will be implemented in an appropriate N release model as a basis for CFD based NO<sub>x</sub> formation simulations.

## 6 NOMENCLATURE

C	carbon content of fuel [g kg <sup>-1</sup> w.b.]
d.b.	dry base
GVC	gross calorific value [kJ kg <sup>-1</sup> d.b.]
H	hydrogen content of fuel [g kg <sup>-1</sup> w.b.]
N	nitrogen content of fuel [g kg <sup>-1</sup> w.b.]
NCV	net calorific value [kJ kg <sup>-1</sup> w.b.]
w.b.	wet base

### Greek letters

$\lambda_{red}$	stoichiometric air ratio in reducing zone
$\lambda_{tot}$	overall stoichiometric air ratio

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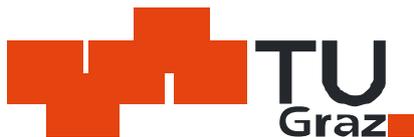
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## 9 LOGO SPACE





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Power Production from Biomass



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