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Characterisation of fuel bound nitrogen in the gasification process and the staged combustion of producer gas from the updraft gasification of softwood pellets

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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 characterisation of fixed-bed gasifier operation and to investigate the release behaviour and the conversion of fuel-bound nitrogen during gasification and subsequent staged combustion of the producer gas using softwood pellets. Spatial temperature profiles and the composition of the producer gas of the gasifier have been measured for different air flow rates. In addition, the concentrations of relevant nitrogenous gas species including tars have been measured in the producer gas and at different positions in the combustion chamber. The air flow rate has a significant influence on the composition of the producer gas and the temperature profile of the packed bed of the gasifier. Results show that during updraft fixed bed gasification almost the entire fuel-bound nitrogen is released as N bound in tars from the packed bed and is then subsequently released as HCN, NO, NH₃ and N₂ as a result of tar cracking during combustion. This strong N-fixation in the tars was not expected and is of great relevance concerning NO_x formation during combustion of the producer gas.

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1. Introduction

The gasification of renewable solid biomass to produce CO₂neutral fuels for heat and electricity production is still in the development stage. Amongst the thermo-chemical conversion technologies, biomass gasification has attracted a high interest as it may offer higher efficiencies in relation to combustion. Softwood pellets are presently used in small-scale residential combustion units. As the market is expected to further increase within the next years, pellets may also be used in small-scale fixed-bed gasifiers for heating purposes as well as regarding micro-CHP applications (e.g. Stirling engine or micro-turbine) in the near future.

Due to the high tar content of the producer gas (up to 150 g m^{-3}), updraft gasifiers are not suitable for engines and gas turbines without comprehensive gas cleaning [1]. Therefore, updraft gasifiers are primarily used for heat production. The updraft gasifier has a high thermal efficiency as the sensible heat of the producer gas is recovered by direct heat exchange with the entering feedstock, which is dried, and pyrolysed before entering the gasification zone. The dust content in the producer gas is usually low due to low gas velocities and the filtering effect of the packed bed [2,3]. In

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addition to the high tar content of the producer gas the main drawback of the process is that high combustion temperatures above the grate can cause blocking due to ash fusion.

The scope of this work was the experimental investigation of a laboratory scale fixed-bed updraft gasifier coupled to a combustion chamber for heat production, firstly with respect to temperature profiles of the fuel bed and the composition of the producer gas.

The temperature profile of the fuel bed is relevant for problems related to condensation of tars and ash fusion. The composition of the producer gas is required for the validation of models developed for updraft gasifiers [4]. Small-scale updraft fixed-bed gasifiers have already been investigated by several authors [2,5,6] before. However, focusing on the gasification of softwood pellets, detailed experimental data could only be retrieved from Lucas [2].

Furthermore, the release of fuel-bound nitrogen has been investigated. During gasification of biomass fuels, nitrogen compounds, such as HCN, NH₃, and nitrogen oxides (NO, NO₂ and N_2O are released from the fuel to the gas phase [7]. Several authors in dentified HCN, NH₃ and N₂ to be the major nitrogenous components of the producer gas of gasifiers [7–9]. 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_x, 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 fuelbound nitrogen is of great relevance for a low-NO_x combustion of the producer gas.

This paper presents the experimental results from the 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 fixedbed 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. The char is partially converted into producer gas mainly by the following endothermic reactions (Eq. (1)-(4)) [10]:

$$C + CO_2 \leftrightarrow 2CO$$
 (1)

$$C + H_2 O \leftrightarrow CO + H_2$$
(2)

$$C + 2H_2 \leftrightarrow CH_4$$
 (3)

$$CH_4 + H_2O \rightarrow CO + 3H_2O \tag{4}$$

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_{2} , CO_{2} , CH_{4} , 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_3 and nitrogen oxides (NO, NO_2 and N_2O) from the bed [11]. Several authors identified HCN, NH_3 and N_2 as the major nitrogenous components of the producer gas of gasifiers [7–9]. 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_x 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 [12]. 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 gasification process, as well as the release and conversion of 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 insulated 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_x 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.

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_2 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 temperature profile of the packed bed inside the gasifier was measured by a set of thermocouples (Type K).

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 reducing zone (λ_{red}) of the combustion chamber and the overall stoichiometric air ratio (λ_{tot}) of the entire experimental facility.

Samples of the producer gas have been repeatedly taken at the outlet of the gasifier by means of gas collection tubes (see Fig. 2). The samples have been cooled down to ambient temperature to remove tars. The composition of the producer gas (CO, CO_2 , H_2 , CH_4 , O_2 and N_2) was then measured by a gas chromatograph (GC) equipped with a thermal conductivity analyser. The tar content of the producer gas was measured using a gravimetric method (see Fig. 3) according to [13].

Samples of the producer gas are drawn through the impinger bottles that are filled with isopropanol as solvent and cooled down to minus 20 °C. 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.

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

The major gaseous nitrogenous species, HCN, NH_3 and NO_x , were quantified either online or by off-line analysis of extracted gas samples. A nitrogen species gas sampling system (see Fig. 4) was installed in order to quantify NH_3 and HCN in the producer gas.

The sampling train consists of 2 series arranged in parallel, each with 3 impinger bottles. The first two bottles are filled either with H_2SO_4 (0.1 mol dm⁻³), used to trap NH₃, or with Na₂CO₃ (1.0 mol dm⁻³) to absorb HCN. The last bottle of each series is filled with silica gel in order to remove moisture. The overall volume flow during each sampling campaign was measured by means of volume flow meters. The NH₃– and HCN-concentration in each sample was then measured with ion chromatography. Due to the high tar content of the producer gas, but it can be assumed that the NO and NO₂ concentrations in the producer gas are negligible.



Fig. 2 – Method for sampling of producer gas.



Fig. 3 – Tar sampling train.

Moreover, the composition of the flue gas in the reducing zone of the combustion chamber was determined regarding HCN, NH_3 , NO, NO_2 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 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 an FT-IR in order to quantify NO, HCN, NH_3 and NO_2 .

Moreover, the composition of the flue gas at boiler outlet was continuously measured by using standard flue gas analysers for CO (NDIR), CO_2 (NDIR) and NO_x (CLD).

Standardized softwood pellets (ÖNORM M 7135) were used for the test runs (see Table 1). The pellets have been analysed concerning the following parameters:

- moisture content: determination of weight loss at 105 °C
- ash content: method according to prCEN/TS 14775
- C, H, N: elemental analyser

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. Firstly, characteristic process data of the updraft gasifier are presented in order to provide an



Fig. 4 – Method for sampling NH_3 and HCN in the producer gas.

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Table 1 – Chemical characterisation of wood pellets.			
	units	pellets (softwood)	
moisture content	g kg w.b.	60.0	
С	$g kg^{-1} d.b.$	487.0	
Н	$g kg^{-1} d.b.$	62.0	
Ν	$g kg^{-1} d.b.$	0.5-0.63	
ash content	$g kg^{-1} d.b.$	3.6	
GCV	$kJ kg^{-1} d.b.$	19,676	
NCV	kJ kg ⁻¹ w.b.	17,060	
Explanations: GCV g value.	ross calorific value; NC	CV net calorific	

overview over the gasification process. Then, the composition of the producer gas including the main gas phase species and tars are given for different loads (different air flow rates). Finally, the release and the conversion of the fuel-bound nitrogen are discussed in detail.

4.1. Process data of the updraft gasifier

The major process data of the updraft gasifier at stable load conditions are summarised in Table 2. Experiments have been performed for different air flow rates, presented for 33% and 100% load related to the boiler capacity of the experimental facility, respectively.

The temperature profiles at several positions of the gasifier axis for 33% and 100% load are shown in Fig. 5. It can be seen that with higher air and fuel rates the temperatures along the packed bed increase, which is also indicated by different producer gas outlet temperatures (see Table 3). In fixed bed gasification the temperature profiles are representative for the different reaction zones, stratified along the reactor height. Due to the fact, that overlapping of processes occurs, it is not possible to exactly assign the boundaries between the different zones by means of temperature measurements. However, some considerations can be made based on characteristic temperatures and existing gradients. The high temperatures above the grate are caused by char combustion. Values up to 1480 K have been observed during experiments (see Fig. 6). Due to the high combustion temperatures, the gasifier may cause problems due to ash fusion, especially when biomass fuels with low ash melting temperature (e.g. herbaceous fuels) are used [14].

As a result of the high temperatures in the lower part of the packed bed, gasification reactions of char start, which is indicated by the temperature decrease. It seems that there is

Table 2 – Operating conditions of the gasifier.			
	units	100% load	33% load
fuel rate primary air rate bulk density of fuel air to fuel ratio height of fuel bed	$\begin{array}{c} kg \ h^{-1} \ w.b. \\ kg \ h^{-1} \ w.b. \\ kg \ m^{-3} \ w.b. \\ kg \ kg^{-1} \\ m \end{array}$	3.70 4.10 650 1.1 0.42	1.43 2.27 650 1.6 0.42



Fig. 5 – Temperature profiles of the gasifier at different loads: mean values (10 min).

a long zone, situated in the middle of the gasifier, where the predominant process is heating of the char bed by the hot gases coming from the bottom of the gasifier indicated by almost constant temperature gradients [4,5]. The spatial temperature gradients are higher for 33% load, which can be attributed to the lower convective flow and the lower combustion temperatures.

The outlet temperature of the producer gas varies between 413 K for 33% load and 478 K for 100% load. The fuel is fed at ambient temperature to the gasifier. Due to the existing difference between the temperature of the solid and the gas phase, evaporation of fuel moisture is assumed to take place instantaneously. The absence of steep gradients in the upper region of the packed bed excludes the existence of a well defined pyrolysis zone, as it would be in case of an abrupt decomposition of the fuel. With increasing air flow rate reaction temperatures in the pyrolysis zone increase, resulting in higher heating rates.

With means of characteristic pyrolysis temperatures and the existing gradients, an average residence time for the solid fuel in the pyrolysis zone can be estimated. The length of the pyrolysis zone is defined by two characteristic temperatures of the packed bed. At about 500 K pyrolysis of the fuel starts, which is indicated by an increasing temperature gradient in

Table 3 – Results from test runs (100% load) performed.				
test run	units	1	2	3
CO	volume fraction in %	26.1	24.4	26.6
CO ₂	volume fraction in %	6.2	4.7	5.0
CH ₄	volume fraction in %	1.7	1.6	1.7
H ₂	volume fraction in %	3.9	5.0	6.3
H ₂ O	volume fraction in %	14.6	18.4	15.2
N ₂	volume fraction in %	42.5	41.4	40.4
tar	$\mathrm{g}\mathrm{m}^{-3}\mathrm{w.b.}$	210	186	199
cold gas efficiency	%	85.4	81.7	88.3
hot gas efficiency	%	92.5	88.7	95.3

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



Fig. 6 - Temperature profiles of the gasifier at 100% load: T0.01 and T0.05 ... distance from grate: 0.01 and 0.05 m.

the packed bed (bed height approx. 0.35 m). As the pyrolysis of the fuel is complete, the temperature gradients in the packed bed decrease significantly (see Fig. 5). At a bed height of approx. 0.25 m temperature gradients reach very small values. The temperature of the packed bed at this point is the second characteristic pyrolysis temperature - 700 K at 33% load and 850 K at 100% load. The superficial velocity of the pellets in bulk along the drying and pyrolysis is assumed to be constant and is calculated from the initial bulk density of the fuel, the fuel feed rate and the diameter of the gasifier. Based on these assumptions, the residence times of the fuel vary between 16 (100% load) and 50 min resulting in heating rates varying from 0.1 to 0.5 K/s (100% load). Therefore, it can be stated, that the pyrolysis in a fixed-bed updraft gasifier takes place under conditions described as slow pyrolysis. These results show good agreement with experimental data derived from a labscale updraft gasifier [5]. As a result of different heating rates in the pyrolysis zone for different loads, the equivalence ratio, as shown in Table 2, is higher for 33% load for a constant bed height. This can be attributed to the higher char ratio, which is, as well known, higher for smaller heating rates [15].

can cause ash sintering, especially when no ash is discharged

High temperatures in the combustion zone of the gasifier emperature [K]

Fig. 7 – Ash agglomeration in the gasifier.

from the grate as it has been observed during test runs (see Fig. 6). Due to the growing ash bed, as no ash has been discharged from the grate, and the high combustion temperatures up to 1480 K, resulting in ash sintering and agglomeration, the combustion zone moved quickly upward and the temperature, which has been measured above the grate, decreased continuously. Due to unstable operating conditions caused by non-uniform air distribution, the experimental facility had to be shut down in order to remove agglomerated ash from the gasifier (see Fig. 7).

Therefore, ash must be discharged from the grate periodically, in order to maintain a stable gasification process, even when biomass fuels with low ash content like softwood pellets are used.

The influences of a growing ash layer and ash discharges at regular intervals on the temperature profile of the packed bed of the gasifier are shown in Fig. 8. As a result of the growing ash bed the temperature, measured in the ash bed, decreased significantly. The influence on the temperatures in the upper regions of the fuel bed was considerably smaller.

After an operation time of 24 h ash was discharged from the grate and the temperature, measured in the ash bed, increased immediately. Steady operation conditions could be maintained, indicated by stable temperature profiles in the upper region of the packed bed of the gasifier. After further 24 h of operation ash was discharged again as the temperature of the ash bed decreased below 500 K and the temperature of the ash bed increased again. The results of the test run performed showed that the ash discharge can be operated depending on the temperature of the ash bed. Following this approach, a continuous and stable operation of the gasifier can be provided.

4.2. Producer gas composition

The influence of different air to fuel ratios on the composition of the producer gas is illustrated in Fig. 9. A decrease of the air flow rate, resulting in a higher air to fuel ratio, produces higher values of CO2 and corresponding lower values of CO. The decreasing ratio of CO to CO₂ with increasing air to fuel ratio can be attributed to different temperatures in the combustion and gasification zone of the gasifier. Higher temperatures



Fig. 8 - Temperature profiles of the gasifier at 100% load. Explanations: T0.4 ... distance from grate: 0.4 m.



Fig. 9 – Composition and NCV of the producer: mean values (Table 3).

favour the production of CO from char combustion according to the Boudouard equilibrium and accelerate the reduction of CO_2 in the gasification zone.

Furthermore, the concentration of nitrogen in the producer gas increases with higher air to fuel ratios which results in a dilution of the other components and a lower efficiency of the gasification process (see Tables 3 and 4). This may conclude that the gasification efficiency increases with the air flow rate and consequently with the power input. Furthermore, the production of condensable liquids (tars and water) decreases with an increasing air to fuel ratio (see Tables 3 and 4).

Due to higher temperatures and heating rates in the pyrolysis zone corresponding to smaller air to fuel ratios the amount of liquids produced from pyrolysis is higher, mainly at the expense of char [5,15]. In addition, the temperature of the packed bed decreases with higher air to fuel ratios also indicated by lower outlet temperatures of the producer gas.

Due to the high tar content and the low outlet temperature of the producer gas condensation of tars may occur if the gasifier is not well isolated, particularly in case of operation of the gasifier at low fuel rates, as it was observed during experiments. Therefore, operation of the gasifier at higher fuel rates should be favoured in order to maintain stable operating conditions.

Table 4 – Results from test runs (33% load) performed.				
test run	units	4	5	6
CO	volume fraction in %	20.0	18.9	18.8
CO ₂	volume fraction in %	8.6	9.7	8.6
CH_4	volume fraction in %	1.6	1.8	1.7
H ₂	volume fraction in %	3.4	5.2	4.5
H ₂ O	volume fraction in %	13.8	13.1	14.3
N ₂	volume fraction in %	48.8	48.3	48.3
tar	$g m^{-3} w.b.$	157	126	164
cold gas efficiency	%	81.9	78.8	84.3
hot gas efficiency	%	87.6	84.6	90.0

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

Gas sampling and analyses have been performed under steady operating conditions of the gasifier, but some oscillations have been present during the measurements resulting in unsteady results regarding the composition and the tar content of the producer gas. The oscillations may be attributed to discontinuous feeding of the fuel to the gasifier resulting in unsteady moisture evaporation and decomposition of the solid fuel.

4.3. Fuel-bound nitrogen

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_x 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, preliminary experiments showed that the concentrations of NH_3 and HCN in the producer gas are below $1 \text{ cm}^3 \text{ m}^{-3}$. 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 5 and 6, show clearly that almost the entire 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_2 .

The ratio of N in tars is slightly higher for 100% load compared to 33% load. This can be attributed to the higher tar content of the producer gas at higher power outputs of the gasifier. These findings seem to be contradictory to other studies as HCN, NH_3 and N_2 have been identified to be the major nitrogenous components of the producer gas of gasifiers [7–9].

However, these results have been derived from experiments performed with fluidised bed gasifiers. 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

Table 5 – Release of fuel-bound nitrogen during fixed-bed gasification (100% load).				
test run		1	2	3
fuel rate nitrogen content fuel gasification air rate producer gas tar content nitrogen content of tar nitrogen in fuel nitrogen bound in tar pitrogen ratio (tar/fuel)	$\begin{array}{c} kg \ h^{-1} \\ g \ kg^{-1} \ d.b. \\ m^3 \ h^{-1} \\ m^3 \ h^{-1} \ w.b. \\ g \ m^{-3} \ w.b. \\ g \ kg^{-1} \ d.b. \\ g \ h^{-1} \\ g \ h^{-1} \\ \end{array}$	3.7 0.60 3.2 6.0 210 1.47 2.1 1.8 88 8	3.7 0.60 3.2 6.3 186 1.57 2.1 1.8 86 6	3.7 0.60 3.1 6.2 199 1.62 2.1 2.0 94.1
0 1 (1 1 1 7				

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

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Table 6 – Release of fuel-bound nitrogen during fixed-bed gasification (33% load).				
test run		4	5	6
fuel rate nitrogen content fuel gasification air rate producer gas tar content nitrogen content tar nitrogen bound to fuel nitrogen bound to tar	$\begin{array}{c} kg \ h^{-1} \\ g \ kg^{-1} \ d.b. \\ m^3 \ h^{-1} \\ m^3 \ h^{-1} \\ w.b. \\ g \ m^{-3} \\ w.b. \\ g \ kg^{-1} \\ d.b. \\ g \ h^{-1} \\ g \ h^{-1} \end{array}$	1.4 0.60 1.7 2.8 153 1.50 0.8 0.6	1.4 0.60 1.8 3.0 123 1.70 0.8 0.6	1.5 0.60 1.8 3.1 160 1.50 0.8 0.7
Explanations: air to fuel conditions (273 K, 1013.25	% ratio: 1.6; vc 5 mbar).	84.0 olume rela	79.2 ated to s	88.3 tandard

compared with the reported results. The release of fuel-bound nitrogen during updraft fixed-bed gasification has not been investigated before.

Fig. 10 and Fig. 11 show the results of measurements derived from the reducing and oxidizing 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 m⁻³ (d.b.,volume related to 273 K and 1013.25 mbar) for the investigated test runs and is almost negligible compared to the high tar content of the producer gas. The tars are cracked into lighter gases due to sufficiently high combustion temperatures of about 900 °C 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



Fig. 10 – Measured NH₃, HCN and NO concentrations at different positions in the reducing zone of the combustion chamber. 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 fuel: 0.5–0.63 [g kg⁻¹ d.b.]; λ red: 0.6; λ tot: 1.2. For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.



Fig. 11 – Measured NH₃, HCN and NOx 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 fuel: 0.5–0.63 [g kg⁻¹ d.b.]; λ red: 0.6; λ tot: 1.4. For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.

tar decomposition, the tar-bound nitrogen is released as HCN, NH_3 , NO and N_2 (see Figs. 10 and 11).

In the oxidising zone of the combustion chamber the remaining combustion air is supplied and existing NH_3 and HCN get partly oxidised to NO and NO_2 . Some NH_3 and HCN is reduced to N_2 . The influence of different air to fuel ratios on the measured concentrations of NH_3 and HCN in the reducing zone of the combustion chamber is marginal for the investigated air to fuel ratios.

There is a slight decrease of the NH_3 - and HCNconcentrations 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, Figs. 10 and 11 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_2 due to the high CO-content of the producer

Table flue g	e 7 — Measured NO _x , CO a gas at boiler outlet.	nd O ₂ c	oncent	rations	in the
		100%	load	33%	load
	units	mv	S	mv	S
NO _x CO	$mg m^{-3}$ $mg m^{-3}$	76.9 1.9	8.8 0.7	55.1 15.7	13.0 14.4
O ₂	volume fraction in % d.b.	3.9	_	5.7	-

Explanations: N-content fuel: 0.5–0.63 [g kg⁻¹ d.b.]; mv ... measured mean values; s ... standard deviation; NO_x and CO related to 13% O₂ and dry flue gas.



Fig. 12 – Fuel N converted to NO_x in dependence of N content in the fuel for different biomass fuels and different loads of experimental facility in comparison to conventional combustion plants equipped with air-staging Explanations: data source: [14].

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_3 [12]. 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.

However, the NO concentration in the reducing zone of the combustion chamber and the NO_x concentration in the flue gas at boiler outlet (see Table 7) 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_2 is a function of residence time and temperature [12].

The contribution of thermal NO_x to the overall NO_x 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_x formation over the thermal NO_x path becomes important at temperatures above 1800 K [12].

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 7).

In Fig. 12 the amount of fuel N converted to NO_x versus the N content in the fuel is shown for different biomass fuels and for different loads of the experimental facility.

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_x 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 with basic air staging concept 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 gasification process as well as the release behaviour and the conversion of the fuelbound nitrogen. Test runs have been performed at different operating conditions.

In fixed-bed gasification the temperature profiles are representative for the different reaction zones, stratified along the reactor height. Due to high temperatures above the grate, as a result of char combustion, the gasifier may cause problems due to ash fusion, especially when biomass fuels with low ash melting temperatures (e.g. herbaceous fuels) are used. Temperature profiles indicate that pyrolysis takes place in the upper region of the packed bed with heating rates corresponding to slow pyrolysis. Furthermore, higher fuel rates and lower air to fuel ratios increase the temperature along the fuel bed, which is also indicated by increasing producer gas outlet temperatures.

Test runs showed that high temperatures in the combustion zone of the gasifier can cause ash sintering, especially when no ash is discharged from the grate. Thus, ash discharge from the grate should take place periodically in order to maintain a stable operation of the gasifier. The results of the test runs performed showed, that the ash discharge can be operated depending on the temperature of the ash bed. Following this approach, a continuous stable operation of the gasifier can be achieved.

There is a significant effect of the air flow rate on the composition of the producer gas. As it is increased, the air to fuel ratio decreases and CO increases due to improved char gasification and lower air to fuel ratios with a corresponding increase of the heating value of the producer gas. This may conclude that the gasification efficiency increases with the air flow rate and consequently with the power input.

Due to the high tar content and the moderate outlet temperature of the producer gas, condensation of tars may occur if the gasifier is not well insulated, particularly in case of operation of the gasifier at low air flow rates.

Furthermore, the release behaviour and the conversion of the fuel-bound nitrogen have been investigated carefully. The experimental results clearly show, that during updraft fixedbed gasification almost the majority of fuel-bound nitrogen is released as N bound in the tars from the packed bed and is then subsequently released as HCN, NO, NH_3 and N_2 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_3 and HCN are partly oxidised to NO and NO_2 . 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

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 N_2 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_3 . Further investigations in order to gain detailed information about the reaction mechanisms involved are needed. 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_x combustion chambers and their accurate modelling.

Nomenclature

С	carbon content of fuel [wt.% d.b.]
d.b.	dry base
GVC	cross calorific value [kJ/kg d.b]
Н	hydrogen content of fuel [wt.% d.b.]
Ν	nitrogen content of fuel [wt.% d.b.]
NCV	net calorific value [kJ/kg w.b.]
OXZ	oxidising zone of combustion chamber
RZ	reducing zone of combustion chamber
w.b.	wet base
Greek lett	ers

λ_{red}	stoichiometric air ratio in reduction zone
λ_{tot}	overall stoichiometric air ratio

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