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Strategies and technologies towards zero emission biomass combustion by primary measures

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Abstract

To satisfy the market demand for fuel-flexible low-emission biomass combustion systems a new approach based on an updraft gasifier directly coupled with a multi-stage gas burner and a boiler has been developed. In the gasifier semi-volatile ash forming elements (i.e. mainly K) are released in the hot charcoal combustion zone and are precipitated again in the upper regions of the fuel bed. Therefore, an almost particle free product gas can be achieved which can then be efficiently combusted at low excess air ratios in the subsequent multi-stage burner. In this paper the principle of ash embedding in the gasifier is described with a special focus on the K chemistry. Combining this approach with appropriate fuel bed cooling measures, the new technology can be applied for a wide spectrum of biomass fuels reaching from conventional wood fuels over short rotation coppice to agricultural fuels/residues such as miscanthus and olive stones. Test runs performed at a lab-scale testing plant confirmed this high fuel flexibility and the possibility to operate this system at almost zero CO and OGC emissions as well as dust emissions of less than 5 mg/MJ_{NCV}.

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1. Introduction

During the last three decades a steep further development of biomass combustion systems towards low emissions and high efficiencies has been achieved. In the residential heating sector for instance, according to type tests performed

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with wood chip boilers at FJ-BLT [1], CO emissions were decreased from more than 10,000 mg/MJ in the 1980ies to less than 50 mg/MJ. At the same time the boiler efficiency could be increased from about 76% to about 92%. The same trend regarding emission reduction could also be observed for the medium-scale capacity range up to 10 MW_{th}.

One drawback of present systems is their restricted fuel flexibility. If a plant is operated with other fuels than the design fuels, elevated gaseous and particulate emissions and in many cases plant internal ash related problems (deposit formation, slagging) occur. These problems are more pronounced if ash, N, S and Cl-rich agricultural fuels are utilised. In the medium capacity range emission related problems can be overcome by the application of costly secondary measures like filters while in the small capacity range the application of secondary measures is not economically meaningful. Therefore, present residential biomass heating boilers almost exclusively utilise wood fuels (log wood, high quality pellets according to EN ISO 17225-2-A1 and wood chips with moisture contents up to 35 wt% w.b.). In the medium capacity range a higher flexibility regarding fuel type, ash and moisture content is given, however, there are still no technologies available in which wood fuels and agricultural residues can be applied without major adaptations to the respective fuel.

To gain a higher flexibility regarding fuel purchase and thereby to reduce fuel costs, the market clearly demands for fuel flexible systems. To make such system economically feasible they should ideally control emissions by primary measures. Since such systems are presently not available a new highly fuel-flexible low-emission approach for biomass combustion in the capacity range up to 10 MW_{th} has been developed and is presented in the paper.

2. Technological approach

For state-of-the-art biomass combustion technologies no significant further improvements towards lower emissions and significantly enhanced fuel flexibility are possible. Therefore, a new approach has been selected for the technology to be presented. The approach is based on an updraft gasifier, which is directly coupled with a multi-stage gas burner and a boiler. The updraft gasifier thereby provides the basis for a fuel flexible low-dust operation.

In updraft gasifiers the fuel is fed from above to a fuel bed and the gasification air passes upwards trough the fuel bed. Thereby, zones with different conversion processes and consequently a typical pronounced temperature profile over the fuel bed are established. In the bottom (grate near) zone of the fuel bed, charcoal combustion takes place at temperatures above 1,000°C. The gas then passes in counter-current through the fuel bed and heats it up while getting cooled down in the gasification/pyrolysis zone. At the top of the bed drying of the fuel with the product gas takes place. Depending on the moisture content of the fuel, the product gas leaves the fuel bed with about 100°C (for moist fuels) to more than 400°C (for dry fuels).

As it also happens in fixed-bed combustion systems, semi-volatile ash forming elements are released to the gas phase at high temperatures in the charcoal combustion zone. Later they undergo gas phase reactions and form submicron particles by nucleation and condensation. Moreover, they contribute with a major share to deposit formation on heat exchanger surfaces by direct condensation. For the combustion of chemically untreated biomass fuels these mechanisms are clearly dominated by K. In contrast with combustion systems, where K-components almost directly leave the fuel bed with the flue gas, in updraft gasifiers they have to pass with the product gas through the whole fuel bed and react with the char. Thereby, they are embedded in the ashes and consequently the product gas leaving the fuel bed is very poor in condensable ash forming elements, and thus fine particulate matter emissions and ash deposit formation can be substantially reduced.

Moreover, the gas volume flow in an updraft gasifier is, compared to a fixed-bed combustion system, very low and therefore, gas velocities at the fuel bed surface of below 1 m/s can be achieved. This leads to a very low entrainment of fuel and char particles with the product gas and consequently to, compared with fixed-bed combustion systems, one to three orders of magnitude smaller coarse fly ash emissions.

One well known drawback of updraft gasifiers are the high contents of tars in the product gas. The tars have to be removed when the gas shall be utilised in a combustion engine for instance. However, if the gas is directly fed to a gas burner, as in the approach presented, they are used as fuel.

The product gas leaving the fuel bed is combusted in a multi-stage gas burner directly coupled with the gasifier. With such burners even at very low excess air ratios (around 1.2) an almost complete gas burnout regarding CO and organic compounds can be reached and due to the possibility to apply flue gas recirculation and multiple air staging,

also the NO_x emissions can be kept low. The direct coupling of the gasifier and the multi-stage burner as well as the grate design, which are key issues regarding the new technological approach, have already been patented.

3. Objectives

During the development of the new combustion approach it turned out that the behaviour of K determines both, the low dust emissions of the system and its operational stability due to the risk of ash melting in the fuel bed. Therefore, one major objective of the work presented was to investigate the K-behaviour in the gasifier when using different biomass fuels. Moreover, it was the aim to investigate the gaseous and particulate emissions in order to prove the potential of the new technology regarding highly fuel flexible biomass combustion at ultra-low emissions, which represents the achievement of a new technological milestone.

4. Methods

Based on lab-tests as well as on simulations with a model for updraft gasifiers [2], [3], a geometric concept for the gasifier has been developed. Moreover, a gas burner has been designed and optimised by CFD (Computational Fluid Dynamics) simulations. The aim was to find a burner configuration (geometry, number and orientation of combustion air injection points) which enables an almost complete burnout and therefore almost zero CO, OGC and organic PM emissions at very low excess air ratios (high efficiencies). In order to carry out the CFD simulations a special code considering the conversion of tars released from the fuel bed has been developed and applied [4]. It includes an empirical in-house fuel conversion model for fixed beds [5], the realizable k-ɛ-turbulence model, a Discrete Ordinates Model for radiation and an Eddy Dissipation Concept (EDC) using a reduced detailed gas phase combustion mechanism. To consider tar conversion, an extension of the gas-phase-model with nitrogen-free and nitrogen containing tars, each with oxidation and thermal decomposition reactions has been developed [4]. Figure shows a scheme of the lab-scale gasifier and the burner as well as results from the CFD simulations regarding CO burnout.



Figure 1. Scheme of the lab-scale plant (left) and results of CFD simulations of the gas burner (Explanations: Iso-surfaces of the oxygen concentration [m³ O₂/m³ w.b.] in several horizontal planes through the combustion chamber (middle); Iso-surfaces of CO concentrations [ppmV] in the vertical cross-section (right); fuel: softwood pellets; boiler load: 50 kW)

Within comprehensive testing campaigns, the new technology has been stepwisely optimised. After the technology had proven its suitability for low-emission combustion of pellets and high quality wood chips, a series of test runs with different biomass fuels (reaching from different wood based assortments to agricultural residues) has been performed in order to investigate the operation stability, ash transformation processes and the emissions of the technology under the perspective of fuel flexibility. Since also fuels with ash melting temperatures <1,000°C have been applied, the gasifier was equipped with fuel bed cooling measures in the grate-zone, where the highest temperatures occur. Each test run thereby lasted for about 8 hours of stable operation. A strong focus was put on the evaluation of the ash transformation processes since they determine both, the particulate emissions as well as the operation stability of the system.

During the test runs characteristic process data such as air and flue gas flow rates, temperatures along the fuel bed and in the gas burner as well as the boiler load were recorded continuously. The flue gas composition downstream the boiler was analysed regarding the contents of O_2 (paramagnetic sensor), CO, NO (NDIR) and OGC (FID). The total suspended particulate (TSP) matter emissions were measured with a filter method according to VDI 2066 and the particle size distributions and concentrations of particles <1 μ m (PM₁) were determined with a 9-stage Berner-type low-pressure impactor (BLPI) and an electrical low-pressure impactor (ELPI). Moreover, the gaseous HCl and SO_x emissions have been determined using a wet chemical method according to VDI 3480.

Fuel, ash and if occurring slag samples have been collected and subsequently analysed regarding all relevant ash forming elements as well as carbonaceous species. Thereby the moisture content of fuel samples has been determined according to EN 14774, the ash content according to EN 14775, the C, H and N contents according to EN 15104 and the Cl content according to EN 15289. For the determination of the contents of major and minor ash forming elements as well as S in fuel and ash samples a multi-step pressurised digestion with HNO₃(65%)/HF(40%)/H₃BO₃ followed by measurement by inductively coupled plasma optical emission spectroscopy (ICPOES) or inductively coupled plasma mass emission spectroscopy (ICPMS) (depending on detection limits) has been performed.

The determination of the organic and inorganic carbon contents of the grate ash has been carried out according to EN13137. Selected samples of grate ashes have been analysed by SEM/EDX (scanning electron microscopy / energy dispersive X-ray spectrometry) and XRD (X-ray diffraction) in order to identify different phases formed and the elemental composition and chemical compounds prevailing in the grate ash.

The data gained from the test runs have been evaluated regarding emissions, slagging tendencies, general aspects of plant operation and especially regarding ash transformation processes. For the latter, mass, energy and element balances have been calculated to evaluate release ratios for relevant ash forming elements with a main focus on K. The evaluation of the results was supported by thermodynamic high-temperature equilibrium calculations (TEC). Therefore, the software FactSage 6.2 with the component database Fact 53 and the solution databases FToxid (slags and other oxide mixtures) and FTsalt (liquid and solid salt phases) has been applied. Moreover, to define the constraints for the calculations, gasifier bed simulations with the in-house code for updraft fixed-bed gasifiers mentioned above, have been performed.

5. Results and discussion

In the following firstly ash formation in the updraft gasifier technology is discussed as it is the basis for the low particulate emission operation of the new technology. Then, results regarding gaseous and particulate emissions are presented. Forest residues and miscanthus pellets are thereby used as case studies. Forest residues since they are a very common wood fuel in heating plants in all capacity ranges and miscanthus as a typical representative of herbaceous biomass fuels.

In Table 1 the chemical compositions of the forest residues and miscanthus as well as of the grate ashes gained from the test runs are summarised. Moreover, two fuel indexes [6] relevant for the potential for aerosol formation in conventional fixed-bed combustion processes (K+Na+Zn) and regarding the ash melting behaviour (Si+K+P)/(Ca+Mg+Al) are evaluated. Elevated values of the fuel index K+Na+Zn in common fixed-bed combustion systems thereby indicate an increase of the PM₁ emissions and an increased risk for deposit formation on boiler tubes. In this respect forest residues are expected to show moderate and miscanthus high fine particulate matter emission and deposit formation potential (see Figure). Regarding the ash melting index (Si+K+P)/(Ca+Mg+Al), decreased ash melting temperatures occur with increasing value of the index since Si, K and P are known to decrease the ash melting temperatures while Ca, Mg and Al contribute to the formation of phases with high melting temperature. The values determined indicate a shrinkage starting temperature above 1,200°C for forest residues and of about 800°C for miscanthus (see Figure). According to the fuel compositions, for forest residues a typical Ca-K-dominated fuel ash system is expected while miscanthus ashes are dominated by the behaviour of the K-Ca-Si-system.

To provide information about the general framework conditions of the test runs, in Table 2 some relevant plant operation data are summarised. As it can be seen from the low standard deviations of the boiler load, the air ratios and the flue gas temperature at boiler outlet, stable and constant operation conditions prevailed. Moreover, the data show a good comparability of the two test runs regarding primary air ratio and grate temperatures.

| | | | Forest residue | 5 | | Miscanthus | |
|---------------------|--------------|-------|----------------|---------------|-------|------------|---------------|
| | | fuel | grate ash | TSP emissions | fuel | grate ash | TSP emissions |
| Moisture content | wt% (w.b.) | 35.8 | | | 8.0 | | |
| Ash content | wt% (d.b.) | 2.4 | | | 2.9 | | |
| С | wt% (d.b.) | 48.3 | | | 47.3 | | |
| Н | wt% (d.b.) | 6.1 | | | 6.1 | | |
| Ν | wt% (d.b.) | 0.1 | | | 0.2 | | |
| S | mg/kg (d.b.) | 220 | 1,900 | 49,800 | 930 | 14,000 | 86,900 |
| Cl | mg/kg (d.b.) | 68 | 67 | 136,000 | 504 | 1,683 | 126,000 |
| Si | mg/kg (d.b.) | 2,410 | 90,200 | n.a. | 5,810 | 172,000 | n.a. |
| Al | mg/kg (d.b.) | 742 | 28,700 | 9,900 | 71.5 | 1,900 | 12,100 |
| Ca | mg/kg (d.b.) | 5,210 | 246,000 | 40,800 | 1,420 | 40,300 | 15,900 |
| Mg | mg/kg (d.b.) | 518 | 26,900 | 6,860 | 637 | 20,100 | 2,230 |
| K | mg/kg (d.b.) | 1,690 | 104,000 | 220,000 | 6,900 | 211,000 | 302,000 |
| Na | mg/kg (d.b.) | 79.3 | 3,050 | 5,000 | 264 | 8,050 | 17,300 |
| Fe | mg/kg (d.b.) | 531 | 34,000 | 21,500 | 81.4 | 2,880 | 5,700 |
| Р | mg/kg (d.b.) | 232 | 12,800 | 4,560 | 764. | 26,800 | 2,500 |
| Mn | mg/kg (d.b.) | 92 | 3,990 | 1,240 | 102 | 3,160 | 244 |
| K+Na+Zn | mg/kg d.b. | 1,779 | | | 7,185 | | |
| (Si+K+P)/(Ca+Mg+Al) | mol/mol | 0.8 | | | 6.3 | | |

Table 1. Chemical composition of the forest residues and the miscanthus applied as well as of ash and dust emission samples from the test runs (Explanations: d.b. ... dry basis; w.b. ... wet basis; n.a. ... not available since quartz filters have been used for TSP sampling).

Table 2. Mean values and standard deviations of relevant plant operation data during the test runs

| | | Forest residues | | Miscanthus | |
|---------------------------------------|-----------|-----------------|------|------------|------|
| | | mean | s | mean | S |
| boiler load | kW | 31.6 | 0.7 | 29.4 | 0.8 |
| grate temperature | °C | 902.8 | 25.0 | 939.4 | 46.8 |
| flue gas temperature at boiler outlet | °C | 161.2 | 1.3 | 144.7 | 2.5 |
| O ₂ content flue gas | vol% d.b. | 6.0 | 0.2 | 5.8 | 0.2 |
| primary air ratio | - | 0.20 | 0.01 | 0.20 | 0.01 |
| total air ratio | - | 1.40 | 0.02 | 1.38 | 0.02 |
| average pressure drop over fuel bed | Ра | 40.7 | 3.8 | 45.8 | 22.2 |

The grate ashes from forest residues were grey and mostly finely granulated (see Figure 2). Only loose ash agglomerations have been observed. The colour of these agglomerates was grey-brown. They were comparably soft and easy to destroy which indicates starting of ash sintering but no ash melting. In contrast the discharged grate ash from miscanthus was grey, partly finely granulated but partly also still in the form of pellets. These pellets were easy to destroy. Additionally, it contained rather big (some cm in diameter) sintered ash agglomerates. This was expected since the grate temperatures amounted to 939°C on average (see Table 2) and the ash melting index indicated the start of ash melting processes already at about 800°C. Representative grate ash samples from both test runs have been forwarded to chemical (see Table 1) and XRD analyses. Moreover, hard sintered resp. molten ash pieces from the test run with miscanthus were analysed by SEM/EDX.

Based on the operation data recorded (see Table 2), the results of the fuel, ash and dust analyses (see Table 1) and the results of the TSP, HCl and SO_x emission measurements (see Table 3) mass, energy as well as element balances for K, S and Cl have been calculated. From these data very low release ratios for K (0.44 wt% resp. 0.33 wt% of the K contained in the fuel is found in the TSP emissions) could be calculated. For comparison, in conventional grate fired systems the release ratios for K at 900 - 1,000°C are, depending on the fuel composition, in the range between 20 and 60 wt% [7], [8]. The S and Cl release ratios are as high as in conventional grate combustion plants (S: 74 and 47 wt%; Cl: 91 and 87 wt% for forest residues and miscanthus respectively).

The ash chemistry of forest residues is dominated by the behaviour of the Ca-K-system as most of the Si contained in the fuel comes from mineral impurities which show a low reactivity during ash formation. According to the results of the chemical analyses of the grate ash enough carbonate is present to bind the whole K and a part of the Ca. This is confirmed by the XRD analyses of the grate ash which indicate that K is mainly bound as $K_2Ca(CO_3)_2$ (Fairchildite). Ca is besides $K_2Ca(CO_3)_2$ also found in other carbonates such as $CaCO_3$ (Calcite) and $Mg_3Ca(CO_3)_4$ (Huntite). Ca not bound as carbonate (about 64 wt% of the Ca) occurs as CaO (Lime), Ca(OH)₂ (Portlandite) and Ca₃Al₂O₆. According to the XRD analyses, Si does not react with the other ash forming elements but forms SiO₂.



Figure 2. Pictures of the grate ashes of forest residues and miscanthus as well as results of XRD analyses (Explanations: left: grate ash - forest residues; middle: grate ash - miscanthus; right: result of XRD analyses of the grate ash from forest residues).

A comparable result has been achieved from TEC. As Figure 3 shows, at 700°C, which is a typical temperature for the gasification zone of the gasifier, $K_2Ca_2(CO_3)_2$, $CaCO_3$, as well as Ca-Mg-oxides and silicates dominate the ash composition. With increasing temperature $K_2Ca_2(CO_3)_2$ and $CaCO_3$ decompose to $K_2Ca(CO_3)_2$ and CaO and then $K_2Ca(CO_3)_2$ decomposes to K_2CO_3 and CaO. At temperatures above 800°C K_2CO_3 starts to form KOH and CO₂, which are released to the gas phase. The grate temperature measurements indicate temperatures above 900°C at which according to TEC the whole $K_2Ca(CO_3)_2$ should already have been transformed. However, the XRD analyses and also the wet chemical analyses prove the existence of this component which means that the decomposition process cannot be completed within the residence time of the ashes in the hot charcoal combustion zone. Therefore, K is mainly kept in Fairchildite and discharged with the grate ash. Consequently, in this K-Ca-ash system only very low amounts of K get released to the gas phase.



Figure 3. Results of TEC regarding ash formation under the constraints of an updraft gasifier fuel bed (Explanations: left: forest residues; right: miscanthus; below: miscanthus additivated with 6 wt% d.b. kaolin)

For miscanthus the ash chemistry is dominated by the behaviour of the Si-Ca-K-system. The grate ashes were mainly amorphous and therefore the XRD analyses revealed no results. According to TEC, K is mainly bound in low

687

melting K-silicates. SEM/EDX analyses of slag pieces found in the grate ash confirm that K-Ca-silicates with about 30 wt% K₂O, 60 wt% SiO₂ and 6 wt% CaO, which show according to the three-phase diagram of this system melting temperatures of about 950°C, caused the ash agglomeration. This temperature range is in good agreement with the grate temperature measurements (see Table 2: 939 +/-47 °C). The formation of K-silicates on the one side causes a severe slagging risk but on the other side is responsible for an almost complete embedding of K in the bottom ash.

For fuels with even more disadvantageous K/Ca/Si-ratios and thus lower ash melting temperatures, ash melting in the charcoal combustion zone of the gasifier may block the fuel bed. For such fuels, additivation with kaolin (an alumo-silicate mineral) may be an option, as the results of TEC in Figure 3 and also the evaluation of the ash melting index (Si+K+P)/(Ca+Mg+Al) indicate (see Figure). With kaolin addition K-Al-silicates, which have higher melting temperatures than K-silicates, are preferably formed. Besides reduced ash melting an enhanced embedding of K in the grate ash can be achieved (no K-release according to TEC).

Almost all chemically untreated biomass fuels can be attributed to one of the two ash systems explained and therefore, K retention in the grate ash should work accordingly. Since K is the most relevant element with regard to fine particulate matter emission formation, this results in very low PM emissions of the new technology. In addition, with the staged gas burner concept applied a combustion of the product gas with almost zero CO and OGC emissions can be achieved. This is proven by the emission data gained from the test runs (Table 3). About the same results have also been achieved for the other fuels investigated within the testing campaign. Besides CO emissions <10 mg/Nm³ for all fuels the TSP emissions for poplar, willow, agropellets, olive tree prunings, vineyard pruning, and olive stones could be kept below 6.5 mg/Nm³ (related to dry flue gas and 13 vol% O₂) without application of a filter.

Table 3. Gaseous and particulate emissions determined during the test runs with forest residues and miscanthus

| | | Forest Residues | | Miscanthus | |
|--|---|-----------------|------|------------|------|
| | | mean | S | mean | S |
| CO emissions | mg/Nm ³ (related to dry flue gas and 13 vol% O_2) | 6.9 | 2.3 | 6.1 | 0.6 |
| HCl emissions | | 0.85 | 0.24 | 30.06 | 3.04 |
| SO_x emissions (as SO_2) | | 18.89 | 2.83 | 66.29 | 8.78 |
| PM ₁ emissions (measured with BLPI) | | 1.5 | 0.16 | 2.2 | 0.13 |
| TSP emissions | | 2.9 | 0.33 | 2.5 | 0.01 |

In Figure 4 the relation of the fuel index K+Na+Zn with PM_1 emissions is shown to make the ultra-low particulate emission levels of the new technology in comparison to conventional grate-fired biomass combustion systems obvious. Moreover, the relationship of the molar (Si+K+P)/(Ca+Mg+Al) ratio of the biomass fuels tested with the related shrinkage starting temperatures are presented to underline the big differences regarding ash melting and the positive effect that additivation may have.



Figure 4. Evaluation of the test runs results with respect to the fuel indexes K+Na+Zn and (Si+K+P)/(Ca+Mg+Al) (Explanations: diagram on the right hand: dashed red line: miscanthus with 6 wt% d.b. kaolin)

It has also to be mentioned that the new technology causes a certain increase of HCl and SO_x emissions. Since S and Cl release takes place at comparably low temperatures (Cl starts at about 200°C and about 75% of S is released during the pyrolysis phase [9]) these elements only pass through the upper part of the fuel bed and can therefore not be efficiently embedded. Due to the lack of gaseous and solid K-compounds and Ca-containing coarse fly ashes in the flue gas almost no reaction partners are available for the formation of chlorides and sulphates and thus, S and Cl are mainly emitted as gaseous HCl and SO_x (mainly SO₂). The range of the emissions however, is usually on such a low

level, that no problems regarding SO_2 emission limits, as they are prescribed by the EC medium-scale combustion directive [10] have to be considered.

6. Summary and conclusions

A new ultra-low emission combustion concept based on a fixed-bed downdraft gasifier directly coupled with a multi-stage gas burner and a boiler has been developed. The concept has already proven its applicability for small-scale residential wood chip and pellet boilers and has also been introduced into the market in 2016.

As the investigations presented in this paper have shown, the new concept also represents a promising approach regarding fuel flexible combustion. If the grate zone of the gasifier is cooled appropriately even fuels with rather low ash melting temperatures (down to 900°C) can be applied. Even in case of the utilisation of ash and K-rich biomass assortments, the gasifier concept can keep the fine particulate and the TSP emissions on such low levels (<5 mg/MJ_{NCV}) that no additional filter as in conventional combustion systems is needed. The low dust contents of the product gas at burner inlet also enable an operation at low excess air ratios without slagging problems.

Moreover, the test runs have shown, that also for (in terms of combustion) more difficult fuels such as forest residues and miscanthus almost zero CO and OGC emissions are possible. One additional advantage of the new concept is, that these low emissions are achievable at very low excess air ratios (a total excess air ratio as low as 1.2 is possible with the gas burner concept applied) which increases efficiencies.

With the new fuel-flexible combustion concept a technological milestone regarding efficient biomass combustion could be achieved. Further work is planned regarding the implementation of the new technology in the medium capacity range and regarding the further development and optimisation of the gas burner in order to achieve also ultralow NO_x emissions in the future.

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