Oscillating combustion of different fuel types for NO_X reduction in grate furnaces and coal burners

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A B S T R A C T

Oscillating combustion is a method to reduce NO_X emissions and is to be understood as a time-resolved staged combustion where either the fuel or the secondary air supply is oscillated. These two operating options with oscillating combustion have been the focus of the research group behind this paper for several years. In this work, experimental investigations of an oscillating air supply demonstrate nitrogen oxide reductions in the combustion of hard coal, lignite, wood pellets and waste wood chips by up to 35 %. Investigations were carried out in a test facility for pulverized fuel called BRENDA, in a laboratory fixed bed reactor called KLEEA and an industrial vibrating-grate firing system.

1. Introduction

Despite numerous alternative and renewable energy sources, fossil fuels today account for 80 % of global primary energy demand. In Germany, coal still accounted for 28 % of electricity generation in 2021 [1]. The energy conversion from chemical to electrical energy takes place almost exclusively via combustion processes of fossil or alternative energy sources such as waste or biomass. Combustion processes produce numerous environmentally harmful air pollutants. Against the background of climate change and the public debate on environmental pollution, the reduction of pollutants is of great importance. For this reason, in Germany, the framework conditions for air emissions from industrial plants are defined by the Bundes Imissionsschutz Gesetz (BImSchG). NO_X still plays a special role in the assessment of pollutant emissions. To comply with the limits for nitrogen oxide emissions two different approaches for the reduction of NO_X concentrations in the flue gas of industrial plants are utilized. The so-called primary measures address the combustion-related fundamentals of nitrogen oxide formation. These formation mechanisms can be inhibited by appropriate air distribution in the combustion chamber. The secondary measures comprise the subsequent denitrification of the flue and include processes such as Selective Catalytic Reduction (SCR) and Selective Non-Catalytic Reduction (SNCR). Post-treatment of the flue gases is usually associated with higher investment and lower plant efficiency, while the primary denitrification is more economical and ecological [2]. Oscillating combustion is one of the primary nitrogen oxide reduction measures. Oscillating fuel addition has so far led to significant reductions in NO_X emissions. The changing stoichiometry that occurs in the flame results in less NO_X. By now, the investigations carried out have been largely limited to the oscillating addition of natural gas, which is easy to oscillate. In contrast, examinations on oscillating combustion of solid fuels are still scarce.Table 1

The first trials of operating in an oscillating combustion mode for solid fuels have been undertaken in a pilot plant called BRENDA by oscillating the addition of pulverized hard coal [3] and in a grate furnace by oscillating the secondary air mass flow [4–6]. The trials demonstrated the potential of oscillating combustion to reduce NO_X emissions. The research was expanded further to the oscillating addition of the air

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Table 1
Overview of investigated setups.

Test facility	Fuels	Frequency
BRENDA (pulverized fuel boiler)	hard coal lignite	2 Hz
KLEAA (fixed bed reactor)	wood pellets waste wood chips	1 Hz
Industrial grate furnace (Polzenith)	waste wood	0.19 Hz 0.38 Hz 0.75 Hz

flow rates in the coal burner of the pilot plant BRENDA. This approach results in a similar NO_X reduction potential, while avoiding the technical challenge of oscillating an abrasive hard coal dust flow [2,7]. The oscillating combustion of solid fuels is influenced significantly by the fuel *N*-mechanism. Since different solid fuel types as hard coal, lignite and biomass show different contents of fuel bond nitrogen, this work focuses on the oscillating combustion of these fuels, as well as the transfer of the operating conditions on different combustion technologies.

The novelty of the current work is the demonstration of the oscillating combustion in different combustion setups for solid fuels where the impact of the fuel itself plays a minor role on the reduction potential for NO_X emissions.

The following table shows a brief overview about the investigated test facilities, fuel types and the corresponding oscillation frequency for the secondary air. Since the oscillation frequency is not part of this research, reference is made at this point to [7].

The purpose of this paper is to demonstrate that oscillating combustion is a general principal for the reduction of NO_X in solid fuel combustion systems. It will be shown that this holds true for combustion systems with completely different working principles, for fuels of different nature and particle size. Hence, in this paper, pulverized fuel combustion in a swirling flame, and a packed (lab scale reactor) and a moving (industrial grate) bed combustion system serve as examples to demonstrate the effectiveness of oscillating combustion for NO_X reduction. Note that it is not our goal to re-examine all the details of these three combustion principles as many detailed studies on pulverized fuel combustion and packed and moving bed combustion can be found in literature. We will restrict the examinations to the fundamental difference in NO_X emission of oscillating and non-oscillating combustion in the sense of a proof of principle.

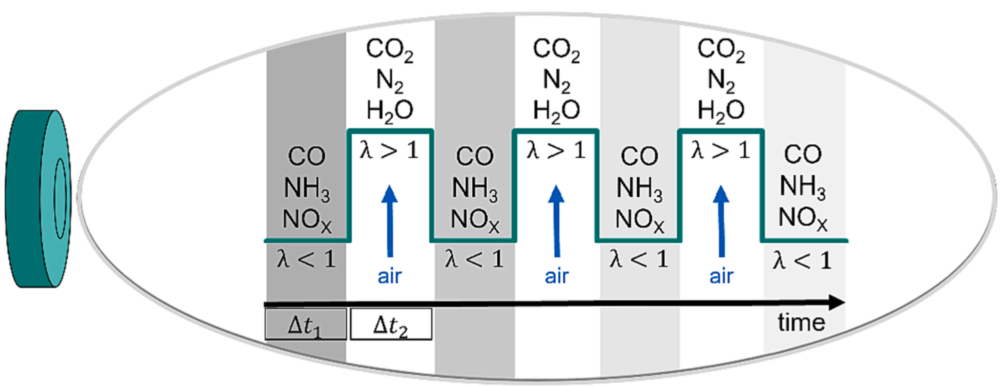

Fig. 1. Principle of oscillating combustion.

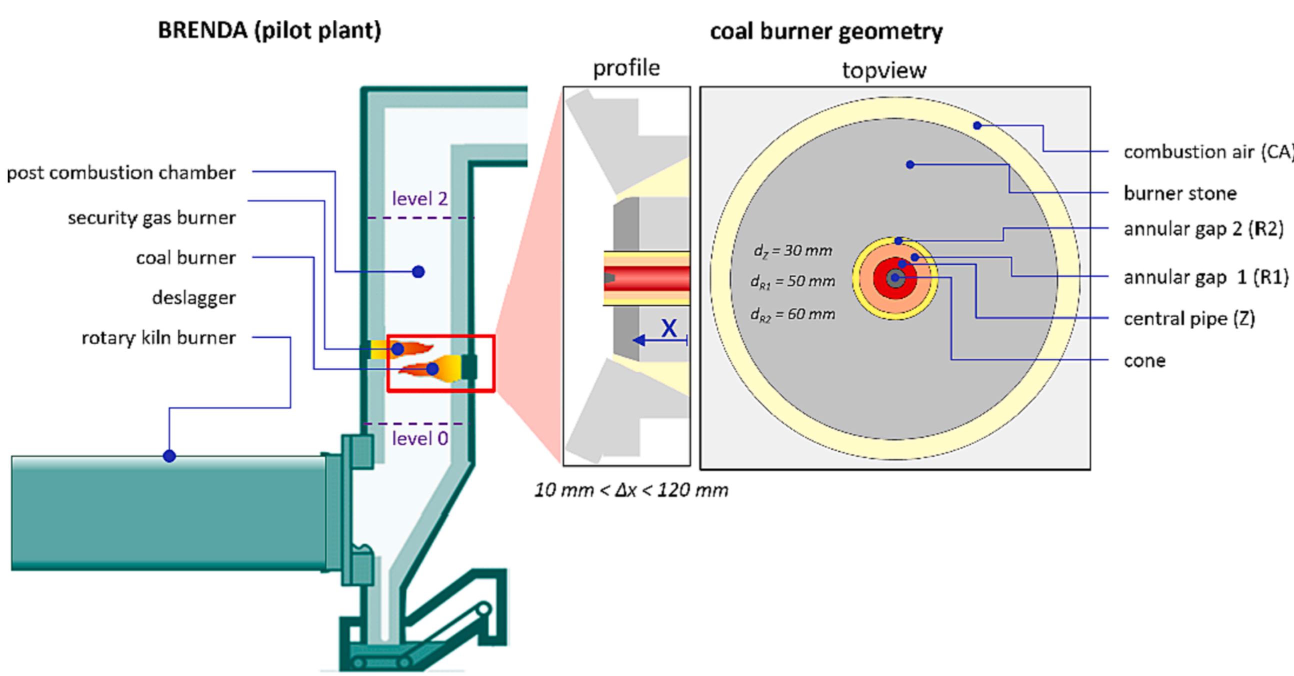

Fig. 2. Setup of BRENDA facility.

1.1. *Theory of oscillating combustion*

For pulverized solid fuel combustion in most cases fuel NO formation is dominating. A detailed description of the formation mechanism can be found in [2]. In contrast to the well-known secondary measures for NO_X reduction such as the above-mentioned SCR and SNCR, the primary measures affect the combustion process itself. Among these are fuel and air staging, which are usually local, as well as the novel time-dependent oscillating combustion.

In the recent years, the oscillating combustion gained the interest of many researchers as a primary reduction measure for NO_X [3,4,6,8,9]. The result of a field study from 1996 to 2003 on oscillating combustion with gas flames [10] shows a significant reduction of NO_X emissions of 31–67 %. The results were confirmed by [3,4,6,8,9] for other combustion systems.

The basis is the oscillating addition of fuel or air into the reaction zone of the flame, which causes a change of the local stoichiometry [4,6]. The different reaction zones occur immediately after the fuel leaves the burner. As the distance to the burner increases, the axial cross mixing of the combustion air progresses. Fig. 1 shows this behavior schematically.

The oscillating addition of fuel/air leads to oscillating reaction conditions in the reaction zone of the flame. Lean-fuel ($\lambda > 1$) and fuel-rich reaction zones ($\lambda < 1$) follow each other. Fig. 1 shows schematically the air staging combustion mode. The reaction conditions, however, are not adjusted by locally directed dosage of secondary air, but by oscillating air addition. Accordingly, oscillating combustion can be regarded as a time-resolved variant of staged combustion [9]. In the fuel-rich time periods Δt_1, CO, NH_3 are increasingly produced while in the lean-fuel time periods Δt_2 CO reacts to CO_2 and NO_X with NH_3 reacts to N_2 and H_2O [4,6].

Previous work investigated the NO_X reduction potential by an oscillating addition of natural gas [8–10]. It was illustrated that an oscillating addition of the secondary air during the combustion of chipboard cubes in a fixed-bed reactor results in a significant reduction of NO_X emissions. None of these studies, however, show the effect of oscillating combustion on NO_X emissions if a solid fuel is used. The high abrasion inside the pinch valve used to realize the oscillating supply of the hard coal dust is probably the most challenging issue. In this paper, the results of the oscillating combustion of pulverized hard coal and lignite with a coal burner are presented.

2. Materials and methods

2.1. *Test facility BRENDA*

The so-called BRENDA test rig comprises a rotary kiln, which is connected to a post combustion chamber. The rotary kiln is equipped with an oil burner which supplies the process heat required for the combustion chamber. Fig. 2 shows the section of the post combustion chamber where the investigations were carried out.

A swirl burner (made by SAACKE[1]) for pulverized coal firing with a thermal output of 1 MW and a gas burner are staggered at the same level on the wall of the post combustion chamber in an opposite manner. This ensures that in an event of a failure of the pulverized coal burner, the temperature in the post combustion chamber will not drop down below 850 °C. This has to be ensured according to the 17 BImSchV[2] for guaranteeing the safe operation of the plant. The gas burner is shown in Fig. 2 as well.

The NO_X concentration is measured at level 0 and level 2. The NO_X emission resulting from the pulverized coal firing is determined by the difference between the NO_X load of the dry flue gas from level 2 and level 0. It is assumed that the nitrogen oxides entering level 0 are inert and so no longer react with other gas compounds. Fig. 2 shows the geometry of the pulverized coal burner in detail. The burner, the fuel-bearing central tube, the annular gap 1 (R1), the annular gap 2 (R2), quart block stone and the combustion air inlet (CA) are arranged concentrically. A cone serves to stabilize the flame and is set at a constant distance of 15 mm in x-direction during the entire test campaign (the maximum diameter of the cone is 15 mm).

The pulverized coal enters the combustion chamber through the central pipe Z or the annular gap R2. The combustion air enters the furnace through the exterior gap and is fed tangentially into the burner, entering the combustion chamber with a swirl. During operation, the burner block can be displaced along the marked x-axis into the furnace wall, changing the gap width of the combustion air inlet.

Table 2
Experimental setup for BRENDA (combustion of hard coal and lignite.

parameter	coal	lignite	unit
coal in Z	57	45	kg/h
air in Z	75	75	m_N^3/h
combustion air in CA	370	208	m_N^3/h
air in R2	130	200	Hz
oscillation in R2	0/2	0/2	–
swirl number	0.37	0	–
λ	1.16	1.26	–
λ_{loc}	0.42	0.71	–

Table 3
Experimental setup for BRENDA (investigation of λ_{loc}).

	trial							
parameter	1	2	3	4	5	6	7	unit
λ_{loc}	0.51	0.46	0.42	0.36	0.31	0.29	0.17	kg/h
air in R1	180	150	130	100	75	68	0	m_N^3/h
air in R2	180	150	130	100	75	68	0	m_N^3/h
combustion air in CA	258	288	308	338	363	388	438	m_N^3/h
	constants							
coal in Z	57							kg/h
air in Z	75							m_N^3/h
oscillation in R2	0/2							Hz
swirl number	0.3							–
λ_{burner}	1.5							–

2.1.1. *Test settings for the BRENDA test trials*

The test settings of the incoming air, the mass flows, the pulsation frequency and the position of the pinch valve can be found in Table 2. Hard coal and lignite have been used as fuel, the fuel data can be found in chapter 2.4. For the comparison of the results, the *N*-conversion rate calculated according to Equation (5) is used (see chapter 2.2.1).

In addition, to the comparison of hard coal and lignite for a base case, the local air ratio λ_{loc} and the swirl number have been varied for hard coal.

2.1.2. *Variation of local air ratio λ_{loc}*

The local air ratio λ_{loc} is calculated as a function of the hard coal mass flow, the minimum required air quantity l_{min} and the volume flows $\dot{V}_Z$, $\dot{V}_{R1}$, $\dot{V}_{R2}$ according to Equation (1).

$$\lambda_{loc} \quad \frac{\dot{V}_Z + V_{R1} + \dot{V}_{R2}}{l_{min} \bullet \dot{m}_{coal}} \tag{1}$$

Another stoichiometric parameter λ_{burner} which considers the combustion air flow entering the pulverized coal burner $\dot{V}_{CA}$ is defined by

[1] https://www.saacke.com/de/home/.

[2] Bundesimmissionsschutzverordnung (engl. Federal immission protection ordinance).

Table 4
Experimental setup BRENDA (swirl investigation).

	trial						
parameter	1	2	3	4	5	6	unit
swirl number	0.99	0.82	0.71	0.59	0.51	0.36	–
	constants						
coal in Z	57						kg/h
air in Z	75						m_N^3/h
oscillation in Z	0/2						Hz
combustion air in CA	438						m_N^3/h
λ_{burner}	1.03						–

Equation (2).

$$\lambda_{burner} \quad \frac{\dot{V}_Z + \dot{V}_{R1} + \dot{V}_{R2} + \dot{V}_{CA}}{l_{min} \bullet \dot{m}_{coal}} \tag{2}$$

Constant swirl number and stoichiometric air ratio are achieved by the adjustment of the parameters listed in Table 3. The experiments 1 to 7 were carried out in non-oscillating and oscillating operational mode at 2 Hz, respectively.

2.1.3. Variation of swirl number

The pulverized hard coal enters the post combustion chamber through the central tube Z. The combustion air is supplied through the exterior gap and is fed tangentially into the burner, entering the combustion chamber with a swirling flow. During operation, the burner block can be shifted along the x-axis (see Fig. 2), thus, changing the gap width of the combustion air inlet. Since the swirl number S is determined only by geometrical considerations it can be calculated as a function of the traversed path Δx according to Equation (3) [11]. The distance Δx 0 mm corresponds to a swirl number S of 0, the maximum of Δx is 110 mm.

$$S \quad 0.01466 \frac{1}{mm} \bullet \frac{130mm + \sin^2(30^\circ) \bullet \Delta x}{130mm + \sin(30^\circ) \bullet \Delta x} \bullet \Delta x \bullet Q^2, with \Delta x in mm \tag{3}$$

For calculating the swirl number S according to Eq. (3), the following Equation (4) for the ratio Q is needed. Q is the ratio of the swirled combustion air V_{CA} to the total incoming air volume flow of the burner.

$$Q \quad \frac{\dot{V}_{CA}}{\sum_i^n \dot{V}_{i,air}} \tag{4}$$

In order to investigate the swirl influence, all incoming mass flows are listed in Table 4. The variation of the different swirl numbers listed in Table 4 were carried out in non-oscillating and oscillating operational mode at 2 Hz. All incoming mass flows were kept constant.

2.2. Lab reactor KLEAA

The experimental facility KLEAA represents a fixed bed reactor, in which the combustion behavior of solid, lumpy fuels is characterized (see Fig. 3).

The facility comprises three units: a combustion chamber, which contains the fuel, which is passed by primary air, a post combustion chamber, where the gases from the combustion chamber are post-burned by secondary air addition and a flue gas cleaning system. After the flue gases leave the post combustion chamber they enter the flue gas cleaning system, where they are cooled down to approx. 170 °C, followed by dust removal in a baghouse filter and an activated coal adsorber. The gas composition is measured at two locations, directly downstream of the combustion chamber (gas sampling 1, raw gas) and downstream of the bag house filter (gas sampling 2, clean gas).

The secondary air can be added in an oscillating manner by a pinch valve.

2.2.1. Operation mode

The fuel – in this case wood pellets with a length of approx. 1 cm and a diameter of ca. 6 mm is filled inside the cold fixed bed. The fixed bed stands on rails and after filling the fuel into it, it is pushed under the already electrically heated top part of the combustion chamber. The

Table 5
Elemental composition of the examined fuels.

parameter	coal	lignite	wood pellets	waste wood	unit
$x_{Fuel,\ C}$	0.8280	0.6100	0.4710	0.4794	kg_C/kg_{Fuel}
$x_{Fuel,\ H}$	0.0530	0.0450	0.0580	0.0591	kg_H/kg_{Fuel}
$x_{Fuel,\ O}$	0.0480	0.2158	0.3907	0.3765	kg_O/kg_{Fuel}
$x_{Fuel,\ N}$	0.0160	0.0065	0.0012	0.0409	kg_N/kg_{Fuel}
$x_{Fuel,\ S}$	0.0120	0.0077	0.0001	0.0003	kg_S/kg_{Fuel}
$x_{Fuel,\ Cl}$	0.0020	0.0000	0.0001	0.0017	kg_{Cl}/kg_{Fuel}
$x_{Fuel,\ w}$	0.01100	0.0610	0.0760	0.0310	kg_w/kg_{Fuel}
$x_{Fuel,\ a}$	0.0300	0.0540	0.0030	0.0112	kg_a/kg_{Fuel}
min. oxygen demand	2.59	1.77	1.33	1.37	kg_{O2}/kg_{Fuel}
min. air demand	11.17	7.65	5.97	5.89	kg_{air}/kg_{Fuel}
net calorific value	33.8850	22.807	17.300	17.800	MJ/kg_{Fuel}

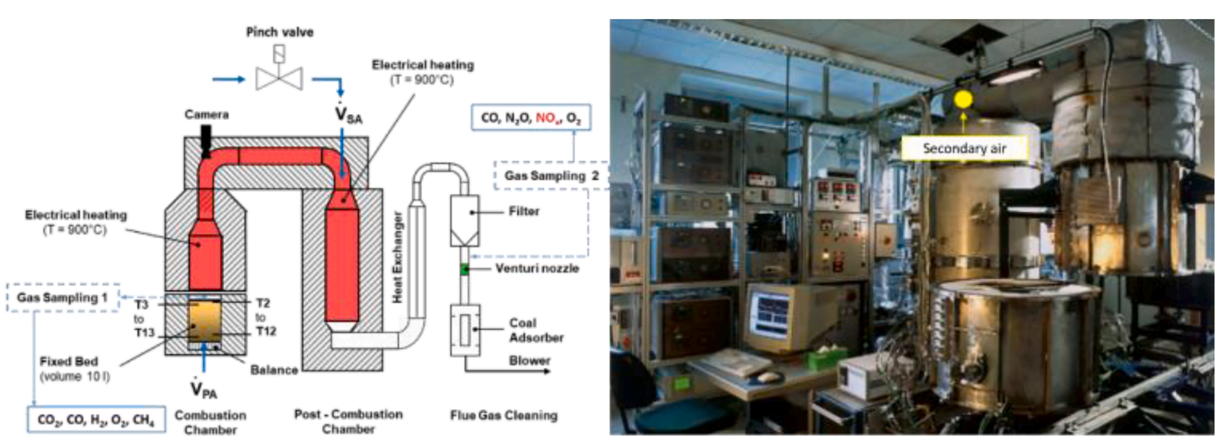

Fig. 3. Schematic layout of the KLEAA fixed bed reactor.

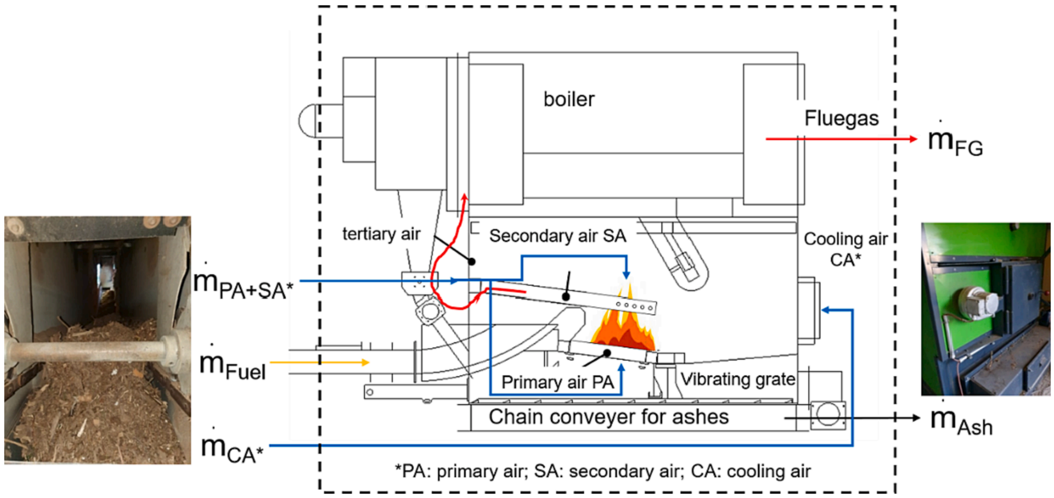

Fig. 4. Balance system of the plant at Polzenith.

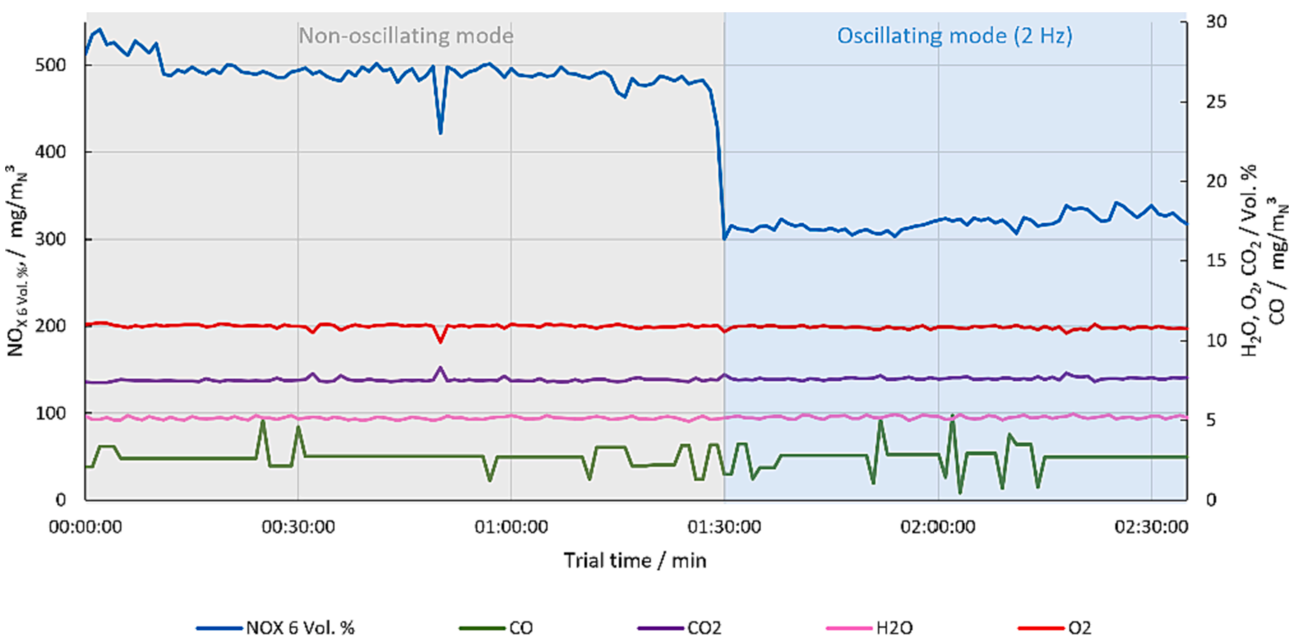

Fig. 5. Base case with hard coal.

primary air is supplied with 10 m$_N^3$/h. The mean value of the secondary air is set to 25 m$_N^3$/h and is either constant or oscillated with a frequency of 1 Hz.

After a short drying period, the released volatile components ignite. Note that the electrical heating is in operation throughout until the end of the combustion process.

The combustion front migrates in the opposite direction of the primary air towards the bottom of the grate, where depending on fuel and operating conditions the char burnout follows. The experiment is considered completed, when the oxygen content in the clean gas is close to 20 vol%.

2.3. *Test facility grate furnace*

The pilot plant at Polzenith[3] is used for hot water supply and thus, it is not operated in a continuous mode instead it underlies an interval operation. Therefore, balancing and correlation of measured NO_X concentrations by comparable operating parameters is rather difficult. Waste wood (see Table 5) has been used as a fuel. Besides the non-continuous operation, the inhomogeneous fuel adds another “variable”. The facility layout, as well as the mass balance space frame are schematically represented on Fig. 4.

The image on the left side peers at the metering/dosing and the fuel, the cooling air blower for the ash discharge is seen on the right.

Primary air is supplied to the grate from below. The distribution of the air on the grate cannot be controlled, so it depends strongly on the fuel bed height and the local pressure drop how the air gets into contact with the fuel. Above the fuel supply over a “knee” the secondary air is injected from both walls left and right in the direction of the fuel flow. The secondary air distribution is not known, only one ventilator on the right side of the wall supplies the secondary air. The cooling air is supplied with a fixed volume flow. All volume flows are unknown, the overall stoichiometry is controlled by the oxygen concentration in the flue gas. For our application the primary air flow, the secondary air flow, the flue gas flow as well as the fuel mass flow were determined with the

[3] https://www.polzenith.de/.

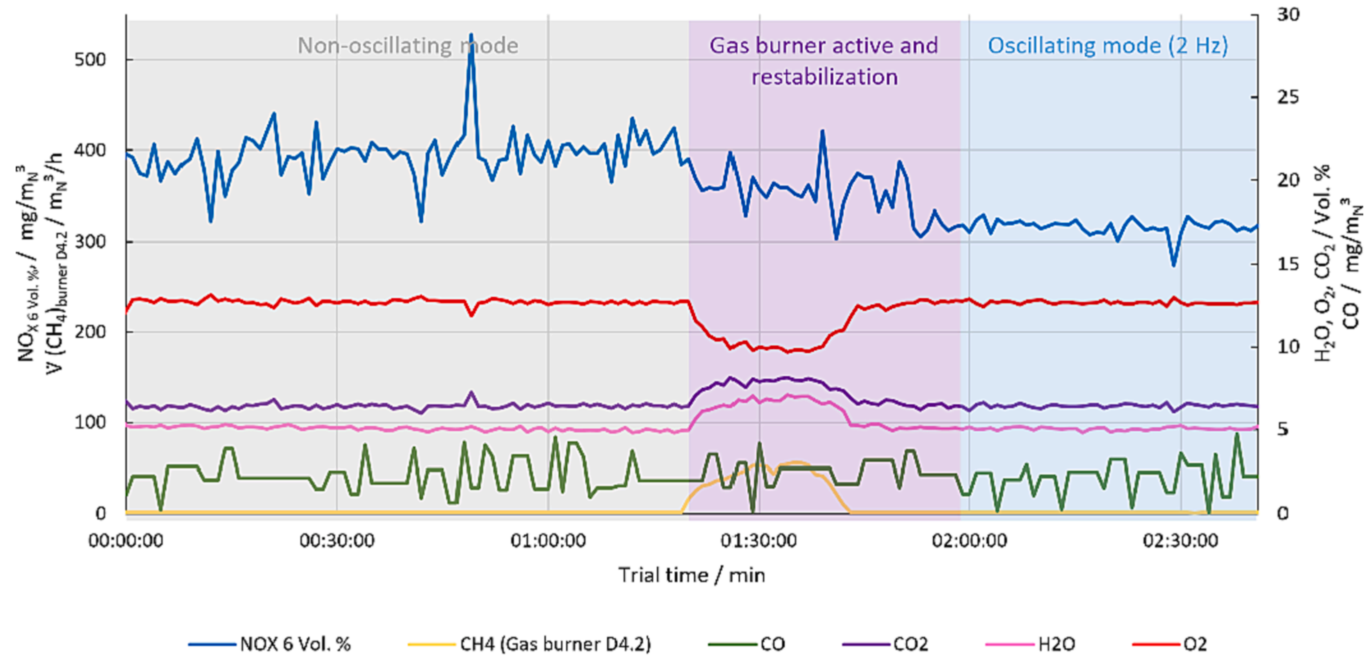

Fig. 6. Base case with lignite.

help of additional measurements with pitot tubes and weighing.

2.4. Fuel characterization

Various fuels were used in the three different experimental facilities BRENDA (see chapter 2.1), KLEAA (see chapter 2.2) and the grate furnace (see chapter 2.4). In the pilot plant BRENDA, hard coal and lignite were fired in a pulverized coal burner. The fuel for the KLEAA setup was wood and waste wood pellets. In the grate furnace only waste wood (as delivered) was fired. The chemical composition of the used fuels hard coal, lignite, wood pellets and waste wood (as delivered and pellets) are shown in Table 5. The difference between waste wood and its pellets is only the water content. For the waste wood it is about 5.2 wt-%, for the pelletized waste wood it is about 3.1 wt-% as it is shown in Table 5. The comparison illustrates the difference in fuel-bound nitrogen, which is largely responsible for the nitrogen oxide formation by the fuel N mechanism in the tests carried out.

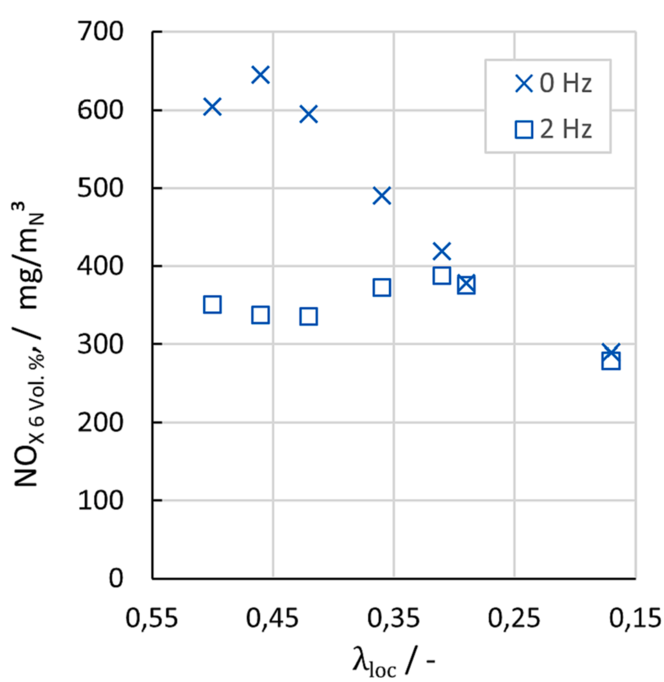

Fig. 7. Results of investigation of λ_{loc}.

2.4.1. N-conversion rate

To compare the effects of oscillating combustion for the different tested fuels the conversion rate CR_N for the fuel bound nitrogen was calculated according to Equation (5).

$$CR_N \quad \frac{c_{NO_X} \bullet \dot{V}_{offgas} \bullet (1 \quad x_{H_2O,offgas})}{\dot{m}_{coal} \bullet x_{N,coal}} \bullet \frac{\widetilde{M}_{NO_2}}{\widetilde{M}_N}, in\% \tag{5}$$

3. Experimental results

3.1. Brenda

3.1.1. Base case with hard coal and lignite

Two long-time runs, one with hard coal and one with lignite, respectively were carried out at BRENDA. The results are summarized in the following figures. Fig. 5 shows the NO_X, 6 vol% concentration over the experimental time for the test run with hard coal. The hard coal mass flow, the entering air flows and the stochiometric air ratios λ and λ_{loc} were kept constant. With the beginning of the oscillating operational mode at 01:30 with 2 Hz, the NO_X emissions decline about 34.8 % from 492 mg/m$_N^3$ down to 321 mg/m$_N^3$. The concentrations of the measured flue gas components H_2O, O_2, CO and CO_2 do not show any significant changes over the trial period and stay stable. The results are in a good agreement with previous findings [2,3] and confirm the positive influence of oscillating combustion on NO_X emissions.

Fig. 6 shows the results for the base case with lignite where an average NO_X concentration of 384 mg/m$_N^3$ can be observed for the non-oscillating operational mode. Here, after 80 min of trial time the gas burner had to be activate due to operational security issues. During the operation of the gas burner the concentrations of H_2O and CO_2 are increasing because additional fuel (natural gas) is entering the burning chamber. With the beginning of the oscillating operational mode (2 Hz at 02:00), the NO_X emissions stabilize at a value around 316 mg/m$_N^3$ resulting in an average reduction of 17.7 %. The results clearly show that

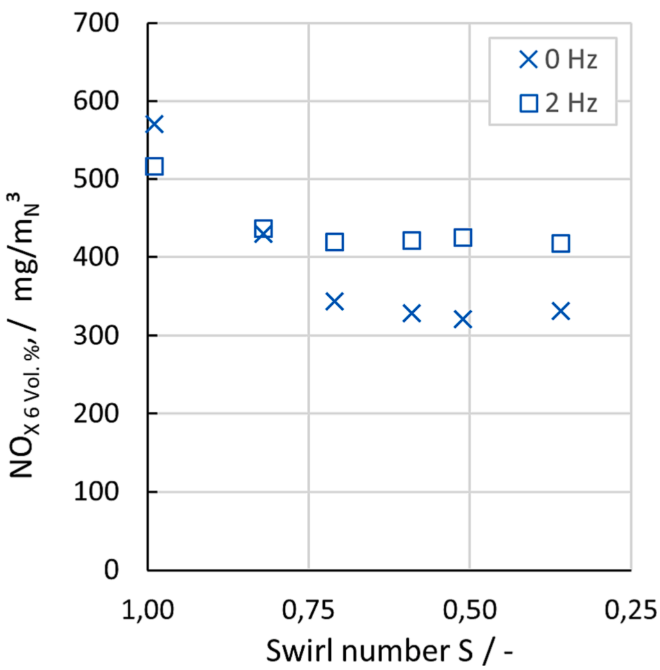

Fig. 8. Results of investigation of swirl.

the positive effect of oscillating combustion is also transferable on the combustion of lignite with a coal burner. Besides the effect of the activation and deactivation of the gas burner no significant changes in the concentrations of the flue gas components H_2O, O_2, CO and CO_2 are observed during the period of investigation.

3.1.2. *Local stoichiometric air ratio λ_{loc}*

The results of the changing stoichiometric air ratio λ_{loc} are shown in Fig. 7 for the hard coal flame. In non-oscillating mode, the staged reduction of λ_{loc} is leading to a reduction of the NO_X concentration for $\lambda_{loc} < 0.45$. Decreasing λ_{loc} from 0.45 down to 0.17, which is associated with an increasing oxygen deficit, shifts the equilibrium via the NO-reburning mechanism to HCN, resulting in the lowest NO_X concentration of about 170 mg/m_N^3. Under oscillating combustion conditions, the same staged reduction of λ_{loc} does not result in a staged reduction of the NO_X concentration. The oscillating operational mode seems to diminish the influence of the local stoichiometric air ratio on NO_X. The results match well with the previous findings [2] where the influence of the momentum of the secondary air (resulting in different λ_{loc}) on the NO_X concentration showed a reduction for $\lambda_{loc} > 0.28$. The difference of the NO_X concentration of oscillating and non-oscillating operational mode peaks at λ_{loc} 0.46 for non-oscillating mode showing a reduction of the NO_X concentration of 48 %. For values of $\lambda_{loc} < 0.3$ the NO_X concentration of oscillating and non-oscillating combustion are almost similar. The threshold value of λ_{loc} at 0.3 can be explained by the fact that with decreasing λ_{loc} a parallel shift of the air from the inner gap R2 to the outer annular gap CA occurs (see Fig. 2). This shift is needed to ensure a constant λ_{burner}. The momentum of the oscillated air flow in R2 causes the typical time-resolved staged combustion as shown in Fig. 1. With decreasing the air flow rate in R2 and increasing the air flow rate in CA the momentum of R2 declines. At a $\lambda_{loc} < 0.3$ this momentum is too small to cause an oscillating time resolved change of the reaction conditions in the flame area.

3.1.3. *Swirl*

A clear correlation between NO_X emissions and swirl number can be observed. Fig. 8 shows the NO_X concentrations for non-oscillating and oscillating (2 Hz) operation modes for swirl numbers below 1.

In a non-oscillating operational mode, the NO_X concentrations decrease with the reduction of the swirl number. The minimum is reached at a swirl number of 0.5. The observed behavior of an increase of the NO_X concentration with an increasing swirl number is contrary to the literature [12] where the recirculation zone in type I and type II flames leads to a NO_X reduction of swirl stabilized flames. The recirculation zone develops at a critical swirl number of $S > 0.5$ for gaseous flames. However, [12] clearly states that the swirl optimization of oil or coal flames is difficult due to the different ignition stability limits and the reaction kinetics. At 2 Hz oscillation frequency, the NO_X concentration for a swirl number of 1 is slightly reduced by 14 % compared to the non-oscillating operational mode. A further reduction of the swirl number leads for $S < 0.79$ to a minimum increase of the NO_X concentration. The difference of the dependency of NO_X concentration on swirl number for non-oscillating and oscillating combustion mode, can be explained by the fact of the intermittent operation for oscillating combustion (inner air flow R2 is turned on and off), preventing the formation of a stable recirculation zone. The absence of the recirculation zone, suppresses the NO reburning mechanism, resulting in higher NO_X concentrations for the oscillating operating mode. According to previous studies [2,13] no significant effect of swirl number on NO_X could be observed since the low quantity of data points did not allow for a reliable statement. Here, the results illustrate that the oscillating operational mode has a negative effect on the NO_X concentration of swirl stabilized hard coal flames. The lowest NO_X concentrations are achieved at swirl numbers $S < 0.7$ and non-oscillating conditions.

3.2. *KLEAa*

Results for the experiments with waste wood pellets are summarized in Fig. 9.

The graphics on the left side show the measured raw gas concentrations above the fuel bed, the supplied primary air amount, and the balance signal. The graphics on the right side show the clean gas concentrations after the supply of the secondary air. The graphics at the top are the experiments without oscillation and the bottom ones with secondary air oscillation with 1 Hz. Note that fuel consumption is very uniform indicated by the steady mass loss of the fuel bed indicted by the blue line. At first, the fuel is heated, dried and starts to release volatiles. After 3 min, the volatile components ignite and the reaction products CO_2, CO, increase, while O_2 is consumed (see Fig. 9 top left). Furthermore, some unreacted CH_4 is left from the pyrolysis gases. In addition, char can react with the water vapor from the lower fuel layers to H_2 and CO. This is indicated by a slightly delayed increase of the H_2 formation. At the clean gas side, the O_2-decrease correlates with the beginning of the ignition and the burn-off of the solid material, the CO_2-concentration increases and reaches a peak after approx. 11 min, almost at the same time with the peak in CO-formation above the fuel.

By the end of the fuel ignition in the combustion chamber and with the start of the char burn-off NO_X reaches its maximum; at this point the fuel ignited completely and the maximal release rate of NH_3, as a part of the volatile components is reached. The peak of N_2O at the beginning of the combustion (3 min) is due to the release of HCN from the pyrolysis. Combined with O_2, N_2O is formed at lower temperatures (below 900 °C). The maximal temperature of the fuel bed up to the minute 4 is about 850 °C (see Fig. A1 in the annex). The N_2O peak is related to a smaller peak of CO; this results from the reaction of NCO and NO to N_2O and CO [10]. With the increase of the released hydrogen N_2O is reduced again. By the application of oscillation, NO_X can be significantly lowered down from 1800 mg/m_N^3 to 880 mg/m_N^3 at low CO concentrations at the same time. The O_2 level is below that of the experiments without oscillation, which is caused by the higher flue gas mass flow from the fuel bed. Using the equation for the conversion rate of fuel nitrogen into NO_X the conversion rate CR gives 6.3 % for the mode without oscillation, with oscillation 4.3 % is achieved.

The results for the wood pellets are shown in Fig. 10 for raw and clean gas without oscillation (top) and for the experiment with

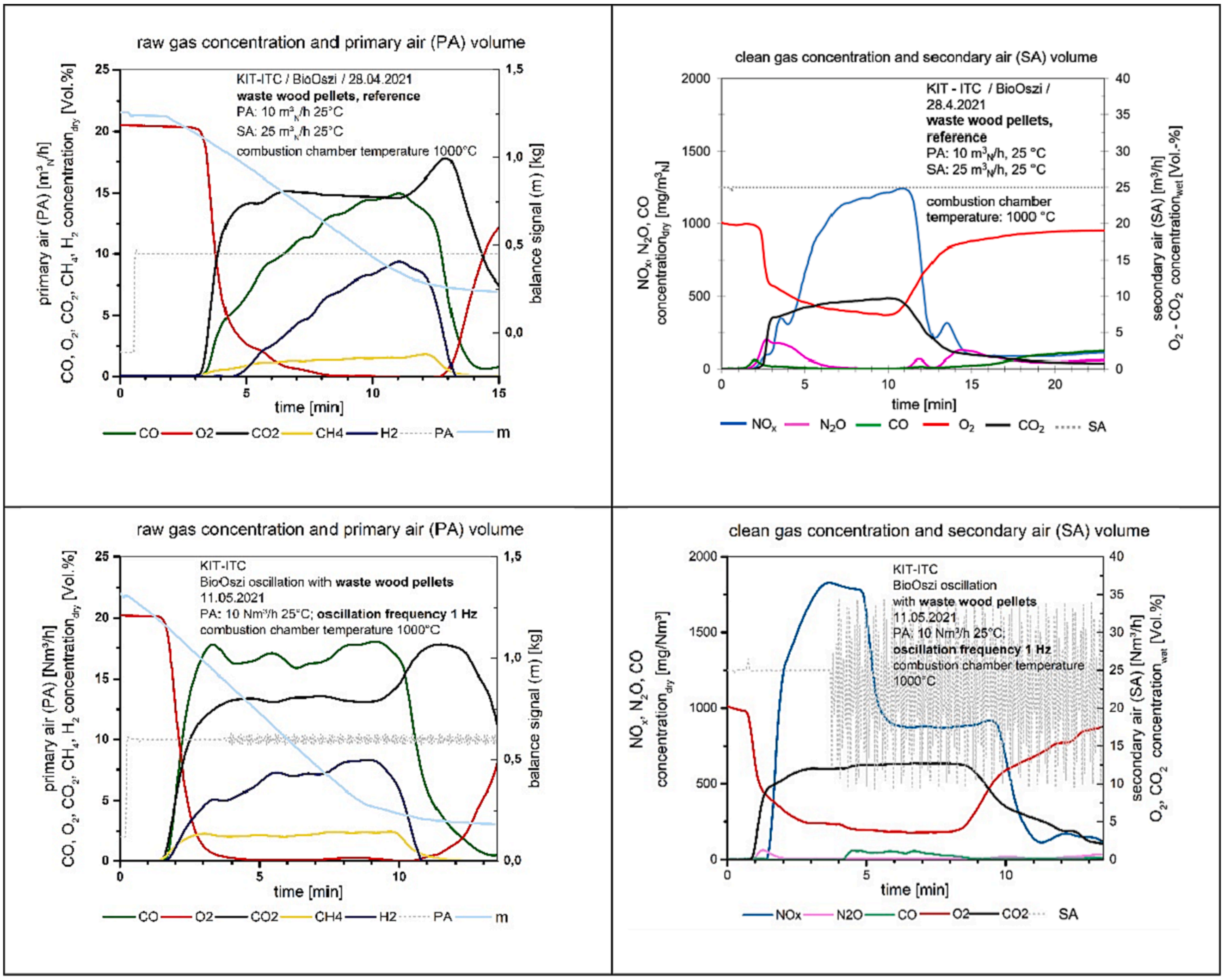

Fig. 9. Experimental results by non-oscillating (top) and oscillating combustion (bottom) of waste wood pellets at KLEAA.

oscillation 1 Hz (bottom), respectively. Wood pellets have a significantly lower fuel *N*-content (0.12 wt%) than the waste wood (4.1 wt%) as indicated in Table 5.

The general trend of the evolution of gas concentrations and fuel bed mass loss are very similar to the waste wood, and, hence, are not discussed here further for brevity.

However, it is important to note that without oscillation, roughly 180 mg/m_N^3 as referred to 11 vol% O_2 are measured, with oscillation approx. 125 mg/m_N^3, which corresponds to reduction rate of 30 %. Applying the equation for the conversion rate CR, it is interesting that with much lower nitrogen content in the fuel, the CR is higher: In the mode without oscillation the CR is about 35 %, with oscillation a reduction to 25 % is achieved.

3.3. *Grate furnace*

In order to evaluate the measured gas concentrations a mass balance based on the measured volume flows was calculated. For the mass balance the "Input" mass flows (sum of the primary air, secondary air and cooling air amounts, as well as the fuel) were compared with the "Output" flows (sum of the flue gas and ash quantities). The comparison for a couple of experimental times is represented in Table 6.

The represented values are average of up to 5 min. Since the working mode of the boiler depends strongly on the load, averaging of the values for a longer time periods are of limited sense. Considering possible error sources (amount of cooling air, leakage air amount and errors of the measuring equipment) the error of the balance has a good value of maximum 24 %. As already mentioned, the facility depends on the load i.e., for supply of hot water for heating and use. Due to this reason, there is no constant operation for longer time periods as for instance of 3–4 h. Therefore, in the following discussion next to the "classical" representation of NO-concentrations referred to 11 vol% oxygen in the flue gas in mg/m^3, a load referred NO-concentration in mg/kJ will be introduced. This translation is done as the NO-concentration referred to 11 vol% O_2 is multiplied by the mass flow of the flue gas and consequently referred to the thermal output of the facility (product of fuel mass flow and heating value). Fig. 11 represents the change of the NO-concentration for one setting without a pulsator and with pulsator with a frequency of 0.75 Hz.

Due to the operation of the pulsator, which interrupts the secondary air with a frequency of 0.75 Hz, a reduction of roughly 4 % can be achieved. It needs to be considered that an important prerequisite for a higher reduction efficiency, the non-stoichiometric combustion of the fuel on the grate with primary air is not presented, as seen in Fig. 11. The primary air ratio λ_p is determined by the ratio of the primary air amount and the product of the minimum required air quantity (5.15 kg air/kg fuel) and the fuel amount. If $\lambda_p < 1$ (gasification), then one significant requirement for the functioning of the oscillating combustion is fulfilled. Additionally, the ratio of the primary air to the total air from primary and secondary air is represented (see Fig. 12). It shows where the main

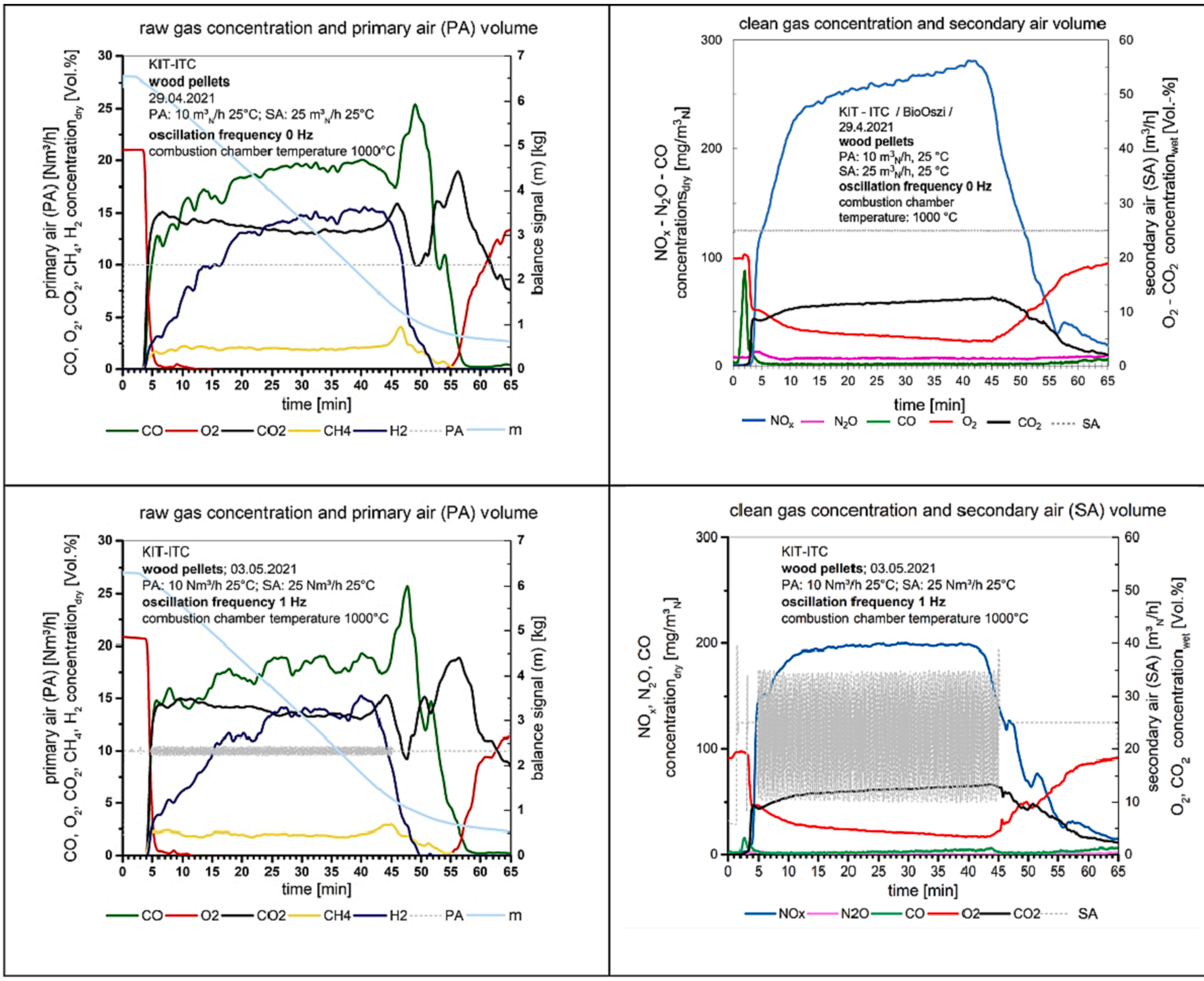

Fig. 10. Experimental results of wood pellets combustion without oscillation (top) and oscillating combustion (bottom) of wood pellets at KLEAA.

Table 6
Mass balance and balance error for selected experimental times.

parameter	Date/Time					
Mass flow [kg/h]	7.3.; 13:35	7.3.; 13:55	7.3.; 14:10	8.3.; 11:05	8.3.; 11:30	8.3.; 12:35
Fuel	108	111	103	139	94	139
Primary Air (PA)	900	786	897	960	868	757
Secondary Air (SA)	386	315	379	265	265	436
Cooling Air	130	130	130	130	130	130
Input	**1,524**	**1,342**	**1,509**	**1,494**	**1,357**	**1,462**
Fluegas	1,703	1.661	1,689	1,441	1,601	1,451
Ash	2	2	2	3	2	3
Output	**1,705**	**1,663**	**1,700**	**1,443**	**1,603**	**1,454**
Difference	181	322	191	−51	246	−9
Deviation [%]	12 %	24 %	13 %	−3%	18 %	−1%

air quantity is supplied.

The primary air ratio is not < 1 for the whole investigation time period considered, which means that there are no gasification conditions on the grate, while the main air is supplied through the primary air. The ratio is between 70 % and 75 %. Furthermore, small effects can be seen. The reduction results probably, due to local reducing conditions, which cannot be pictured by the averaging here. If referred to the thermal output, the NO-concentration is reduced by 6 % in case of oscillation mode in comparison with the non-oscillation mode of operation.

On the second day of experiments different pulsator requirements were set and it was attempted to shift the air ratio in favor of the secondary air.

Fig. 13 shows the NO-concentrations in mg/m^3, and the conversion rate of the fuel bound nitrogen on the secondary y-axis.

If the greatest difference between the NO-concentrations without and with oscillation is considered, the maximum NO-reduction from 340 mg/m^3 to 300 mg/m^3 is about 12 %.

Despite the shifting of the primary and secondary air it was possible to achieve gasification conditions on the grate only for short periods, as can be seen on Fig. 14.

In the short intermissions with gasification conditions the NO-concentration referred to the energy content decreases significantly, as shown by the yellow arrows. Unfortunately, these time intervals are too short to be able to compare systematically settings without oscillation and with oscillation. By the initial air distribution with approx. 75 % fraction of the primary air and oscillation of 0.75 Hz the NO-concentration decreases from 0.112 down to 0.109 mg/kJ, which corresponds to a reduction of about 3 %, but it lays well in the “background” noise of the measurement accuracy. By the setting with reduced primary air amount of about 65 % comparable time periods without and with oscillation with a primary air number $\lambda_p < 1$ does not exist.

Fig. 15 shows an image of the fuel bed on the grate for the setting

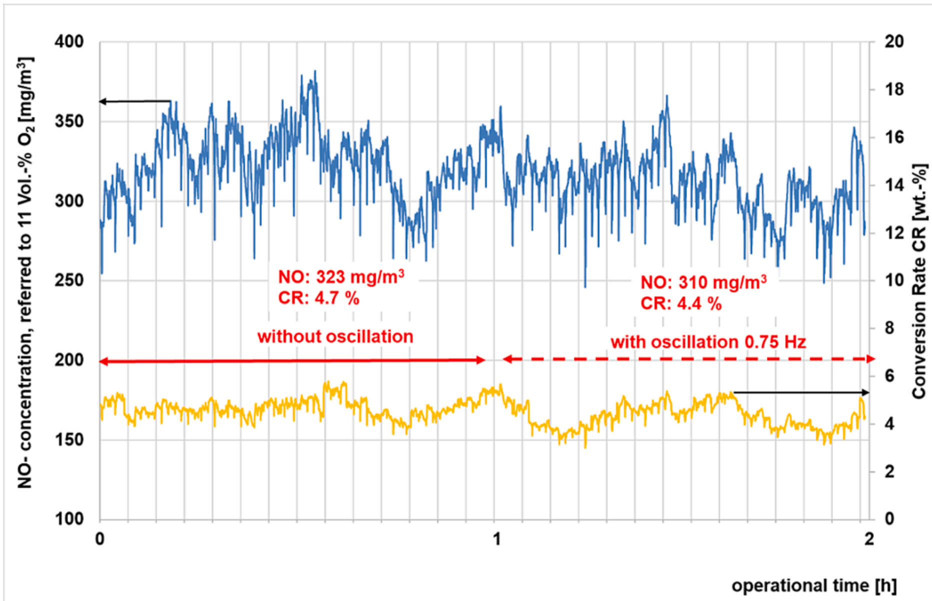

Fig. 11. NO-concentrations and conversion rate for experiments without and with oscillation.

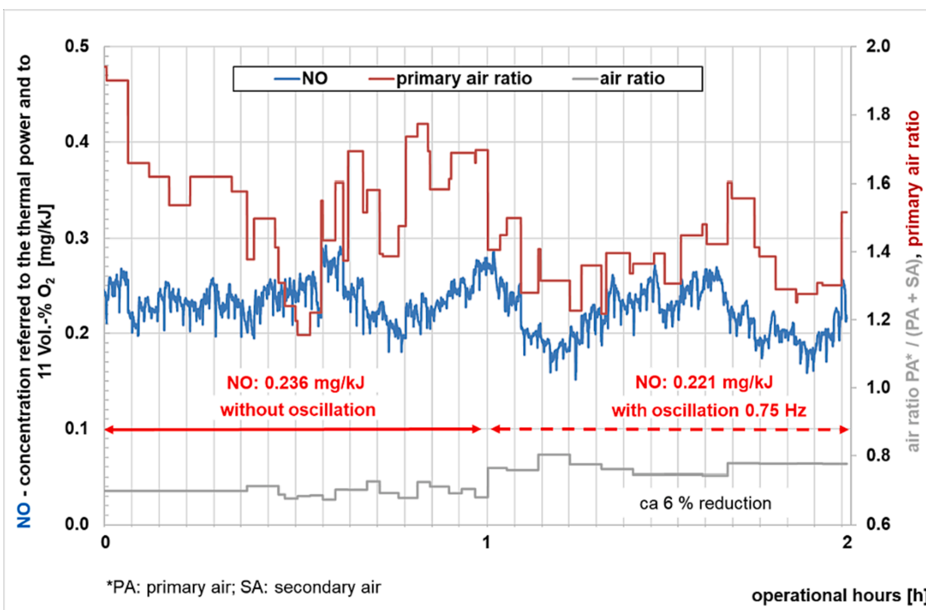

Fig. 12. NO-concentration referred to the thermal power, primary air ratio and air ratio.

with reduced primary air amount.

The reduction of the air quantity results obviously in a minimized heat release, which not only promotes the slag formation, but also reduces the solid matter conversion. The facility disadvantage is apparent here, since it is not equipped with an ignition arch, which stores the released heat and emits it again. In general, the process is not possible to be fully implemented on the facility at Polzenith, since significant components in the plant i.e., ignition arch, spatial separation of primary conversion and gas burnout are not available in a sufficient manner. Nevertheless, NO-concentrations could be reduced by the shifting of the primary and secondary air amounts, which needs to be investigated, while the plant is in operational mode.

3.4. Comparison of fuel types

To compare the different types of fuels and furnaces the effect of the NO_X reduction due to the oscillating operating mode the conversion rate of the fuel bound nitrogen (see Equation (5)) of the different combustion facilities (BRENDA, KLEAA and the grate furnace of POLZENITH) is shown for the used fuel types in Fig. 16.

In all test plants, by applying an oscillating addition of the secondary air, a reduction of the conversion rate for the species N from fuel bound nitrogen to NO_X in the flue gas could be achieved. The maximum reduction potential of the conversion rate of about 35 % can be observed for hard coal. The firing of the waste wood (as delivered) in the grate furnace shows the minimum of the reduction potential of the conversion rate with 10 %. The minimum value for the conversion rate, an interesting observation of the results is that the reduction potential of the

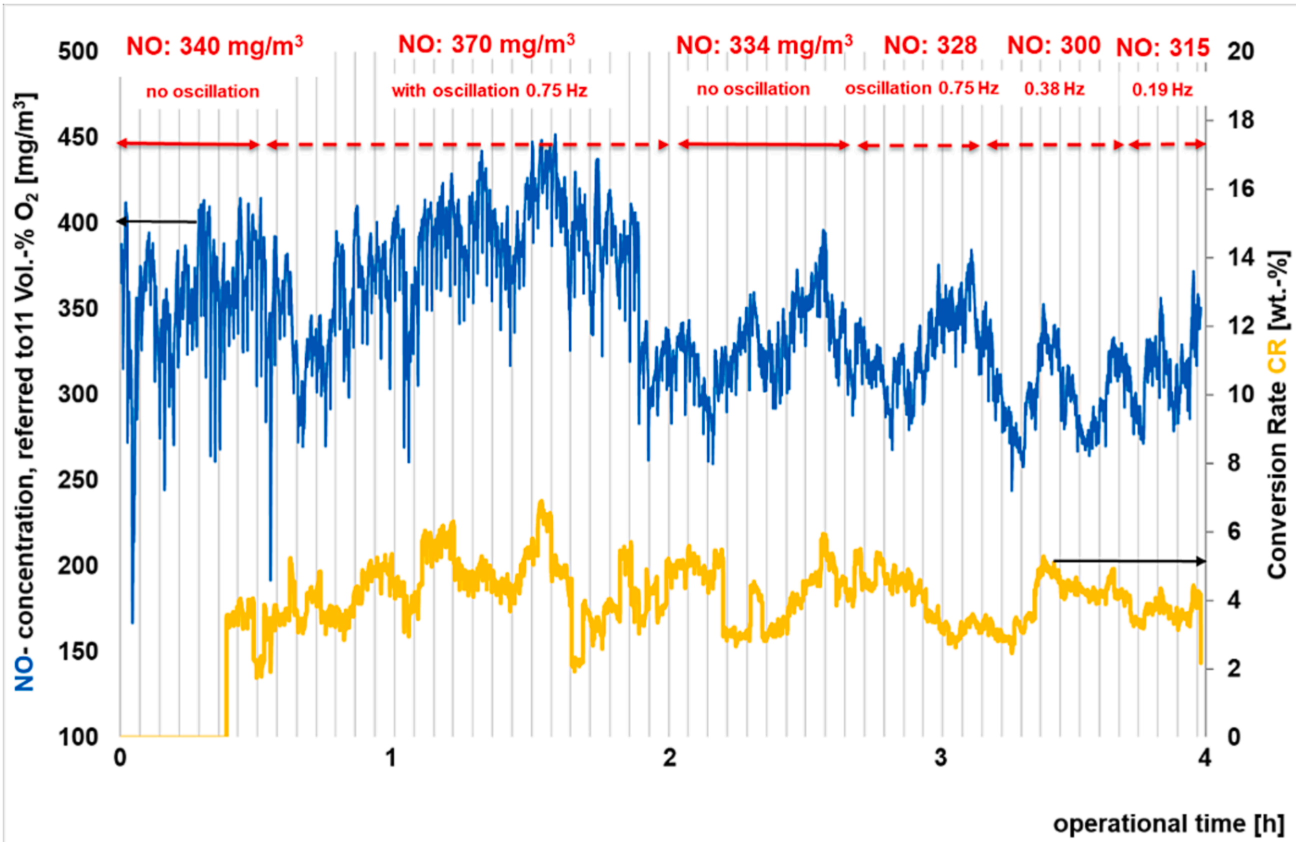

Fig. 13. NO-concentrations and conversion rate for experiments without and with oscillation.

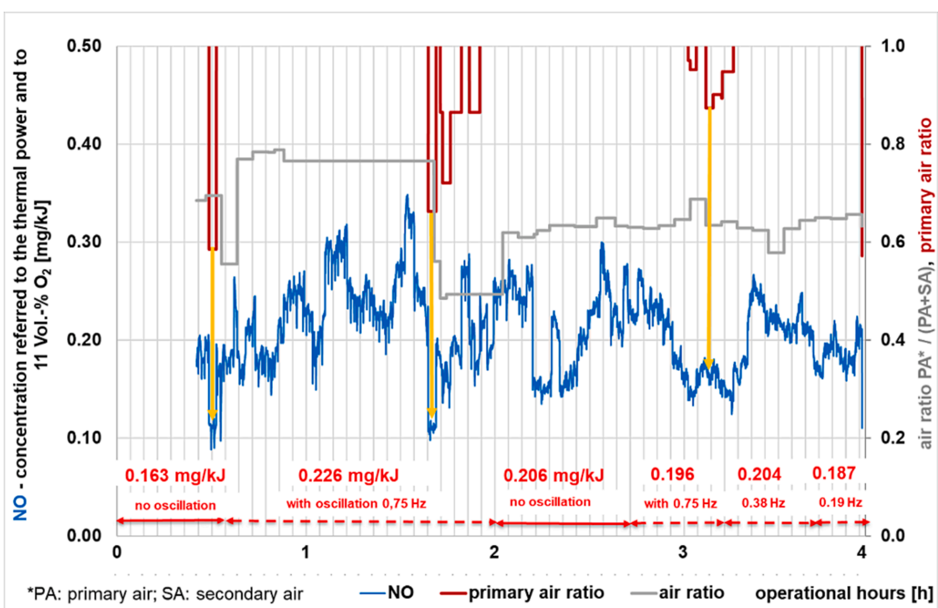

Fig. 14. NO-concentrations referred to the thermal power, primary air number and air.

conversion rate stays constant at around 30 % for hard coal, lignite, and the biomass-based fuel types despite their different fractions of fuel bound nitrogen.

4. Discussion

As shown above the oscillating combustion with the hard coal dust burner at BRENDA enables reduction of NO_X emissions with either fuel, hard coal or lignite. The oscillating combustion of hard coal with a nitrogen content of 1.6 % resulted in a reduction of the nitrogen oxide concentration in the flue gas by 34.8 % from 492 mg/m_N^3 down to 321 mg/m_N^3.The results correlate with the previous studies on oscillating combustion at the pilot plant BRENDA [2,3]. With the oscillating combustion of lignite with a nitrogen content of 0.65 %, a reduction of the

Fig. 15. Fuel bed of investigated grate furnace.

nitrogen oxide concentration in the flue gas by 17.7 % from 384 mg/m^3_N down to 316 mg/m^3_N could be achieved. Thus, the feasibility of nitrogen oxide reduction by oscillating operation in lignite grading with the pilot plant BRENDA could be demonstrated.

The investigation of the local stoichiometric air number shows that the increase of the oscillated air impulse which is responsible for the essential interference of the hard coal dust leads to a NO_X reduction (see Fig. 7). It is striking that for a lower air number both curves converge and there is no difference between the two operating modes. The explanation can be derived from the proportion of the oscillated added air in comparison to the total combustion air of the coal burner. From a local stoichiometric ratio of<0.31, the proportion of oscillated added air is only<15 % of the total combustion air entering through the burner. The remaining impulse through the air flow shall be reduced, in order to create the changing reaction conditions characteristic of the oscillating combustion.

The results of the study on the influence of the swirl number show no positive effect of the oscillating air addition on the nitrogen oxide concentration (see Fig. 8). Consistently higher values of NO_X concentration for the oscillating combustion from a swirl number < 0.81 are observed. Assuming that at low swirl numbers the oscillating air addition influences the flame stability this behavior can be easily described. Similarly, to the case of the local stoichiometric air number, the oscillated air content is only 17 % of the total air flow supplied via the burner. The positive effect of oscillating combustion becomes clear only from an oscillating air content of about 30 % [7].

Based on the investigations on the dust firing, a transfer of the primary measure oscillation to a grate firing could also be demonstrated. In contrast to dust firing, where both, the fuel and part of the combustion air were oscillated only oscillation of the secondary air is sensibly possible in the two-stage laboratory reactor as well as in the small grate firing system of the POLZENITH company.

In the KLEAA laboratory plant, model fuels with different nitrogen contents were used to ensure the boundary conditions necessary for the effect of the reduction. These include:

- The formation of stable gasification in the first stage for the conversion of the fuel.
- A secondary air supply that can post-combust the raw gas locally separate from the first stage

The pelletisation of the waste wood, which was used untreated in the grate firing system of POLZENITH, was necessary for the laboratory plant to ensure uniform combustion in the first stage, which can be seen in Fig. 9, top left and bottom left, from the mass decrease and the course of the gas concentrations. By starting oscillation, the nitrogen oxide concentrations can be lowered significantly. For future tests, however, the oxygen content, which decreases on average due to the interruption of the secondary air in the flue gas (comparison of Fig. 9, top right diagram with the bottom diagram), should be balanced out again by a higher inlet pressure in the secondary air, so that the oxygen content in the flue gas is approximately the same. The oscillation frequency is measured by the remaining CO concentration in the flue gas, which is at the same low level during oscillation as without oscillation. This frequency varies from plant to plant, as the explanations for grate firing show. The nitrogen content of the fuel here (4 wt- %) is of the same order

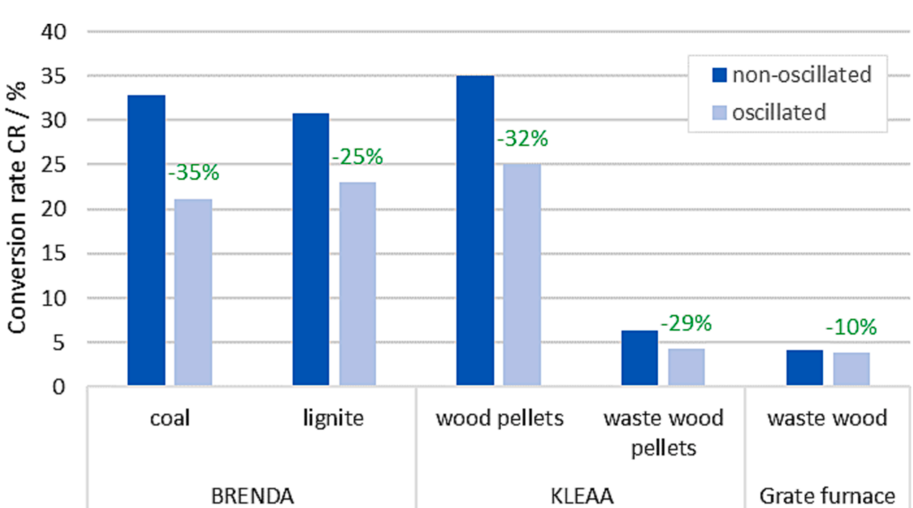

Fig. 16. Fuel bed of investigated grate furnace.

of magnitude as the nitrogen content of hard coal (1.6 wt- %), the natural pellets used here with 0.12 wt-% are of the same order of magnitude as lignite with 0.65 wt- % nitrogen. In both cases, the question was, whether oscillation can also lead to a significant reduction in NO_X concentrations for fuels with a lower nitrogen content. The question can be answered with"yes" in both cases, for the lignite and for the untreated natural wood pellets the NO_X concentrations can be reduced, however, the reduction rate of about 18 % for the lignite and 30 % for the untreated wood pellets is lower than with higher nitrogen contents in the fuel. This can presumably be explained by the higher conversion rate to CR with higher nitrogen content in the fuel, which is known from the"Fenimore curve" for the non-oscillated case [12]. The curves of the gas concentrations in the raw gas (Fig. 10, top left and bottom) reflect the uniform gasification of the fuel in order to achieve the prerequisite for a good reduction of the oscillating secondary air. From Fig. 4 showing the schematic structure of the grate firing system at POLZENITH, it can be seen that there is no spatial separation of primary zone and secondary zone as in the laboratory reactor (Fig. 3): The secondary air is blown into the flames from the combustion bed directly above the grate; thus, one of the prerequisites for successful oscillation is not or insufficient fulfilled. In addition, the primary air cannot be added in a controlled manner along the length of the grate because there is only one grate zone. Thus, the primary air seeks"the path of least resistance" according to the local pressure conditions on the grate and thus does not enter the fuel in a defined way. To characterize the system as a whole and to obtain reliable data for determining the primary air rate, appropriate measurement technology was first installed to enable a mass balance to be drawn up. However, within the limits of the measurement inaccuracies, the results can be considered satisfactory according to Table 6. The pulsator, the design of which is a trade secret, is installed here outside the grate system and interrupts all the air supplied to the kiln; the air is then distributed via channels to the left and right along the length of the grate. Unfortunately, it was not possible to check to what extent the impulse of the interruption arrives in the kiln. Since the system is heat-controlled, the output and thus the fuel and air quantities change permanently with constant fuel and air distribution ratios. Therefore, the profiles shown here must always be related to the output (Fig. 13) to establish comparability of the NO_X concentrations. With a trimming of the primary air in favor of the secondary air (from about hour 2 in Fig. 13) from originally 75/25 to 65/35, it was nevertheless only possible to achieve sub stochiometric conditions in the combustion in very short periods of time. Therefore, the effect of oscillation, e.g., at 0.19 Hz, is only about 9 % lower than the value without oscillation. Due to the less good aeration of the fuel on the grate, fuel accumulation occurred, so that this setting was taken back again. For a transfer of the good results achieved in the laboratory reactor, a plant must be selected in a follow-up project in which the primary and secondary zones are spatially separated from each other, and which allows stationary operation for several hours.

A comparison of the different fuels with different nitrogen contents shows that oscillating combustion equally leads to a reduction of the conversion rate of species N from fuel-bound nitrogen to nitrogen oxide by 25 – 35 % for all used fuel types. The oscillating operational mode thus favors the N_2 formation mechanisms, whereby about a third of the fuel-bound nitrogen reacts to N_2 under oscillating operational conditions instead of NO_X under non-oscillating operational conditions. Exceptions here are the results of the grate furnace, where a reduction of the conversion rate of species N from fuel-bound nitrogen to nitrogen oxide of 5 % was achieved due to the above-mentioned setup limiting conditions.

The optimization potential of the considered test facilities shows differences due to the different technical equipment and the possible operation of the respective system. At the two test plants BRENDA and KLEAA, a high NO_X reduction potential with oscillating combustion for hard coal and lignite as well as for biomass-based fuels such as wood pellets and waste wood pellets was demonstrated. On the other hand, in the grate furnace of untreated waste wood, a comparatively low optimization potential with regard to the nitrogen oxide concentration in the flue gas was identified. To compile, a reduction in the nitrogen oxide concentrations in the flue gas could be achieved in all combustion systems under oscillating operational conditions.

5. Conclusion

Nitrogen oxide emissions contribute significantly to air pollution and global warming and must be reduced from combustion processes in accordance with legal regulations. Primary measures that directly intervene in the combustion process are the first choice for optimized operation to save or reduce secondary reduction measures. In addition to locally resolved fuel and air staging, primary measures also include the oscillating addition of fuel and air as a time-resolved staging alternative. The aim of this work was the investigation of the NO_X reduction potential of oscillating combustion using various fuel types and different combustion technologies. In this paper, results from dust firing of hard coal and lignite, as well as waste wood and natural wood pellets in a grate firing system are presented. It is shown that nitrogen oxides can be reduced by up to 35 % through time-resolved fuel and air staging. However, ensuring a sub-stoichiometric first stage is a prerequisite for a high reduction.

CRediT authorship contribution statement

Nicklas Jolibois: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Writing – original draft, Visualization. **Hans-Joachim Gehrmann:** Conceptualization, Methodology, Validation, Formal analysis, Investigation, Resources, Writing – original draft, Visualization, Supervision, Project administration, Funding acquisition. **Krasimir Aleksandrov:** Conceptualization, Methodology, Validation, Formal analysis, Investigation, Writing – original draft, Visualization. **Manuela Hauser:** Validation, Investigation. **Dieter Stapf:** Resources. **Bo Jager:** Validation, Formal analysis, Visualization. **Siegmar Wirtz:** Validation, Investigation. **Viktor Scherer:** Resources, Writing – review & editing. **Gregor Pollmeier:** Validation, Investigation, Resources. **Philipp Danz:** Validation, Investigation. **Jorg Matthes:** Writing – review & editing. **Markus Vogelbacher:** Writing – review & editing. **Patrick Waibel:** Writing – review & editing.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Hans-Joachim Gehrmann reports financial support was provided by Federal Ministry of Education and Research Bonn Office.

Data availability

Data will be made available on request.

Annex

The temperatures are located in the fixed bed, starting with T2 from the top to T13 near to the bottom.

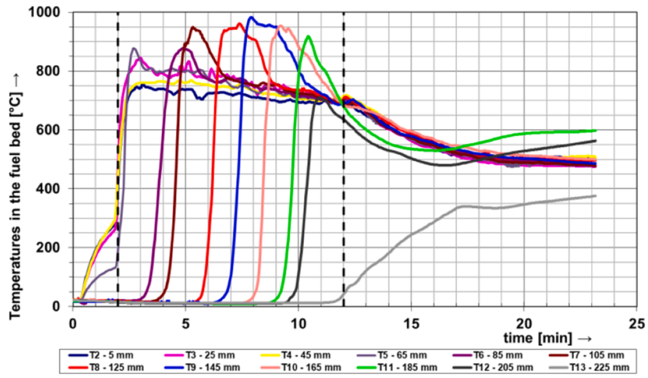

Fig. A1. Temperature profile of combustion of waste wood pellets, non-oscillating mode.

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