Experimental and Numerical Investigation of NO_x-emissions by a Biomass Combustion With Oscillating Secondary air

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Abstract

With the Paris Climate Agreement EU aims to reduce the emissions of greenhouse gases by at least 40% by 2030 compared to 1990. In order to achieve this goal the importance of biomass as a solid, lumpy fuel increases steadily. In Germany the substitution of fossil fuels by biomass in heat generation saves 15.7 Mio.t CO_2-equivalent yearly. However, the combustion of biomass results in emissions of NO_x, fine particles and CO. Since these emissions equally harm humans and environment, their limit values are regularly tightened by the legislature. Hence, in order to minimize the emissions, new and innovative solutions are needed.

One possible primary measure for NO_x-emissions reduction is the oscillating supply of the secondary air in a staged combustion, which was applied in this work.

With the help of this technological approach the study targets the reduction of NO_x-emissions down to concentrations below the required maximum permissible values according the German emission standard for clean air.

At the Institute for Technical Chemistry (ITC) experiments in a batch-operated reactor have been carried out, in which the combustion behavior of solid fuel discharges as well as the conversion of the formed flue gas under different oxidizing conditions were investigated. If the fuel is burnt sub-stoichiometric in the primary stage, through oscillation of the secondary air supplied to the post-combustion chamber above the fuel bed, the NO_x-concentration can be reduced by as much as 40% compared to the non-oscillating operation mode. However, this requires homogeneous conversion of the solid fuel during the primary stage.

Complementary to the experiments 3D-combustion modelling of the fuel transformation and nitrogen oxide formation after the secondary air supply to the post-combustion chamber of the reactor have been carried out at LEAT. For the NO_x-formation modelling the Eddy-Dissipation-Concept (EDC) combined with In-Situ-Tabulation (ISAT) and a detailed kinetics were applied. The NO_x-formation in the lab-reactor was successfully simulated and the results overlap well with the experiments.

Introduction

In order to act against the climate change the shift away from fossil energy sources is inevitable. In this context the lower and the upper house of the German Parliament scheduled the stepwise withdrawal from the coal power plants until 2038 [1]. Consequently, the importance of climate neutral energy carriers, as for example biomass, increases. In Germany through the combustion of solid biomass 167,000 TJ heat are generated every year, while thereby 15.7 Mio.t CO_2-equivalent of greenhouse emissions less are released if compared with fossil energy sources [2]. However, by the combustion of biomass do form emissions, which are harmful for human and environment i.e NO_x, fine dust and CO. For this reason maximum permissible values (limiting values), dependent on the thermal output of the facility have been ordered by the legislature. For firing plants with a thermal output > 1 MW the German clean air regulation decisive, which includes tightening of all limiting values, as of Fall 2021 [3]. Due to this tightening all existing plants must be upgraded with corresponding technologies for emissions reduction in order to avoid the danger to be taken out of operation.

For the reduction of NO_x-emissions well-established processes, such as SCR and SNCR are existing on the market already. Both technologies belong to the so-called “secondary measures” – after NO_x is formed it is decomposed by the addition of a reducing agent. These processes are particularly suitable if the formation of NO_x is unavoidable i. e. if nitrogen-rich biomass (> 1 wt. %, i. e. chipboards) is incinerated. Disadvantage of both technologies are the high investments and operational costs, which are too much for smaller facilities with an output of 20 MW and below.

Primary measures prevent the NO_x-formation during the combustion, as with the help of design measures the oxidant supply, temperature and residence time are influenced. Nonetheless, the existing facilities have been installed under different circumstances and even after a changeover are not in condition, if possible at all, to align with the tightened limiting values. Furthermore, these measures can hardly influence NO_x-formation due to nitrogen contained in the fuel.

Oscillating supply of the secondary air in a two-stage combustion is another option for NO_x-reduction and investigated in this work. Through the oscillation temporally change in the local stoichiometry takes place, whereby reduction of NO_x-emissions can be achieved [4-8]. The advantage of this technique is that it can be implemented in already existing firing plants and is likely more cost-efficient than the alternatives mentioned above.

With regard to the potential use of this approach in biomass firing in the course of this study, experimental investigations in a batch-operated reactor (KLEAA) at the KIT are carried out. Biomass pellets of waste wood with an elevated nitrogen content are utilized as a fuel. Two-stage combustion takes place inside the reactor, whereby in the second stage, the flue gas combustion, the air is supplied oscillating. The potential of NO_x-reduction for different frequencies of oscillation are studied.

Complementary, CFD-simulations of the flue gas combustion in KLEAA are carried out. The modelling have to add to the better understanding of the relationships between transient air supply and NO_x-reduction. In a case of a good agreement with the experiments the modelling approach can be transferred to larger facilities and the reduction potential of the oscillating method within the plant can be estimated.

Experiments at the lab-reactor KLEAA

The experimental investigations started with the raw material, which have been accumulated from POLZENITH. For better correlation of the combustion behavior without and with oscillation, the waste wood was pelletized. The experiments represented here show the results of the waste wood pellets. At first, the experimental set-up will be described briefly, followed by the fuel characterization and explanation of the experimental schedule. Selected experiments will be discussed.

Design KLEAA

The experimental facility KLEAA represents a fixed bed reactor, in which the combustion behavior of solid, lumpy fuels is characterized (Figure 1).

The facility comprises three units: a movable combustion chamber, where the fuel is filled, a post-combustion chamber, where the gases from the combustion chamber oscillating or non-oscillating, respectively are post-burned and a flue gas cleaning system. At first, the flue gases are cooled down to approx. 140°C in a boiler, followed by dust removal by a baghouse filter and finally cleaned by an activated coal adsorber. The chemical composition of the gas is determined from the raw gas and after the baghouse filter. The water condensates from the raw gas and after the baghouse filter are analyzed for NH_3 and HCN. The mass flow of the raw gas can be determined from the balance signal. Evaluation of the ignition behavior is realized with the help of thermocouples ordered in a shifted way inside the combustion chamber. These for the simulation necessary starting parameters have been determined experimentally.

The gas concentrations for stationary, non-oscillating condition after the secondary air supply were compared with the experimentally determined values. The mixing condition of secondary air with the flue gas were not changed. The post combustion of the flue gases from the primary stage was reached with 25 m^3/h stable secondary air injection in all cases. To vary the mixing conditions in future and keep the air ratio constant, the diameter of the nozzles has to be changed to influence the momentum.

Since, the water-vapor concentration was not measured experimentally it was determined from an element balance.

Operation mode and used fuel

The fuel – in this case pellets with a length of approx. 1 cm and a diameter of ca. 6 mm is filled inside the cold combustion chamber, which afterwards is pushed under the already preheated to 1000°C top part of the combustion chamber. The primary air is supplied with 10 m^3_N/h. According Table 1, characteristic for the use fuel is the high Nitrogen content of 4.09 wt. %. The secondary air is adjusted at 25 m^3_N/h. After a short drying period the released volatile components ignite with the air oxygen and the radiation energy from the electrically heated combustion chamber, the combustion process begins, but the electrical heating continues to operate. The combustion front migrates in the opposite direction of the primary air towards the bottom 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.%. The combustion process is optically evaluated by the means of a video camera.

Results

The time sequence of the gas concentrations, as well as the measured clean gas concentrations are summarized in Figure 2. The graphics on the left side show the measured gas concentrations above the fuel bed, the supplied primary air amount and the balance signal. The uniform burn-off of the pellets in both experiments can be well distinguished. The graphics on the right side show the clean gas concentrations after the supply of the secondary air, both graphics at the top the experiment without oscillation, the bottom ones with oscillation. The values for HCN and NH_3 are averaged for the time of the degasification. At first, the fuel is heated, dried and degasified. After 3 minutes the volatile components ignite and the reaction products CO_2, CO, CH_4 and H_2 increase, while O_2 is consumed (Fig. 2 top left). Henceforth, in the direction of the bottom grate every following fuel layer reacts in the same manner. After 8 minutes no free O_2 is available anymore, so that increasingly CO is formed, but it can't however react further to CO_2. Unreacted char from the pyrolysis can react with water vapor from the lower fuel layers to H_2 and CO, consequently, H_2-concentration increases. After approx. 11 minutes the ignition front reaches the bottom grate i. e. degasification is over and CH_4 as well as H_2 decrease, O_2 increases again, the formed CO can further react to CO_2 and the char finally burns out. 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-conceantration increases and reaches a peak after approx. 11 minutes, 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. NH_3 reacts with oxygen from the secondary air to NO_x, the nitrogen from the char generates between 13th and 14th minutes additional small peak and it correlates with the maximum in CO_2 concentration above the fuel bed. The peak of N_2O at the beginning of the combustion (3 minute) 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 4th minute is about 850°C.

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 [8]. With the increase of the released hydrogen N_2O is reduced again.

By the experiments with oscillation the flue gas mass flow from the fuel bed is with 0.0020 kg/s somewhat larger than that of the experiments without oscillation (0.0015 kg/s). Thus, by constant primary air supply the concentration level of the unburnt components such as CO and CH_4 increase, while the CO_2 content is comparable. The higher release rate is obvious from the NO_x level at the clean gas side: its maximum is at 1800 mg/m^3_N NO_x, while the maximal value for the experiment without oscillation is 1250 mg/m^3_N NO_x. By the application of oscillation, NO_x can be significantly lowered down to 880 mg/m^3_N 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. The clean gas concentrations are not normalized.

Certainly, the application of oscillating combustion air is suitable for fuels with a high nitrogen content, as in the case shown here.

The application of the wood pellets, which are usually used in firing with pellets and have a significantly lower nitrogen content of 0.12 wt.%, according Table 1, show however that this operation mode is suitable for Nitrogen-poor fuels as well.

The corresponding results are shown in Figure 3 for raw and clean gas without oscillation (top) and for the experiment with oscillation (bottom), respectively.

Due to the homogeneous fuel (pellets) the raw- and clean gas concentrations, except that of H_2, run very smooth and constant, once the ignition starts. By H_2, as already described above, the heterogeneous water gas reaction plays a role. As the part of the unburnt char from the top layers pyrolysis increases it reacts with water vapor from the gasification, combustion and drying of the bottom layers, as well as with C to CO and H_2. Hence, the CO-content of the raw gas increases continuously. Here too, the different stages are easily distinguished: heating, drying and ignition, followed by quasi-stationary burn-off stage and char burnout with a CO-peak at the 48th minute.

Without oscillation, roughly 180 mg/m^3_N as referred to 11 vol.% O_2 are measured, with oscillation approx. 125 mg/m^3_N, which corresponds to reduction rate of 30%.

CFD simulation

Combustion modelling

The simulations focus on modeling the combustion of the flue gas in the post combustion chamber (pcc) and the formation of NO_x under oscillating and non-oscillating secondary air (SA) supply. The domain of the simulation is depicted in Figure 4. Flue gas from the first stage enters the pcc via a connecting channel. Secondary air nozzles are installed radially at the top of the pcc and shortly beneath the connecting channel. The flue gas burning and NO_x formation occur in the top half of the pcc where air and flue gas are mixed. As the models used in the modelling approach are computationally expensive, the simulations were run in parallel and the domain was restricted to the depicted top half of the pcc. Therefore, the burned flue gas outlet lies at half of the pcc's actual height. In total, the mesh contains approximately 120,000 hexahedron cells.

The simulations are run with the commercial CFD solver from ANSYS Fluent (17.2). Table 2 lists the physical and chemical models used in the modelling approach. The turbulent flow is modelled with the Detached Eddy Simulation (DES) Modell and the SIMPLEC algorithm is used for the pressure velocity coupling. Radiative heat transfer between walls as well as wall and gas is modelled with the Discrete Ordinates Model (DOM) using a 3x3 discretization of the angular space. Absorptivity of the gas is calculated with the WSGGM model [13]. To model the interaction between turbulent mixing and chemical reaction the Eddy Dissipation Concept (EDC) [9] is utilized. Integration of the reaction rates is carried out by the ISAT algorithm [10]. Application of EDC-ISAT enables the modelling of "slow" chemistry like NO_x formation. These reactions occur on time scales of order 10-2 to 100 and are therefore much slower than for example the combustion reactions of carbon hydrogens. The "LI32" with 32 species and 144 elemental reactions [11] is used as kinetic mechanism. More elaborate mechanisms like the one from Glarborg [12] were deemed as to computationally expensive for the simulations.

Boundary conditions are derived from the experimental conditions of the KLEAA. The secondary air (SA) flow rate over time for the oscillating case is idealized into a sinusoidal progression (Fig. 5). For the non oscillating case the SA volume flow rate is constant at 25 m^3_N/h. To achieve NOx reduction in the clean gas an oscillation frequency of 1 Hz was used with an average volume flow of 17.5 m^3_N/h and an amplitude of 7.5 m^3_N/h. Fig. 4 also depicts the corresponding total air fuel ratio in the pcc. It is evident that the overall combustion process is strongly overstochiometric in the non oscillating case. Consequently, local combustion conditions are strongly oxidative which is unfavorable for NO_x reduction. By oscillating the SA, the total air fuel ratio periodically approaches a value of one while the local value of the air fuel ratio in the pcc even drops beneath one. Therefore, reductive conditions in the pcc are present and NO_x will be reduced.

Results

Fig. 6 shows the NO_x concentration at the flue gas outlet of the pcc for both, the oscillating and non oscillating case. The NO_x concentration for the non oscillating case is at 980 mg/m^3_N-dry (11 Vol.-% O_2) at all times because of the stationary behavior of the combustion. In case of oscillating SA, the NO_x concentrations quickly drops to about 680 mg/m^3_N-dry approximately 0.2 s after the start of the oscillation. The NO_x curves progression is slightly delayed compared to the SA flow rate curves progression because flue gas and air have to spatially mix first before the reduction zones, in which NO_x is reduced, are formed. With increasing SA flow rate after 0.5 s, the NO_x increases as well because the reduction zones grow smaller. Also, the NO_x curves progression starts to deviate stronger from the sinusoidal curves progression because a higher mixing effect is induced by the oscillation and the already formed reduction zones are mixed with zones of (again) higher stoichiometry. The NO_x concentration increases up to 940 mg/m^3_N-dry, which is slightly lower than the initial concentration. From then onwards, the NO_x concentration only decreases slightly and not as strong as at the beginning. The highly turbulent mixing induced by the oscillation probably prevents local under stoichiometric conditions similar to those that occurred at the beginning. Nonetheless, the oscillation leads to a reduced concentration of NO_x in the burned flue gas of approximately 820 mg/m^3_N-dry. It is expected that this concentration will decrease further with lower local stoichiometry and larger sub-stoichiometric zones. Such conditions can be achieved, for example, with a smaller average volume flow rate of the secondary air.

Discussion

In Table 3 the gas concentrations of O_2 and CO_2, dry state, for the non-oscillating operation mode are compared. The somewhat higher part of CO_2 in the simulation is probably due to a too high CO-amount in the raw gas above the fuel bed compared to the experiment. There is a good agreement between experiment and model calculation of the O_2 concentration. Concerning the NO_x concentration the simulation and the experiment show a satisfying agreement. In non-oscillating operation mode the experimental NO_x concentration is about 1250 mg/m^3_N, while the simulated one has a value of 980 mg/m^3_N. The lower simulated concentration is mainly due to two reasons: on the one hand the water content of the raw gas is too high, because too moist pellets were assumed. A lower water content in the raw gas will result in a higher NO_x concentration in the simulation and consequently, in a better match with the experiment. On the other hand the NH_3/HCN concentrations in the raw gas were determined by the means of an empirical release model [14]. Higher NH_3/HCN concentrations in the raw gas in the simulation will lead additionally to a better fit with the experiment.

In oscillating mode the simulation confirmed basically the trend observed in the experiment: over the considered time frame a reduction of NO_x is observed. Unfortunately, a direct quantitative comparison is not possible. The simulation calculation is based on the stationary case represented before. The experimental parameters of the experiment with non-oscillating air serve as boundary conditions for the simulation. However, the experimental parameters of the experiments with and without oscillating air differ, provided that the experiment with the oscillation shows a higher mass conversion rate of the fuel bed. Consequently, the raw gas mass flow in the post-combustion chamber is higher and the combustion is profoundly more sub-stoichiometric. This can be seen in the O_2 concentration in the clean gas, which in non-oscillating case is just below 10%, while for the case of oscillation it is almost 4%. The low O_2 concentration inside the post-combustion chamber is advantageous with regard to NO_x reduction. The NO_x reduction rates averaged over the time are about 44% for the experiment and 16,5% average for the simulation. The minimum reduction rate value, in the simulation is 30% (see Figure 7).

It can be assumed, that the reduction rates agree better, if the stoichiometric conditions inside the post-combustion chamber in the experiment, as well as in the simulation are matched better, as for instance through the specification of a higher raw gas mass flow.

Summary

The application of biomass will be further an integral part of the power and heat generation with regard to the energy transition. With the continuous adjustments of the emission standards, including NO_x ,and the related pressure on costs for the operators, options for NO_x reduction through primary measures have to been looked for in order to avoid the application of expensive secondary measures, particularly SCR technologies. The time resolved air staging of the “oscillating combustion” can contribute significantly towards the reduction of NO_x in combustion plants, next to other primary measures i. e. local air staging.

In the project presented here, the experimental investigations with waste wood pellets show satisfactory agreement with the simulations. By an oscillation frequency of 1 Hz the NO_x concentration can be reduced by 44% for waste wood pellets and by 30% for wood pellets, respectively by simultaneously low CO content in the clean gas; basically, the simulation results repeat this effect, but the reduction rates were somewhat below the experimentally determined values (Figure7). In future the mixing conditions should be varied to evaluate the influence on the NO_x levels beside the influence of different oxygen levels in the exhaust gas.

In cooperation with the industrial partner POLZENITH the good results from the lab-scale have to be transferred to a grate firing with a thermal output of 400 kW. Additional to the development of a suitable pulsator, issues concerning operation with non-pelletized waste wood must be addressed i. e how the operation mode or the arrangement of the secondary air nozzles must be customized in order to secure a stable gasification on the grate. This is an essential requirement for the effect of the oscillating secondary air.

Figures and Tables

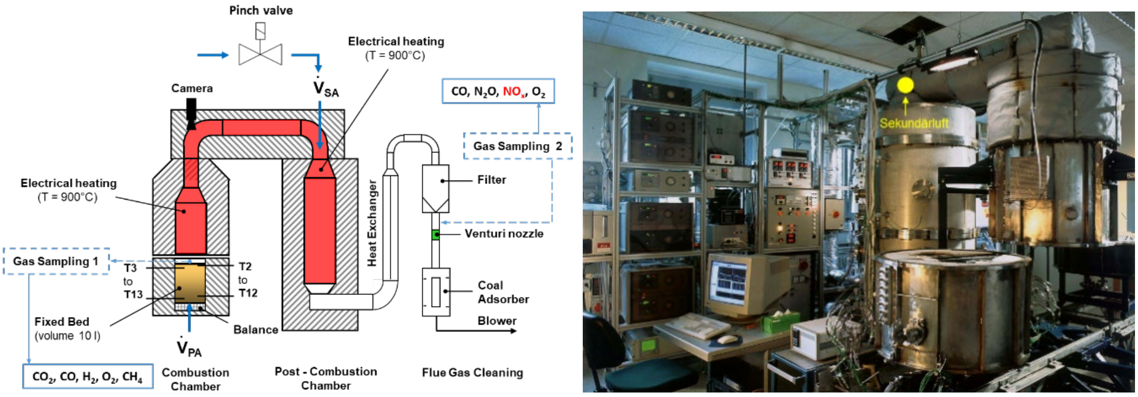

Figure1: Schematic layout of the KLEAA fixed bed reactor.

Table 1. Fuel chemical composition of waste wood pellets and wood pellets

wt. %	Wood pellets	Waste wood pellets (2021)
Water	7.60	3.10
Ash	0.30	1.12
Volatiles	77.80	78.09
Char	14.50	18.71
C_{fix}	14.20	17.58
C	47.10	47.94
H	5.80	5.91
N	0.12	4.09
O_{diff}	39.07	37.65
total - S	0.009	0.03
total - Cl	0.006	0.17
Sum	100.00	100.00
Bulk density [kg/m³]	300	130
Lower heating value [MJ/kg]	17.3	17.8

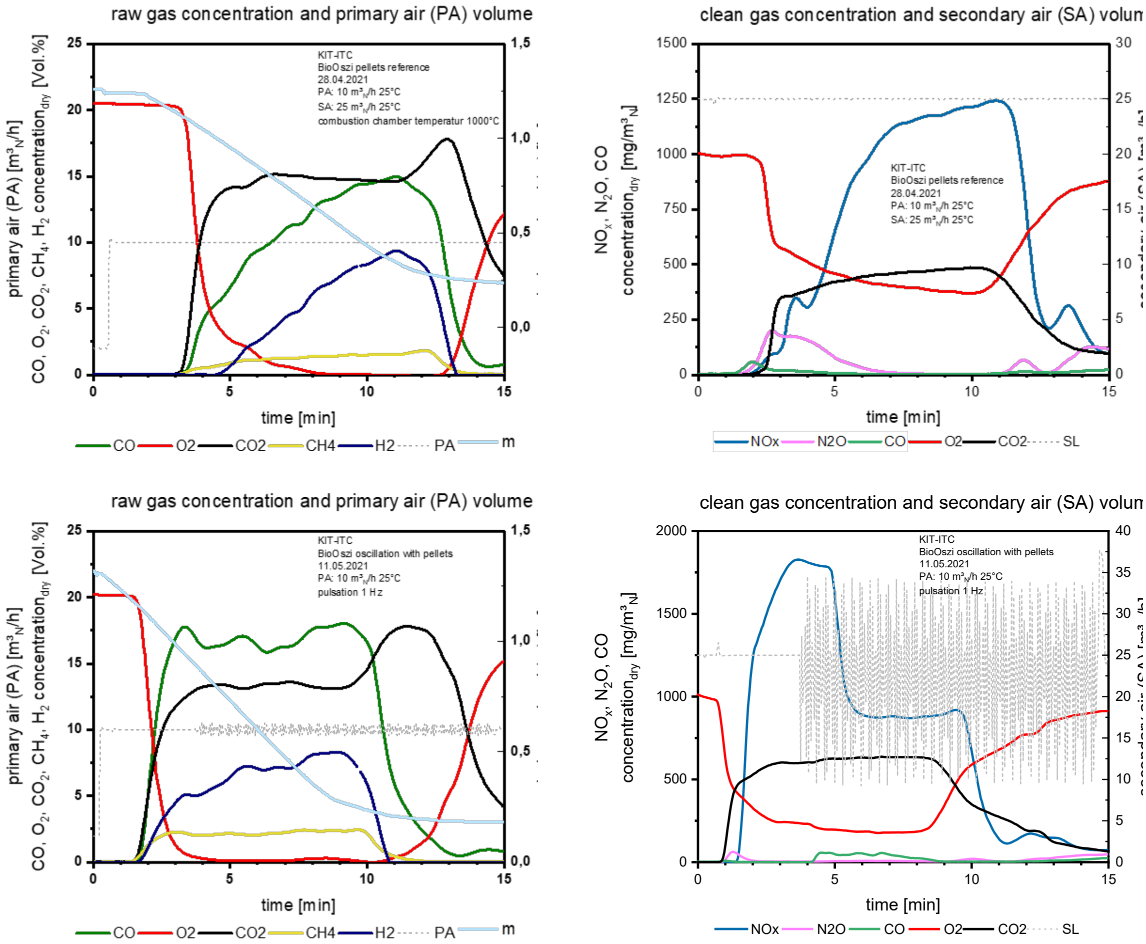

Figure 2. Experimental results by non-oscillating (top) and oscillating combustion (bottom) of waste wood pellets at KLEAA

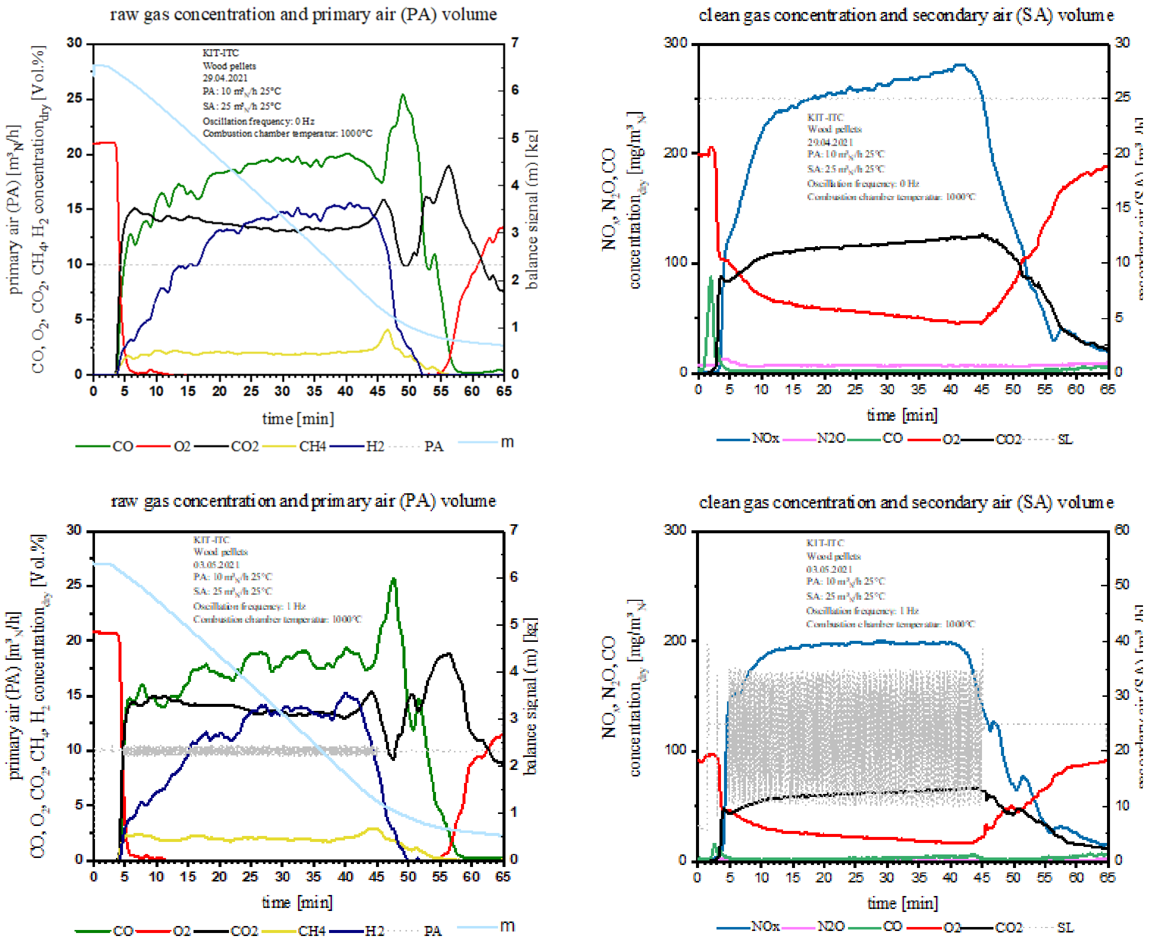

Figure 3. Experimental results of wood pellets combustion without oscillation (top) and with oscillation (bottom) at KLEAA

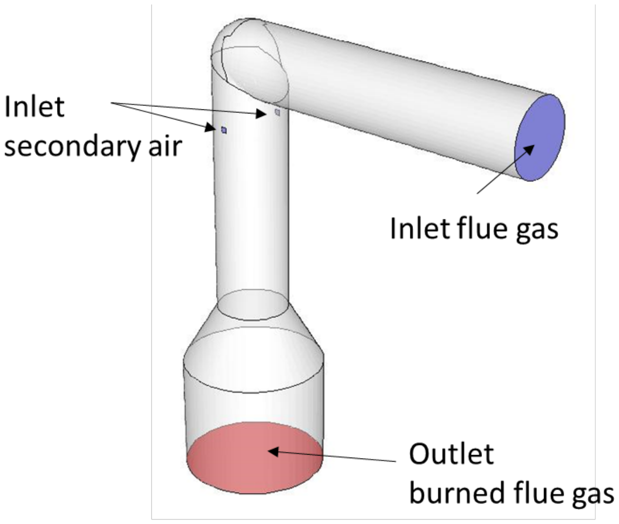

Figure 4. Simulation domain of the post combustion chamber (pcc)

Table 2. Models used in the simulation

Turbulence model	Detached Eddy Simulation
Radiation model	Discrete Ordinates Model
Absorption coefficient	WSGGM
Chemical model	Eddy Dissipation Concept
Reaction mechanism	LI32

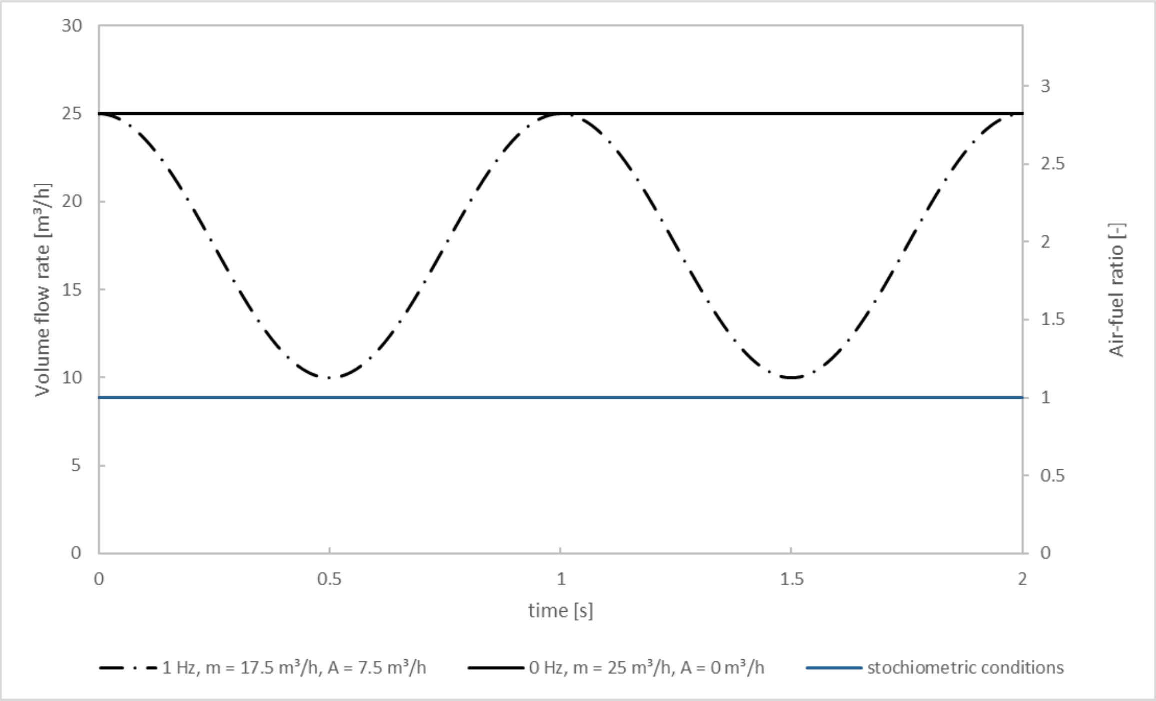

Figure 5. Volume flow rate of the secondary air for the oscillating (dotted line) and non oscillating (solid line) case

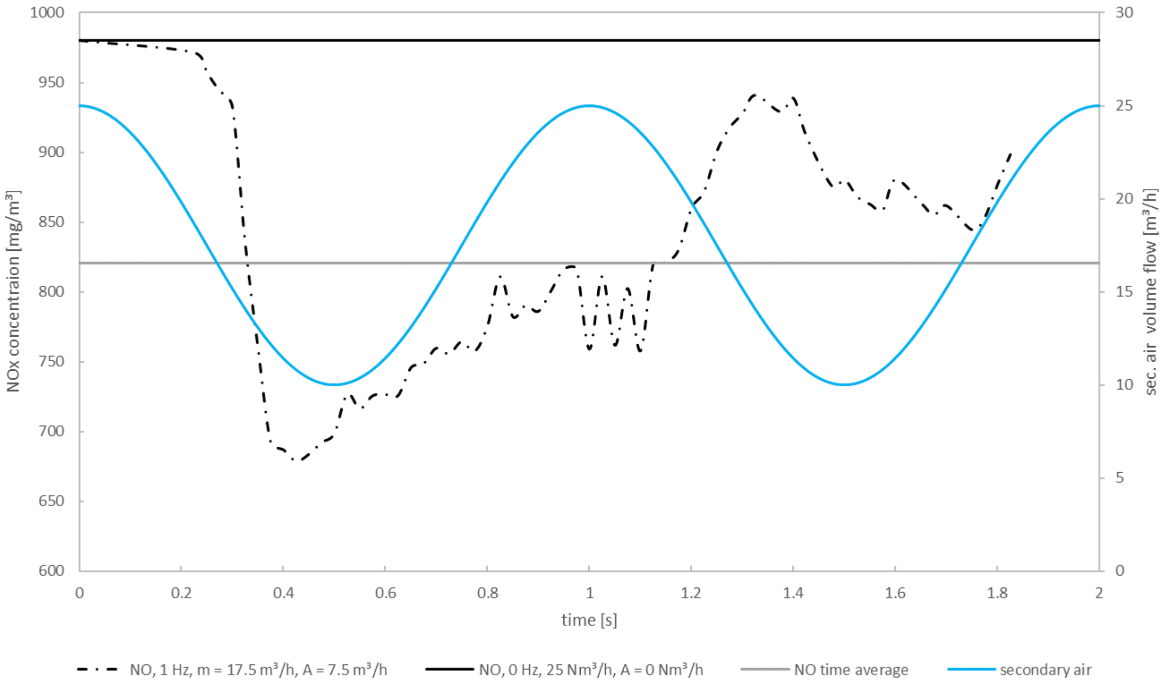

Figure 6. NO_x concentration at the pcc outlet

Table 3. Comparison of experimental and simulated main concentrations in the flue gas after secondary air injection

Gas concentration in clean gas		Experiment	Simulation
CO_2 [vol.%, dry]		9.97	12.19
O_2 [vol.%, dry]		9.35	9.08

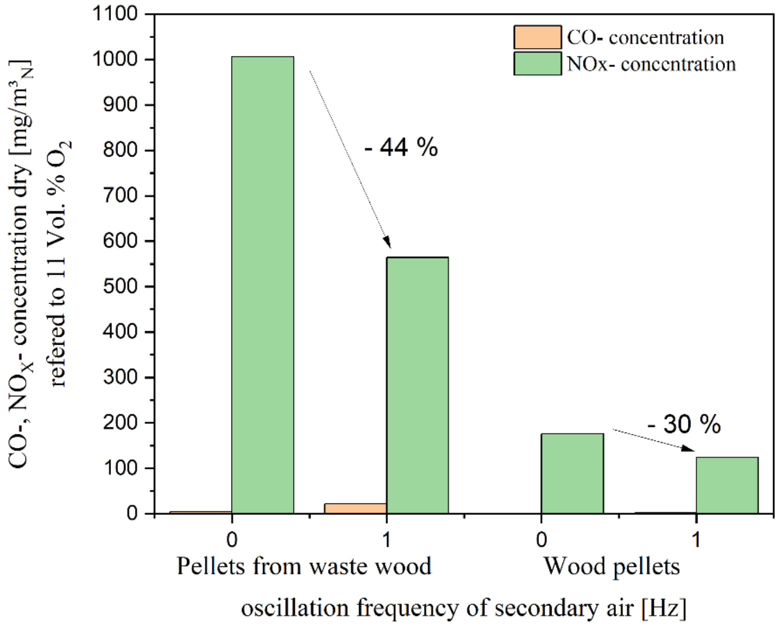

Figure 7. Summarized representation of the experimental and simulation results of the combustion of waste wood pellets and wood pellets in KLEAA

Acknowledgement

The project is funded by the Federal Ministry of Education and Research (BMBF), grant No.: FKZ 01LY1823

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