Oscillating Combustion for NO_x- Reduction in Pulverized Fuel Boilers

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Abstract

Thermal power plants in various fields of application are regularly adapted to the latest emission standards, applying the "Best Available Techniques Reference".

In recent years, new, stricter limit values have been in force in the power plant sector for power plant units with a thermal output of more than 300 MW that are operated with hard coal. In order to achieve the new limits, e.g. for NO_X emissions, downstream reduction processes (Selective Non-Catalytic Reduction, SNCR or Selective Catalytic Reduction) are usually used, which causes an increase of operating fluids (essentially ammonia water).

With an experimentally validated and patented process in which pulverized fuel is fed by oscillation via a swirl burner into a pilot combustion chamber, nitrogen oxides can be reduced without further measures, for example from 450 mg/m_N³ in non-oscillation mode (0 Hz) to 280 mg/ m_N³ in oscillation mode (3.5 Hz), normalized to an O₂ content of 6 % in each case.

Particularly promising are the experiments that use the oscillation of part of the burnt-out air (combustion gas) instead of the fuel in order to minimize, e.g., the wear of the oscillator.

The present results show that fuel oscillation alone is not sufficient to achieve nitrogen oxide concentrations below the values specified by the legislator. Therefore, a combination of different primary (and secondary) measures is required.

This paper presents experimental results on oscillating coal-dust firing based on a novel expert model. Detailed information on flame stability determination is presented in the accepted paper for INFUB 2022 "A camera-based flame stability controller for non-oscillating and oscillating combustion".

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, the framework conditions for air emissions from industrial plants are defined by the Bundes Imissionsschutz Gesetz (BImSchG). The nitrogen oxides NO and NO₂ are grouped together as NO_X and play a special role in the assessment of pollutant emissions.

In order to comply with the limit values for nitrogen oxide emissions, the NO_x concentration in the flue gas of industrial plants is reduced by two different measures. The so-called primary measures address the combustion-related fundamentals of nitrogen oxide formation. These formation mechanisms can be inhibited by clever air distribution in the combustion chamber. The subsequent denitrification of the flue gas belongs to the secondary measures and includes processes such as Selective Catalytic Reduction (SCR) and Selective Non-Catalytic Reduction (SNCR). In these processes, nitrogen oxides are reduced to ammonia with the aid of a catalyst or a reducing agent such as urea. 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 and is part of current research. Oscillating fuel addition has so far led to significant reductions in NO_X emissions. The changing stoichiometry that occurs in the flame result in less NO_X.

The investigations carried out have been largely limited to the oscillating addition of natural gas, which is easier to oscillate in the process. Due to the high abrasive properties and the associated process engineering challenges in the oscillating combustion of coal dust, there have been hardly any investigations in this area.

Based on the findings of [3], experiments with an oscillating pulverized coal furnace will be carried out and evaluated within the scope of this work. Due to the process challenges of oscillating coal dust, the focus of this work was to reduce nitrogen oxide emissions with oscillating addition of the incoming fuel-free air streams.

Based on the results presented in the journal in the Inventions section [2] in this paper, new findings on the effectiveness of oscillating combustion air and assistance in setting the swirl number are discussed

Basic principles

Since the temperature regime in pulverized fuel boiler technology is between 900 °C to 1100 °C, fuel-NO-formation plays an important role. The detailed mechanism are described in [2]. In addition to the well-known technologies for NO reduction with secondary measures such as SCR with catalysts and SNCR with ammonia injection, the primary measures affect the combustion itself, such as fuel and air staging, which is usually local, while the novel oscillatory combustion is time-dependent.

Oscillating combustion as primary measure

Oscillating combustion gained the interest of many researchers as a primary reduction measure for NO_x and is therefore invenstigated [3-7]. The result of a field study from 1996 to 2003 on oscillating combustion with gas flames [7] shows a significant reduction of NO_x emissions of 31-67% and an improved heat transfer of the flame by up to 13%. The results were confirmed by [3-6] for other combustion systems.

The basis is the oscillating addition of fuel or air into the reaction zone of the flame, which causes a temporal change of the local stoichiometry [3]. 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. Figure 1 shows this behavior schematically.



Figure 1: Behavior of stoichiometry in an oscillating combustion [3]

The oscillating addition of fuel/air leads to oscillating reaction conditions in the reaction zone of the flame. Low-fuel ($\lambda > 1$) and fuel-rich reaction zones ($\lambda < 1$) follow each other. Figure 1 shows an analogy to the air staging combustion mode. However, the reaction conditions 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 [6]. In the fuel-rich time periods Δt_1 , CO, NH₃ and NO_X are increasingly produced while in the fuel-poor time periods Δt_2 CO reacts to CO₂ and NO_X with NH₃ to N₂ and H₂O [3].

In the previous work of [5-7] the NO_x reduction potential by an oscillating addition of a gaseous energy carrier such as natural gas is investigated. Although [3] shows that an oscillating addition of secondary air during the combustion of chipboard cubes in a fixed-bed reactor also leads to a significant reduction in NO_x emissions, none of the previous studies show the effect of oscillating combustion of a solid fuel on NO_x emissions. The challenge lies in the high abrasion of the valve during the oscillatory conveyance of coal dust. In this paper, the results of the oscillating combustion of pulverized coal with a coal burner are investigated and complement the previous work of [3].

Methods

Pilot-scale power plant BRENDA

The BRENDA pilot power plant is used to investigate the combustion and emission behavior of conventional as well as alternative pulverized, gaseous or liquid fuels. BRENDA comprises a rotary kiln, which is connected to a post combustion chamber. The rotary kiln is equipped with an oil burner which supplies the required process heat for the combustion chamber. Figure 2 shows the part of the plant section in which the investigations were carried out.

The swirl burner for pulverized coal firing has a thermal output of 1 MW and is manufactured by SAACKE. A gas burner is installed in the opposite furnace wall at the same height as the pulverized coal burner. This ensures that in the event of a failure of the pulverized coal burner, the temperature in the post combustion chamber will not drop down below 850 °C in accordance with the 17th BimSchV, thus guaranteeing safe operation of the plant. Since the gas burner is not the subject of the investigations, it is not shown in Figure 2.



Figure 2: Pilot-scale combustion plant BRENDA

In order to unambiguously determine the NO_x emissions of the pulverized coal burner, 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 no longer react with other gas compounds.

To analyze the flame image during the experiments, a high-speed camera μ Eye UI-5240SE-M-GL from the manufacturer Imaging Development Systems GmbH (IDS) was pointed vertically at the flame from the coal burner above the post combustion chamber. The evaluation of the image material was carried out by the Institute for Automation and Applied Computer Science (IAI), whose publication [8] is referred to here.

Figure 3 shows a closer view of the geometry of the pulverized coal burner. The burner, the fuelbearing central tube, the annular gap 1 (R1), the annular gap 2 (R2), the burner stone and the combustion air inlet are arranged concentrically. The cone serves to stabilize the flame and is set at a constant distance of 15 mm during the entire test campaign (the maximum diameter of the cone is about 15 mm).



Figure 3: Schematic of coal burner

The pulverized coal enters the combustion chamber through the central pipe Z. The combustion air enters the furnace 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 displaced along the marked x-axis into the furnace wall, changing the gap width of the combustion air inlet. By shifting and/or increasing the combustion air flow, the outlet velocity of the combustion air flow can be influenced. Equation (1) shows the simplified calculation formula for the swirl number S resulting from this geometry as a function of the position of the traverse path Δx of the burner block in x-direction according to [9]. The distance $\Delta x = 0$ mm corresponds to a swirl number S of 0, the maximum of Δx is 110 mm.

$$S = 0.014661 \frac{1}{mm} \cdot \frac{130 \text{ mm} + [\Delta x \cdot \sin^2(30^\circ)]}{130 \text{ mm} + [\Delta x \cdot \sin(30^\circ)]} \cdot \Delta x \cdot Q^2, \quad \text{with } \Delta x \text{ in mm}$$
(1)

where Q describes the ratio of the swirled combustion air \dot{V}_{VL} to the total incoming air volume flow of the burner according to equation (2).

$$Q = \frac{\dot{V}_{VL}}{\sum_{i}^{n} \dot{V}_{i,L}} \tag{2}$$

For the description of the stoichiometry at the pulverized coal burner, the local air ratio λ_{loc} was calculated as a function of the 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 (3).

$$\lambda_{\rm loc} = \frac{\dot{V}_Z + \dot{V}_{R1} + \dot{V}_{R2}}{\dot{m}_{coal} \cdot l_{min}} \tag{3}$$

Another stoichiometric parameter was the air number λ_{burner} taking into account the combustion air flow entering the pulverised coal burner \dot{V}_{VL} according to equation (4).

$$\lambda_{burner} = \frac{\dot{V}_{z} + \dot{V}_{R1} + \dot{V}_{R2} + \dot{V}_{VL}}{\dot{m}_{coal} \cdot l_{min}}$$
(4)

In order to evaluate and analyze the influencing parameters on the behavior of the oscillating combustion, several campaigns were performed to supply training data into an expert model and validate it with test data in campaign C. The identified main influencing parameters were further investigated in campaign E. An additional campaign D focussed on the influence of the interaction between oscillating combustion and swirl number.

Trial plan Campaign C

For the modelling, 48 model tests were carried out, taking into account the technical boundary conditions, e.g. minimum air flows through gaps for cooling. In the model tests, the previously determined influences of the oscillation frequency [3] and the level of the volume flows in R1 and R2, the volume flow of the combustion air from the rotary kiln, the quantity of coal supplied and the volume flow conveying coal were examined.

Table 1 shows the varied parameters for campaign C and their factor levels for the pulverized coal burner and the primary burner from the rotary kiln. The oscillation frequencies of the volume flows from annular gap R1 and R2 were examined in 5 factor levels, as a non-linear behavior of these variables was observed in the previous campaign. Due to the technical limitation that the pinch valve does not close properly at frequencies higher than 3 Hz, the parameter space for the oscillation frequency ranged between 0 and 3 Hz. It was known from previous experiments that oscillation has the greatest effect at high frequencies, so an additional factor level was defined at 2.67 Hz. It was also known that the oscillation of the fuel flow leads to lower NO_X emissions and has a primary optimum. Therefore, in this campaign, the effect of oscillating air volume flows in R1 and R2 was investigated as a secondary optimum in 4 factor levels. Due to 50 normal cubic meters per hour (in the following labeled as m_N^3/h) during operation. In these cases, the volume flow in R2 could only be reduced to 65 m_N^3/h and led to a subsequent adaptation of the test plan.

Table 1: Varied parameters and their factor levels in campaign C

System	Parameter	Factor level					Unit	
•		1	2	3	4	5		
Rotary kiln burner	Combustion air rotary kiln burner	1200	1400	-	-	-	m _N ³/h	
Coal burner	Mass flow coal Z	70	90	-	-	-	kg/h	
	Air flow Z	70	90	-	-	-	m _N ³/h	
	Air flow R1	80	90	100	110	-	m _N ³/h	
	Oscillating frequency R1	0	1	2	2.67	3	Hz	
	Air flow R2	50 (65)	70	90	110	-	m _N ³/h	
	Oscillating frequency R2	0	1	2	2.67	3	Hz	

Expert model

The expert knowledge based multivariate regression model was used for the campaign to derive an equation for NO_X concentration as a function of the main influencing parameters. The two interactions between the oscillation frequency and the level of the oscillated air flow are included in the model as additional interactions. In addition to the varied parameters, the swirl number S according to equation (1) and λ_{burner} according to equation (4) are included in the model. Table 2 shows the influencing variables selected for the model and the corresponding abbreviation.

Table 2: Abbreviation of the influencing parameters

Abbr.	Parameter	Unit
а	Oscillating frequency R1	Hz
b	Oscillating frequency R2	Hz
С	Mass flow coal Z	kg/h
d	Air flow Z	m _N ³/h
е	Air flow R1	m _N ³/h
f	Air flow R2	m _N ³/h
g	Combustion air rotary kiln burner	m _N ³/h
ĥ	Swirl number S	-
i	λ_{burner}	-
ae	Interaction in R1	Hz m _N ³/h
bf	Interaction in R2	Hz m _N ³/h

To determine the significance of the respective variables, the F-value and the P-value were calculated for the model created [10, 11]. Within the model, a significance level α of 5% was chosen, as is usual for engineering questions. Moreover, this did not result in a too sharp separation in the reduction of the model parameters in the expert model. This ensured that a parameter tends to remain in the model and that the expert can evaluate its plausibility afterwards.

The acquired expert model was part of the work of this paper. In order to verify the developed expert model the EDI hive model generator, a tool for statistical analysis provided by EDI GmbH¹ [4], was used.

Trial plan Campaign E

Campaign E aimed to investigate a high oscillated air flow through the annular gap R2. Two tests were carried out. The first test was performed to verify the findings from previous experiments in 2018 [3], in which a high-volume flow was oscillated. In the second test the identified phenomenona were investigated. In the test settings from campaign E, λ_{burner} and thus the incoming volume flows and the coal mass flow were kept constant. This results in a smaller step spacing for the influence of the swirl number S, which will be further investigated in campaign D.

Table 3 shows the test settings for the investigation of the influence of the level of the oscillated volume flow in R2. No air from R1 was introduced into the system. In the parameter variation, the combustion air was shifted to the volume flow from R2 with simultaneous change of the swirl setting. As a result, λ_{loc} was changed, but λ_{burner} and the swirl number S remained constant. In tests 5 and 6, the combustion air could not be reduced below 313 m_N³/h due to the minimum air volume technically required for burner cooling. This led to a slight deviation from λ_{burner} of 1.12 instead of 1.11.

¹ for further information please see https://edi.gmbh/en/

Table 3: Trial plan Campaign E: Investigation of the level of the oscillated air flow

System	Parameter	Trial						Unit
-		1	2	3	4	5	6	
Coal burner	Mass flow coal Z			70 (cor	nstant)			kg/h
	Air flow Z	80 (constant)						m _N ³/h
	Swirl number S	0.3 (constant)						
	λ_{loc}	0.28	0.28	0.48	0.48	0.51	0.51	-
	λ_{hurner}	1.11	1.11	1.11	1.11	1.12	1.12	-
	Combustion air	420	420	321	321	313	313	m _№ ³/h
	Air flow R2	65	65	164	164	180	180	m _N ³/h
	Oscillating frequency R2	0	2.67	0	2.67	0	2.67	Hz

In order to determine a statement about the flame image and its influence on the nitrogen oxide concentration in this series of tests, the optical analysis of these tests was carried out using software from IAI. The program determined the relative area of the flame in relation to the total combustion chamber cross-section as a percentage. The relative stable flame area results from an averaging of image sequences consisting of 50 images. If the flame shapes of the images match, a relative flame stability of 100% is calculated, if the flame shapes differ strongly from each other, a relative flame stability of 0% is obtained. The exact calculation bases for the optical analysis as well as the assumptions made for the calculations can be taken from [8, 12].

Trial plan Campaign D

To investigate the interaction between the swirl number and the oscillation at different volumetric flows in R1 and R2, a full-factorial experimental design with two-stage parameter variation of the swirl number S and the oscillation frequency in R1 and R2 was carried out. As in campaign C, no randomization of the experiments was performed due to the technical implementation.

According to equation (1) the swirl number S can only be influenced to a small extent by the process path Δx . In order to achieve a suitable stage spacing, the influence of the volume flow ratio Q according to equation (2) was also taken into account. The influence of the swirl number at S=0.36 and S=0.79 was investigated by changing the process path Δx and the entering volume flow rates.

By increasing the incoming volume flow rates, the local stoichiometry λ_{loc} was kept constant at λ_{loc} = 0.41 by a simultaneous adjustment of the coal mass flow.

The selected parameter settings with the stage spacing are shown in Table 4.

System	Parameter	Trial						Unit
-		1	2	3	4	5	6	
Coal burner	Swirl number S	0.36	0.36	0.36	0.79	0.79	0.79	-
	Oscillation frequency in R1	0	3	0	0	3	0	Hz
	Oscillation frequency in R2	0	0	3	0	0	3	Hz
	λ_{loc}	0.41						-
	Combustion air		420			840		m _N ³/h
	Mass flow coal Z		70			90		kg/h
	Air flow Z		70			90		m _N ³/h
	Air flow R1		70			90		m _N ³/h
	Air flow R2		70			90		m _N ³/h

Table 4: Trial plan campaign D investigation of the swirl number

Results and discussion

Campaign C – Expert Model

The following figures show the measured NO_X concentration related to 6 Vol.% oxygen over the respective influencing parameters plotted in Table 1. The points represent the measured NO_X concentrations of the individual test settings from the statistical test planning.

Figure 4 shows all results from campaign C as a function of the ratio of the volume flow in R2 to the total combustion air entering through the burner, where oscillating combustion was achieved through the annular gap R2. The opposite linear regression from the oscillated data points and the non-oscillated data points underlines the statement that high oscillating air flow through annular gap R2 leads to a reduction in NO_X emissions.



Figure 4: Campaign C: NOx- concentration as function of the ratio of volume flow in R2 to the total combustion air

The outlier at 500 mg/m_N³ in the non-oscillated data series at a ratio from 0.24 demonstrates the wide variety of measurements and underscores why a statistical analysis is needed in this study. In addition, more data points in both series for higher and lower ratios would help to outline the phenomenon examined in this study. Nevertheless, the two regressions demonstrate the advantage of oscillating combustion.

In the following the results of the statistical analysis based on the developed expert model and the verification with the EDI hive model generator [4] are discussed.

The expert model includes all influencing parameters that were specifically modified in the statistical design of the experiment, as well as the air ratio λ_{burner} , the swirl number S and the interaction of volume flow and oscillation in annular gap R1 and annular gap R2. The explanatory variables and their abbreviations for the following considerations are given in Table 2. First, the multivariate regression model with eleven descriptive variables is considered. The approximated NO_x emission data, the calculated predictive accuracy of the model for training and test data and the F-value of the model are shown in Figure 5. For the tabular F-value with the degrees of freedom f₁ = 11 and f₂ = 36, the F-value is F_{11,36,0.95} = 2.07. The model has a higher F-value and therefore contains variables that significantly describes the problem.





Table 5 shows the regression coefficients of the individual influencing variables for the regular and the standardized training data. For the non-standardized coefficients, the set parameter variations can

be used directly to calculate the nitrogen oxide concentration and the units of the coefficients reduce to the unit of the target value to be approximated, the nitrogen oxide concentration in the flue gas in mg/m_N^3 . The unit and the value of the coefficients are given in the table below. For the non-standardized regression coefficients, no comparison is possible in terms of weighting due to the different orders of magnitude of the parameters. For this reason, the standardized dimensionless beta coefficients are also given. With them, the coefficients can be compared with regard to their weighting due to the previously performed standardization of the data.

Table 5: Regression coefficients from expert model

Abbr.	Parameter	Unit	β _j / Unit		$\beta_{j,norm}/ \text{ mg/m}_N^3$
	Interception y axis		8485.27	mg/m _N ³	598.66
а	Oscillating frequency R1	Hz	55.62	mg/(Hz m _N ³)	65.94
b	Oscillating frequency R2	Hz	26.08	mg/(Hz m _N ³)	64.06
С	Mass flow coal Z	kg/h	-43.43	$(h mg)/(kg m_N^3)$	-421.64
d	Air flow Z	m _N ³/h	2.5	(h mg)/m _N ⁶	12.39
е	Air flow R1	m _N ³/h	1.78	(h mg)/m _N ⁶	20.71
f	Air flow R2	m _N ³/h	1.92	(h mg)/m _N ⁶	44.10
g	Combustion air rotary kiln burner	m _N ³/h	0.04	(h mg)/m _N ⁶	3.98
h	Swirl number S	-	-2901.28	mg/m _N ³	-118.61
i	λ_{burner}	-	-3035.93	mg/m _N ³	-444.21
ae	Interaction in R1	Hz m _N ³/h	-0.61	(h mg)/(Hz m _N ⁶)	-66.35
bf	Interaction in R2	Hz m _N ³/h	-0.79	(h mg)/(Hz m _N ⁶)	-83.19

The corresponding model equation from the standardized coefficients from the linear terms a_{norm} - g_{norm} and the interaction terms a_{norm} and bf_{norm} results to the following equation (5).

$$\hat{\mathbf{y}} = \begin{bmatrix} 598.66 + 65.94 \cdot a_{norm} + 64.06 \cdot b_{norm} - 421.64 \cdot c_{norm} + 12.39 \cdot d_{norm} \\ + 20.71 \cdot e_{norm} + 44.1 \cdot f_{norm} + 3.98 \cdot g_{norm} - 118.61 \cdot h_{norm} \\ - 444.21 \cdot i_{norm} - 66.35 \cdot ae_{norm} - 83.19 \cdot bf_{norm} \end{bmatrix} \begin{bmatrix} mg \\ m_N^3 \end{bmatrix}$$
(5)

The largest values for the standardized coefficients $\beta_{i,norm}$ are the parameters c and i, both related to coal mass flow and the stoichiometry. The third-largest value results for h, the swirl number. All three of the above quantities have a negative sign, i.e. the approximated target quantity decreases with an increase in the parameter. This is of interest for the coal mass flow, since with its increase, more nitrogen is introduced into the system and NO_X emissions should increase accordingly. The effect of the stoichiometric air ratio can be explained by the anti-proportionality to the coal mass flow (see equation (3)). Similarly, the linear influences of the oscillation frequency a and b have a positive sign and the interaction terms of the two variables ae and bf have a negative sign. Thus, according to equation (5), the effects are in a contrary relationship to the target quantity. Such dependencies can be used to map the interactions for the oscillation frequency and the volume flow in R2 of Figure 7 with a statistical model. Furthermore, the model assumes that the parameters are varied within the investigated limits of the design space.

Since equation (5) is a mathematically determined model equation for the investigated parameter from Table 1, it is valid only for this particular parameter. Thus, the correlations behind the examined regression coefficients follow the fitted parameter combinations, whereby the parameters can be used to predict the target quantity with an accuracy of 88%.

The results for the examined parameters of the expert model are shown in Figure 6. The red line represents the significance level $\alpha = 0.05$, which determines whether a parameter is significant or not. P-values greater than α are non-significant and P-values less than α indicate significant model parameters, P-values less than $\alpha = 0.001$ are strongly significant. The abbreviations for the influencing variables can be seen in Table 2.



Figure 6: Significance of influencing variables

The evaluation clearly shows that the influencing parameters e- volume flow in R1, f- volume flow in R2 and g- combustion air from the rotary kiln burner have a higher P-value than the selected significance level α of 0.05. According to the performed evaluation, the mentioned parameters have no explanatory content for the formed model. The parameters b- oscillation frequency in R2, c- coal mass flow in Z, hswirl number, i- air ratio λ_{burner} and bf- interaction of oscillation frequency and volume flow in R2 are considered to be strongly significant for the model formed. Furthermore, it can be seen that the interactions ae and bf of the oscillation frequency with the respective volume flow from R1 or R2 can be classified as significant parameters, while the linear influences of the volume flows in R1 and R2 have no significant effect on the model and these only contribute to the model in their interaction with the respective oscillation frequency. As obvious significant effects with a P-value of 0, b- oscillation frequency in R2, c- carbon mass flow in Z, i- air ratio λ_{burner} and bf interaction of oscillation and volume flow in R2 can be identified. The high significance of the two parameters coal mass flow and λ_{burner} with respect to NOx emission is due to the influence of the increased fuel input and is consistent with the findings from the analysis of the regression coefficients from equation (5). The increase in the fuel input is directly reflected in the coal mass flow; the air ratio λ takes into account the incoming air flows and is anti-proportional to the coal mass flow according to equation (4). The oscillation frequency in R2 and the interaction of the oscillation frequency with the volume flow in R2 reflect the investigated effect of air oscillation. Obviously, it is possible to exert a significant effect on NOx emissions through oscillation of the air volume flow. The influence of the swirl number also has a significant effect in the evaluated data. This is consistent with the observation in Equation (5), where the swirl number has the third largest standardized regression coefficient.

In order to verify the expert model the evaluations with the EDI hive model generator [4] for the varied parameters' oscillation frequency and volume flow in R1 and R2, coal mass flow, volume flow of the coal conveying air, volume flow of combustion air from the rotary kiln with regard to the interaction of volume flow and the oscillation frequency in R2 are shown in Figure 7. The provided tool can be used to remove scatter due to parameter variations of the other parameters.



Figure 7: Results for the Interaction in R2 of the EDI hive model generator [4]

The two upper diagrams show the dependence of nitrogen oxide on the oscillation, and the two lower diagrams show the dependence from the volume flow. Now two scenarios are considered, the two left diagrams show the course of the approximated nitrogen oxide concentration for an oscillation frequency of 0 Hz set by the cursor through the orange line. The two diagrams on the right show the NO_X concentration for a set oscillation frequency of 3 Hz.

For the diagram of the volume flow dependence in the left part of the figure, the nitrogen oxide concentration increases with the volume flow, while the dependence of the NO_X concentration at a oscillation of 3 Hz in the right part of the figure shows a clear reduction of the NO_X concentration with increasing volume flow. With the model generator it is possible to determine the oscillation frequency for the case of volume flow reverses in R2 to 0.8 Hz.

The evaluation shows that without oscillation, the increase of the volume flow leads to an increase in the NO_X concentration. With an oscillation of 3 Hz, the increase of the volume flow leads to a significant reduction of the nitrogen oxide concentration.

In the case of an increasing volume flow without oscillation, the supply of more oxygen leads to higher emission values of the nitrogen oxides. The NH_i species react directly to NO under these conditions, and the reaction to molecular nitrogen is inhibited here. Increasing the volumetric flow at an oscillation of 3 Hz should also produce similar results. Apparently, oscillation of higher volume flow rates can produce a more concise oscillatory profile of local stoichiometry. This allows the reaction of the formed NO to molecular nitrogen in the time gaps of the interruption of the volume flow. It should be mentioned that a higher volumetric flow has a larger impulse than a lower volumetric flow, which allows a more efficient interruption of the coal mass flow. Not to the extent that fuel flow oscillation would cause this, but to the extent that there are favorable reaction conditions for the reduction of nitrogen oxide emissions.

Based on the results of the statistical analysis by the expert model and the EDI hive model generator [4] and the results from campaign C, which showed in Figure 4 that the oscillation of a high-volume flow in R2 leads to a lower nitrogen oxide concentration, the oscillating addition of a high-volume flow of 180 m_N^3 /h from the annular gap R2 was investigated.

Campaign E

The measured nitrogen oxide, oxygen and CO concentrations in the experiments conducted to reinvestigate test settings from 2018 in which a high-volume flow was oscillated in R2 are shown in Figure 8 over the test period.

The reduction in NO_x emissions due to oscillation of the volume flow in R2 at 2.67 Hz is clearly visible. For the two experiments, a constant nitrogen oxide concentration of 620 mg/m_N³ for the oscillation-free and 350 mg/m_N³ N for the oscillating process control is achieved over the 60-minute test period. This reduces the nitrogen oxides in the flue gas by 45%.



Figure 8: Oscillation of a high-volume air flow in R2 at 2.67 Hz

This represents the best measurement result of the tests carried out in this paper on nitrogen oxide reduction by oscillation of the air volume flows. The oxygen and carbon monoxide concentrations remain

constant over the entire period shown. This indicates a stable combustion process during the experiments.

At Figure 9 the dependence of the NO_X emissions on the level of the oscillated volume flow in R2 and the oscillation frequency at constant swirl and constant air ratio at burner λ_{burner} is shown. The reference value is the NO_x concentration at the respective non-oscillated state. The other parameter settings of the constant incoming flows are shown in the margin of the figure and in Table 3. Nitrogen oxide emissions at a volume flow of 65 m_N^3/h increase slightly due to the oscillation of 2.67 Hz. Increasing the volume flow entering the annular gap R2 to $164 \text{ m}^3/\text{h}$ significantly increases the nitrogen oxide emissions in the non-oscillated state. In contrast, oscillation of this volume flow shows a significant reduction in nitrogen oxide emissions. At a volume flow of 180 m_N³/h, the nitrogen oxide emissions in the non-oscillated state are slightly higher than at a volume flow of 164 m_N^3/h , but a similar behavior in NO_X reduction can be observed here as in the oscillated state.



Figure 9: Investigation of the volume of the oscillated air flow

Because of the increased oxygen supply in the flame associated with the increase in the volume flow in R2, the nitrogen oxide emissions in the tests increase to 164 and 180 $m_N^{3/h}$ in the oscillated state. Reference [13] emphasizes the careful matching of the different flow fields of the fuel-related primary air streams and the fuel-remote secondary air streams in a pulverized coal burner. Accordingly, the increase in the concentration of nitrogen oxide in the flue gas during the shift of the combustion air into the annular gap R2 can be explained by a poorer preheating of the coal mass flow. This leads to an unfavorable combustion behavior of the coal dust. At the same time, a swirl number of 0.3 is not sufficient to create a recirculation area that would ensure better preheating of the fuel flow.

According to the literature of [14-16], the backflow area for type I and II flames develops only at a critical swirl number of $S_{\text{theo}} \approx 0.5$. However, at the prevailing temperatures, the formation mechanisms are limited to the fuel N mechanism. Accordingly, an increase in oxygen concentration leads to the preferred reaction path from HCN via NHi to NO. Since reducing conditions do not exist in the flame at an increased oxygen supply, the reaction of NHi with already formed NO to N2 is inhibited. Although λ_{loc} < 1 applies to the local stoichiometries of all investigated test settings, λ_{loc} is significantly larger at the higher volume flows in R2. If we now compare the local stoichiometry with the total fuel nitrogen (TFN) curve shown in the NO_x concentration, it increases with increasing air ratio. In addition, the exhaust gas mixture contains more HCN and NH₃ at the local air ratios of 0.28 to 0.51. The HCN and NH₃ concentration in the exhaust gas mixture increases with the local air ratios of 0.28 to 0.51. For both N-species, the residence time in the post combustion chamber is sufficient to react to NO_x at a total air ratio in the post combustion chamber of 1.1 to 1.4 up to the measurement point of nitrogen oxide concentration. Considering the oscillation effect of the two high oscillating volume flows of 164 and 180 m_N^3/h at 2.67 Hz, the nitrogen oxide emission can be reduced despite the high local air ratios. Thus, the interruption of the volume flow leads to alternating reducing and oxidizing conditions in the flame, resulting in NO_x reduction. In this process, more NO is formed under oxidizing conditions. Under reducing conditions, the NO is reduced to N₂. In parallel, the NO molecules can be returned to the reaction path of the fuel-N mechanism via the NO-recycle mechanism. These two mechanisms explain the lower nitrogen oxide emissions. In further considerations, it would be useful to measure the local concentrations of the N-species to draw further conclusions about the formation mechanism under oscillating conditions. With respect to the reference value of the nitrogen oxide concentration at $65 \text{ m}^{3}/\text{h}$ and without oscillating air addition, no reduction of nitrogen oxide emissions is observed at the selected test settings, but the results show that the NO_x formation mechanisms can be substantially reduced by oscillation.

A more detailed analysis of the flame pattern for the phenomenon of increasing concentration of the nitrogen oxide in the flue gas by shifting the air volume flow from the combustion air into the annular gap R2 with oscillation-free process control is explained in more detail below. The visual observation of the flames of the non-oscillated test settings are shown in the image series of 0, 5, 10, 15 and 30 seconds of the test time in Figure 10. A volume flow of R2 of 65 m^3_N /h results in a longer flame and a uniform distribution of the bright flame area along the entire length of the flame. The higher volume flow in the tangentially introduced combustion air results in a more stable, elongated flame pattern.



Figure 10: Photo series of a non-oscillating flame at different air flows in R2

The two lower image series at a volume flow in R2 of 164 and 180 m_N^3/h show a higher flame brightness in the area near the flame root than in the burnout area of the flame. Here, the volume flow of the combustion air is significantly reduced (cf. Table 3), resulting in a shorter and bushier flame.

The evaluations with the IAI program from Figure 11 confirm this fact for the shorter and bushier flame shapes shown in Figure 10 for the high-volume flows in R2 of 164 and 180 m^3N/h , since both the relative flame fraction area and the relative stable flame fraction area are significantly lower for the two flames.



Figure 11: Image analysis of relative flame area

Consequently, with the longer flame at a non-oscillated volume flow in R2 of 65 m^3_N/h , a more uniform burnout of the combustion gases is possible over the length of the flame, which is associated with a lower nitrogen oxide concentration in the flue gas (cf. Figure 1). Definitionally, there are no type I /II flames due to the too low swirl number, so that the results do not agree with the previous studies of [15].

However, the influence of the flame shape on the nitrogen oxide concentration in the flue gas can be clarified from the optical evaluations discussed above.

Campaign D

The results of the investigation of the swirl influence in combination with the oscillation of the air volume flows are shown in Figure 12. The y-axis shows the measured NO_x emissions at two different swirl numbers with a variation of the oscillated air flow rate in the annular gaps R1 and R2 at 3 Hz. As a baseline, the values for the two swirl numbers without oscillation are shown in the middle. The exact step variation of the entering flows is given in the table next to the figure.



Figure 12: Campaign D: NO_x emissions

For all three oscillation settings, the measured NO_x emissions are always slightly lower at a higher swirl rate than at a lower swirl rate. However, the difference is so small that no clear conclusion can be made regarding the influence of the swirl.

Also, with regard to the influence of the oscillation, there is hardly any difference between the values. In addition, the location of the oscillating added air, whether entering through annular gap R1 or R2, does not seem to play a role.

During the test campaign D, a flickering and unstable flame appeared, especially at the higher swirl number of S = 0.79, which was not observed in previous tests. In addition, unburned dust clouds shot out of the side of the flame area at irregular intervals.

Based on the observed flame pattern, more detailed observations of the C_{total} emissions were made. The results for the test settings of Table 4 are shown in Figure 13.





The emission values for C_{total} for the low swirl number are in the same range between 0.3 to 0.5 mg/m_N³, which is acceptable. For the higher swirl number, the C_{total} emissions are about 30 mg/m_N³. The high values of the C_{total} for the higher swirl number indicate incomplete combustion under the given conditions. According to the flame area observations, ignition of the flame was not always possible. The sudden unstable combustion behavior can be partially explained by the visual evaluation of the burner during and after the tests shown in Figure 14.



Figure 14: Burner during and after experiments

Slagging was already observed here during the test. In the outer annular gap through which the combustion air enters the combustion chamber (cf. Figure 3), clear slag build-up can be observed in the upper right-hand area of the burner.

The combustion air flow surrounds the flame like an envelope and stabilizes it by the swirl. If the slagging disturbs it, the flame is no longer stabilized by the swirl. The kinetic energy of the combustion air can therefore not flow into the formation of an internal pressure gradient.

As a result, the hot exhaust gases are not drawn to the root of the flame, which reduces the effect of NO reburning. In NO reburning, NO already formed reacts under reducing conditions as shown in [2] HCN and further to N_2 . Thus, the swirl-induced backflow can be used to influence the effect of the NO reburning.

In addition to slagging, the flame break-off can be attributed to the high outlet velocities of the combustion air. According to [2], the breakaway of highly swirled flames can lead to ignition stability problems. However, with a swirl number of 0.79, there is still no strong swirling of the flame body.

The combination of the high combustion air volume flow with the irregular gap narrowing leads to higher outlet velocities in certain areas of the annular gap, which explains the irregularly occurring bursts of the coal dust jet from the flame body.

At the same time, stable ignition of the coal can no longer be achieved at these velocities. It has also been observed that the constantly changing test settings cause the slagging to change during the test campaign. When the burner is hot enough, the slag begins to melt and flow downward, immediately changing the burner geometry. This can cause the observed change in the geometry of the outer annular gap. The comparison in Figure 14 between the burner during and after the tests shows very well how the slagging pattern changes during the test period. In particular, on the right side of the burner where massive slagging is present during the tests, which no longer adheres to the burner at the end of the test campaign.

At this point, the investigation of the interaction between the swirl number and the oscillated combustion on the NO_X emission ended. The test system is subject to drift and in this case, stable operation was no longer possible under the selected settings. This drift can be remedied by stationary and stable operation. However, due to the continuous parameter variation in this study on oscillating combustion, this was not possible. Accordingly, this phenomenon occurs less in continuously operated plants.

Summary

A statistical experimental design was established to determine influencing parameters for oscillating coal dust combustion without interrupting the fuel mass flow. The oscillating addition of the air volume flows allowed the identification of the secondary optima and obvious influences on the nitrogen oxide concentration in the flue gas. In particular, oscillation at frequencies of 3 Hz reduced the NO_x concentration by up to 25% for the experimental room studied.

However, the exact distribution of the air volume flows and the distance between the addition location and the fuel mass flow played an important role. The coal was added at the burner via the central inlet pipe, and the oscillation of the air volume flows took place in the concentrically arranged annular gaps. The results showed a high effect for the oscillation of the outer annular gap (R2) and hardly an effect for the oscillation of the inner annular gap (R1). This finding can be explained by the dependence of the residence time of the combustion gas before the secondary air is mixed into the flue gas at a coal flame, described by [17]. The oscillation of the locally closer oscillated volume flow from R1 resulted in a maximum nitrogen oxide reduction of 11%, while the oscillation of the air volume flow from R2 further away from the coal mass flow resulted in a maximum nitrogen oxide reduction of 25 %. In addition, the results of the different test settings could be used to determine a relationship between the level of the oscillated volume flow and the oscillation frequency. Accordingly, increasing the oscillated volume flow at a high oscillation frequency of 3 Hz led to a reduction of the nitrogen oxide concentration in the flue gas, while increasing the oscillated air volume flow at a low oscillation frequency of less than 0.8 Hz led to an increase of the nitrogen oxide concentration in the flue gas. This can be attributed to the changing stoichiometric ratio in the flame, which affects the reaction mechanisms of nitrogen oxide formation from fuel-bound nitrogen in a way that inhibits nitrogen oxide formation. It is essential that the impulse of the oscillated air volume flow is high enough to periodically reduce the fuel concentration in the inner flame area. This results in changing local stoichiometric conditions in the inner flame area. Therefore, this effect could only be observed with high oscillated air volume flows from the annular gap R2.

During the reproduction experiments, 180 m_N^3/h of air was added from the annular gap R2 at an oscillating frequency of 2.67 Hz. This reduced the nitrogen oxide concentration in the flue gas by 45 % from 624 mg/m³_N at 0 Hz to 342 mg/m³_N at 2.67 Hz, based on 6% by volume oxygen. This result is in line with the above-mentioned finding that the level of the oscillated air volume flow is essential for nitrogen oxide reduction in the flue gas.

For modelling, the data were divided into training and test data. The training data included all attempts of statistical test planning in order to be able to reproduce the investigated influencing parameters in the model. The test data consisted of a random variation of the investigated parameters and only served to validate the trained model.

Statistical analysis of the experimental results and application of machine learning algorithms identified significant influences such as the oscillation frequency in R1 and R2 and their interaction with the respective volume flows of R1 and R2, the coal mass flow, the air ratio λ , and the swirl number S.

Further investigations on the influence of the swirl number S in combination with an oscillated combustion were not carried out.

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