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## Experiences from oxy-fuel combustion of bituminous coal in a 150 kW<sub>th</sub> circulating fluidized bed pilot facility

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### Abstract

The oxy-fuel combustion in a circulating fluidized bed was investigated in a 150 kW<sub>th</sub> pilot facility. A mixture of oxygen and recirculated flue gas was used for the combustion of bituminous coal. The experiments were set to observe the influence of operating conditions on gas concentrations of CO and NO<sub>x</sub> along with solid burn out. The results in regard to the difference between air and oxy-fuel atmosphere as well as an increase of the inlet oxygen concentration are presented in this paper and compared with results from other research groups.

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### 1. Introduction

The oxy-fuel combustion as a CCS technology has received increasing attention and progress has been summed up frequently in various publications [1] [2] [3]. Compared to the research performed on the oxy-fuel process dedicated to pulverized coal (PC) boilers, there is less to be found on the process applied to circulating fluidized bed (CFB) combustors.

The circulating fluidized bed technology is known for the ability to fire a number of different fuels and the possibility to perform at low levels of NO<sub>x</sub> and SO<sub>x</sub> without additional need for investment in flue gas cleaning equipment. When the oxy-fuel process for CO<sub>2</sub> capture is applied to PC boilers there is a need to recirculate part of

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the flue gas for temperature control. This is true also for CFB boilers, however, the solid inventory of such systems offers an additional means of temperature control. With the use of external heat exchangers some of the needed heat can be taken from the solids, allowing for lower flue gas recirculation and higher inlet oxygen concentrations. In this way the required boiler installation may be smaller than for an air fired plant of comparable power output. While some performed researches dedicated to the external heat exchanger [4] [5] this work aims at the performance of the combustion at the high oxygen concentrations that emphasize the advantage of oxy-fuel CFB combustion.

Although significantly smaller in numbers there have been some publications on the experimental investigation of oxy-fuel CFB combustion. The main differences between their setups is the composition of the gas entering the combustion chamber, being constituted of a mixture of either

- Air and O<sub>2</sub>
- CO<sub>2</sub> and O<sub>2</sub>
- Recirculated flue gas and O<sub>2</sub>

The last option is the actual setup that would be applied in a large scale boiler. While all cases are able to represent the inlet oxygen concentration during oxy-fuel combustion, they come along with differences regarding other gas species.

The first two options lack the high partial pressures of steam, unless additionally injected, that would be part of recirculated flue gas. The high concentration of nitrogen for oxygen enriched air (Air and O<sub>2</sub>) will lead to a reduced partial pressure of CO<sub>2</sub> in the combustion gas, whereas the CO<sub>2</sub> partial pressure observed in the second case (CO<sub>2</sub> and O<sub>2</sub>) might be even higher than in the recirculated flue gas case.

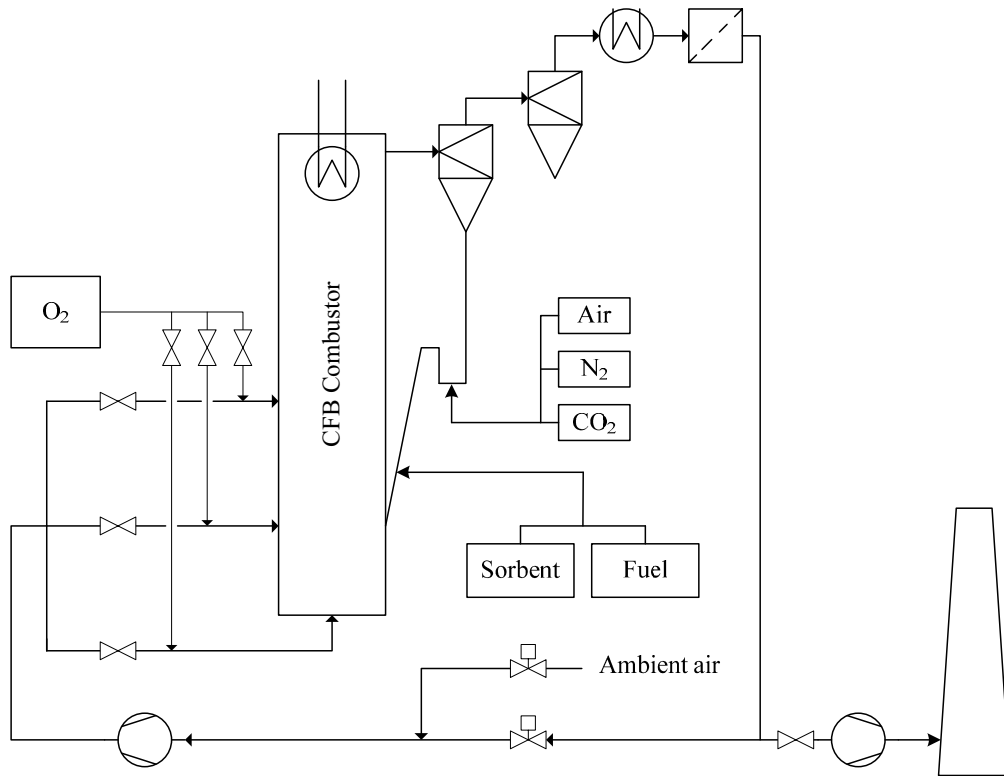
Nonetheless, experimental setups with any of the three gas mixtures listed can be found as their application increases in complexity from the first to the last one. All of them still deliver a valuable contribution to the investigation of oxy-fuel CFB combustion. Considering the difference in the setups the observations from all publications can still be compared to get a better impression of the behavior of combustion under oxy-fuel conditions.

Few results from experimental facilities with an actual mixture of recirculated flue gas and O<sub>2</sub> can be found in open literature though. This is in agreement with Lupianez et al. [6] who only list one research group [7] in his summary that uses this kind of setup. In this paper results from oxy-fuel combustion in a 150 kW<sub>th</sub> CFB pilot facility with recirculation of flue gas are presented.

## 2. Experimental

The facility is located at the Institute of Combustion and Power Plant Technology at the University of Stuttgart. The main components can be seen in Figure 1 and are described below. The core part is the 10 m high refractory lined riser. Combustion gases are supplied by a high pressure blower that may either use air or recirculate flue gases for the oxy-fuel case. In addition to the main gas supply through the windbox at the bottom, the reactor is equipped with two levels for staging purposes. All three supply lines are equipped with mass flow controllers for individual oxygen supply. This setup allows for the staging of gas flows as well as oxygen distribution in the oxy-fuel case.

Solids are separated from the gas in the cyclone and recirculated back to the bottom of the riser via the loop seal that may be fluidized with either air, N<sub>2</sub> or CO<sub>2</sub>. The hot gases leaving the reactor are cooled down to approximately 300°C in the gas cooler before they are cleaned of remaining fly ash in the bag filter and leave the facility through the ID fan. A pressure control valve allows moving the zero pressure point from inside the riser up to a point where the whole recirculation loop would be in overpressure. Behind the filter flue gases for recirculation in oxy-fuel mode may be extracted.

Figure 1 150 kW<sub>th</sub> circulating fluidized bed pilot facility

The reactor has six retractable water cooled rods at the top to extract the heat needed for temperature control in all cases.

Attached to the return leg there is a gravimetric dosing system with two dosing units for fuel (e.g. bio mass, coal) and two for solids in smaller quantities (e.g. sand, limestone).

Combustion tests were carried out with El Cerrejon bituminous coal. The composition is shown in Table 1.

Table 1 El Cerrejon bituminous coal

Ultimate analysis			Proximate analysis		
C	wt.-%, daf	80.3	ash	wt.-%, db	9.6
H	wt.-%, daf	5.3	water	wt.-%, ar	9.4
N	wt.-%, daf	1.9	VM	wt.-%, daf	40.3
S	wt.-%, daf	0.6	LHV	MJ/kg, ar	26.0

Sand was used as initial bed material. The facility was heated up prior to the test points with a natural gas burner. Each test point represents a one hour steady state. During the experiments solid samples from the fly ash directly behind the primary cyclone were taken as well as bed material from the loop seal.

The gas composition was measured continuously with online gas analyzers.

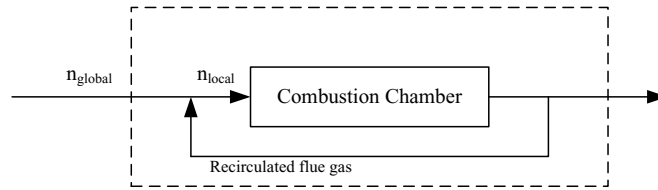


Figure 2 Oxygen ratio for oxy-fuel combustion

To keep residence times and state of fluidization comparable, all points were aimed to be executed at the same fluidization velocity. However, at a constant gas velocity an increase of the inlet oxygen concentration leads to a higher thermal firing power. The cooling rods were used to keep the combustor temperature equal nonetheless. The influence of inlet oxygen concentration as well as the change from air to oxy-fuel combustion on both gas concentrations and solid burnout was investigated this way.

Note that in oxy-fuel combustion there exist two oxygen ratios (see Figure 2), one referring to the oxygen coming from the oxygen supply only, which is referred to as  $n_{\text{global}}$ , which in a commercial boiler is linked to the energy demand for the air separation unit. The other one,  $n_{\text{local}}$ , refers to the oxygen concentration at the inlet of the combustion chamber also including the oxygen that is recirculated with the flue gas. As the oxygen ratio at the combustion chamber is the determining parameter for gas concentrations,  $n_{\text{local}}$  is used below as the reference oxygen ratio.

### 3. Results

The operation at oxy-fuel conditions showed no difference in the stability of the combustion and facility operation compared with the air points. As big efforts have been made in the past years regarding the tightness of the facility, no considerable false air could be seen during initial test runs for oxy-fuel combustion. Nonetheless, the whole gas recirculation loop was operated in overpressure to prevent air ingress.

The results of the measurements for three steady-states are presented and discussed below. All of them represent combustion at an upper riser temperature of  $\sim 880^\circ\text{C}$  at an oxygen ratio of  $\sim 1.1$ . Note that concentrations for CO and  $\text{NO}_x$  are in  $\text{mg}/\text{m}^3$  at STP conditions measured in the dry flue gas without reference to a specific outlet oxygen concentration.

Some additional operating conditions for the points shown are given in Table 2.

Table 2 Test point operating conditions

		Air	Oxy26	Oxy36
superficial velocity	m/s	5.4	4.4	4.4
$\text{O}_2$ at inlet	vol.-%	20.9	26.1	36.2
$\text{H}_2\text{O}$ at outlet	vol.-%	7.7	25.7	26.8
$\text{O}_2$ at outlet	vol.-%,dry	$1.7 \pm 0.3$	$2.9 \pm 0.2$	$3.6 \pm 0.4$
$\text{CO}_2$ at outlet	vol.-%,dry	$17.1 \pm 0.2$	$89.4 \pm 0.4$	$89.5 \pm 0.5$
oxygen ratio	-	1.09	1.1	1.09
upper riser temperature	$^\circ\text{C}$	$882 \pm 2$	$876 \pm 3$	$885 \pm 4$

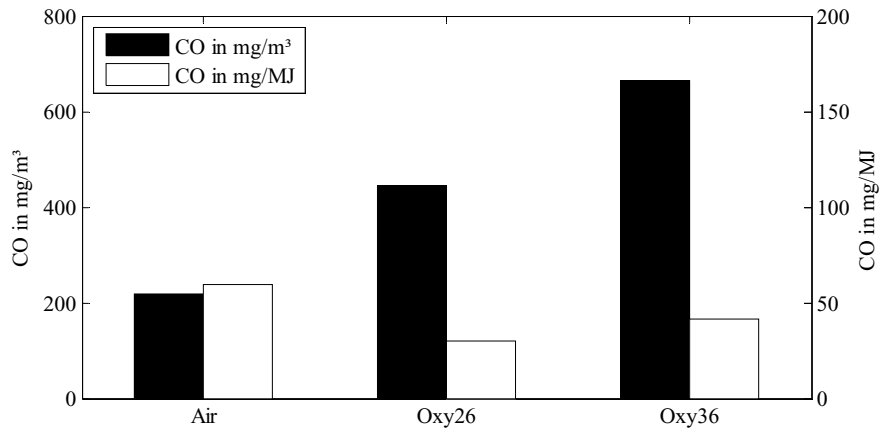


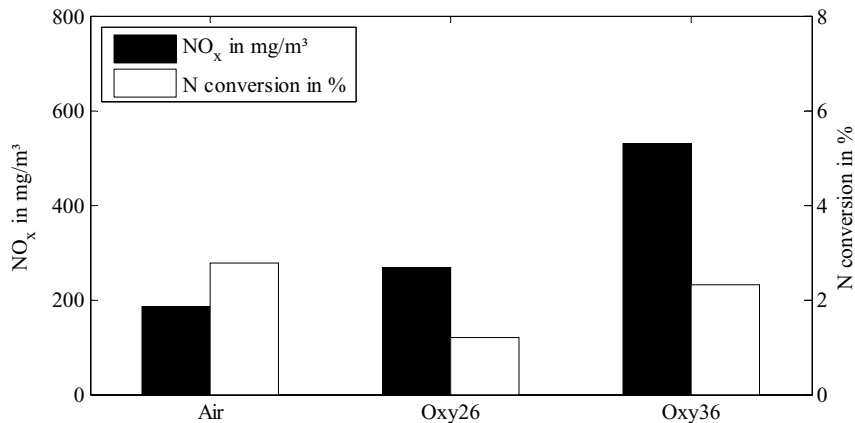
Figure 3 CO concentrations

CO concentrations, both in volume and energy based units, are shown in Figure 3. CO in oxy-fuel mode shows higher concentrations than in air mode and increases with higher oxy-fuel cases. While information on the trend between different oxy-fuel cases is hard to find, Jia et al. reported for both facilities used that CO concentrations in oxy-fuel mode were similar to air firing [7] [8], or slightly higher [9] at later experiments. Czakiert et al. showed lower CO concentrations with increasing inlet oxygen concentration without flue gas recirculation [10] but stated that this effect could partly be contributed to the combustion temperature also rising when increasing the load of the facility.

Energy based CO concentrations during the experiments were on the same level for air combustion and the two oxy-fuel cases investigated.

Summing up the reported findings with the work presented in this paper oxy-fuel CFB combustion seems to show equal or higher CO concentrations on a volumetric basis than air combustion, strongly depending on the fuel and the facility operating conditions.

The analysis of carbon in fly ash content from the solid samples show the same trend between oxy-fuel and air atmosphere as the CO concentrations, with 28.6% carbon in ash for air and 42.6% for the oxy-fuel case at 26 vol.-% inlet oxygen concentration. While the actual numbers are believed to be related to the facility conditions (e.g. the cooling rods at the top) the trend of increasing solid carbon losses observed itself is important to notice. A high partial pressure of  $\text{CO}_2$  as well as the higher thermal power for increasing inlet oxygen concentrations, together with the above mentioned facility setup could all contribute to an increase in CO concentrations and solid carbon losses during oxy-fuel combustion. However, the extent of each factor is hard to determine at this point. As mentioned above variation from low to high inlet oxygen concentrations has been subject to research in smaller numbers and there is very little to be found about this specific behavior, to compare the presented results with.

Figure 4 NO<sub>x</sub> concentrations

Volume based NO<sub>x</sub> concentrations and fuel-N conversion are shown in Figure 4. The nitrogen conversion relates the amount of nitrogen measured as NO<sub>x</sub> in the flue gas to the nitrogen fed to the combustor with the fuel. The fuel-N conversion is proportional to NO<sub>x</sub> concentrations given in mg/MJ. NO<sub>x</sub> concentrations in lower oxy-fuel mode compared to air combustion are at a comparable level and may be slightly higher or lower depending on the actual inlet oxygen concentration used while performing oxy-fuel combustion. A clearly increasing trend for NO<sub>x</sub> for higher oxy-fuel cases can be observed though. Fuel-N conversion in oxy-fuel combustion is noticeably lower than air values for all cases investigated and increases as well for rising inlet oxygen concentrations. Similar observations regarding the behavior for both concentrations and conversion rates were made by Jia et al. [7] [8] [9] during experiments with different fuels. Taking into consideration the fuel dependent behavior of NO<sub>x</sub> production, the results observed are in good agreement.

#### 4. Conclusions

Experiments for oxy-fuel combustion of bituminous coal in a circulating fluidized bed were carried out. A mixture of oxygen and recirculated flue gases was used as the fluidizing agent. No constraints regarding the stability of the facility and the combustion process could be found. Influence of operating parameters on the gas concentrations were investigated and verified for a number of test point. While CO concentrations increase for oxy-fuel combustion they stay at the same level on an energy based value. NO<sub>x</sub> concentrations also go up slightly while N-conversion rates are noticeably lower. All test point presented used oxygen concentrations at the exit of less than 4 vol.-%, dry, allowing for high CO<sub>2</sub> concentrations around 90 vol.-% in the dried gas for storage purposes.

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