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Experiments in a once-through furnace simulating different extents of recycle gas cleaning in coal-fired oxy-fuel combustion: sulphur oxide emission behaviour

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1. Introduction

With a growing awareness on the increased greenhouse gas emissions and its contribution to climate change, CO₂ capture and storage (CCS) technologies for coal-fired power generation were developed to concentrate the CO₂ for processing and sequestration. Oxy-fuel combustion is one of the CCS technologies. The focus of this work is to quantify SOₓ (SO₂ and SO₃) emissions during air and oxy-firing configurations with different extents of recycle gas cleaning. The SOₓ concentrations during oxy-fuel combustion are generally three to five times higher compared to conventional air firing [1-4]. The higher SOₓ concentrations, particularly SO₃ in combination with high concentration of water in the recycled stream, increase the acid dew point temperature of the system, thereby increasing allowable flue gas temperatures and reducing the thermal efficiency of the power plant [5].

2. Experimental setup

This paper presents results on the experimental work carried out at a 20 kW electrically heated once-through furnace of Institute of Combustion and Power Plant Technology (IFK) of the University of Stuttgart. This top fired rig is capable for combustion investigations with 0.5 up to about 3 kg pulverized fuel per hour and highly flexible feeding of oxidant gases such as air or mixtures of O₂, CO₂, H₂O and trace gases (e.g. NO, SO₂ and Hg) to evaluate air but also oxy-fuel combustion of solid fuels with simulated flue gas recycling. The system is illustrated in figure 1.

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The electrically heated furnace was operated at 1350°C, with a constant gas product rate of about 11.5 m³(STP)/hr (and variable fuel feed) to maintain comparable gas residence times in the system for all experimental settings. The top mounted burner is supplied with the pulverized fuel carried by a carrier oxidant gas stream and an additional primary and secondary oxidant gas stream. In conventional air combustion this oxidant streams are fed with cleaned and dried air, whereas for oxy-fuel experiments a mix of CO₂ and O₂ from tanks is fed to simulate flue gas recycling. In this way, oxy-fuel recycle rates can be set up highly flexible by adjusting the O₂ concentration in the oxidant. The oxidant O₂ concentration was fixed at 28 Vol.-% for all oxy-fuel experiments which corresponds to recycle rates of about 70%. To simulate recycling of flue gas impurities, the oxidant feed gas was doped with NOₓ, SO₂, Hg⁰ and H₂O vapour. With this configuration, the experimental rig is capable to simulate virtually every possible pulverized fuel oxy-fuel combustion process layout since the oxidant gas composition can be readily changed to simulate different recycle gas treatment configurations and impurity levels in coal. The application of this method allows an isolation of flue gas variables from other process variables and enables a mechanistic understanding of the impurity behaviour as well as of coal specific ash interactions.

In this study the following configurations that comprise extreme as well as industrially relevant cases (e.g. flue gas recycling with partial recycle gas drying, various degrees of recycle gas desulfurization and Hg removal) were investigated in particular:

- air firing
- oxy-fuel 1: combustion with pure CO₂/O₂
- oxy-fuel 2: simulated removal of 20% of H₂O and SO₂ and approx. 50% of Hg⁰
- oxy-fuel 3: simulated removal of 20% of H₂O, 50% of SO₂ and approx. 50% of Hg⁰
- oxy-fuel 4: simulated removal of 20% of H₂O and SO₂ and approx. 80% of Hg⁰
- oxy-fuel 5: simulated removal of 20% of H₂O, 0% of SO₂ and approx. 50% of Hg⁰
- oxy-fuel 6: simulated removal of 20% of H₂O and SO₂ and 0% of Hg⁰
- oxy-fuel 7: simulated removal of 20% of H₂O, 0% of SO₂ and 0% of Hg⁰
Three Australian coals have been used in this study. Their NCV, proximate, elemental and Hg analyses are presented in table 1.

Tab. 1: NCV, proximate, elemental and Hg analyses of the three investigated Australian coals

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During the experiments concentrations of SO₃ were measured discontinuously via the controlled concentration method at sampling positions before and after a baghouse filter. During the experiments the concentrations of O₂, CO, CO₂, SO₂ and NOₓ were recorded continuously. Besides the flue gas analyses, ash samples were collected from the filter during all experiments and analyzed for their composition.

3. Results

A comprehensive set of SO₂ and SO₃ concentrations was experimentally generated for various oxy-fuel combustion setups and three different coals. Based on that data, conversion of SO₂ to SO₃ was calculated and compared with existing literature. The generated results allow for a detailed analysis and evaluation of the process parameters that influence the SOₓ emissions of an oxy-fuel combustion process. The acid dew point temperatures (ADPs) for the exiting flue gas have been calculated for each of the firing conditions. SOₓ emissions and conversion of SO₂ to SO₃ are in good agreement with existing literature. Measured SOₓ concentrations show an increase when switching from air to oxy-fuel operation for all investigated coals and oxy-fuel settings, even when no SO₂ is injected to the oxidant gas. The ADP temperature has been found to increase by between 30 to 50K when changing from air to oxy-firing with recycle of H₂O and SO₂. The SO₃ capture by fly ash in the baghouse filter has also been investigated and was found to reduce the SO₃ emissions considerably. The reduction in SO₃ concentrations in the bag filter has been found to be equivalent to a reduction of the ADP temperature by as much as 15K.

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References