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Flue Gas Desulphurisation for Hot Recycle Oxyfuel Combustion: Experiences from the 30 MWth Oxyfuel Pilot Plant in Schwarze Pumpe

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1. Introduction

Coal-fired power plants are one of the major sources for CO_2 emissions. Therefore CCS (Carbon Capture and Storage) technologies are seen as an important option for the future operation of coal fired power plants from an environmental but also from an economical point of view.

Compared to other CO_2 capture technologies (post-combustion and pre-combustion) for coal-fired power generation, the oxyfuel combustion CO_2 capture is not a CO_2 selective method [1] but the CO_2 is enriched by recirculating the flue gas back to the combustion. Therefore, the main purpose of flue gas cleaning is to control the non-CO_2 components for both CO_2 capture and the boiler operation, instead of a pretreatment procedure for the selective capture of CO_2 from flue or fuel gases. A special focus is thereby on the desulphurization of the flue gas. In order to test the oxyfuel process Vattenfall has constructed an experimental large-scale pilot test facility. For flue gas desulphurization, a wet FGD system designed by Babcock Noell GmbH using the Babcock and Wilcox´s tray absorber technology with three spray levels and an external oxidation tank to avoid in-leakage of oxidation air into the system is used.

Although these flue-gas cleaning technologies have been proved in industrial scale for air pollution control requirements, the capabilities used in the gas cleaning for CO_2 capture purpose are required to be further investigated due to the following reasons:

- There are significant differences in inlet flue gas compositions under the oxyfuel combustion conditions compared with that under conventional air-firing conditions mainly in terms of CO_2, acidic gas components such as SO_2, SO_3/H_2SO_4 and moisture content,
- The gas cleaning requirements are based on downstream processes such as CO_2 processing/purification, transport, storage and environmental regulation, instead of the air emission limits,
- Removal of multiple components is generally required compared to for example the conventional FGD process,
- Much higher removal efficiencies could be required for the downstream CCS processes, which means significant mass transfer "work" will be required for such flue-gas cleaning processes, and
- Specific requirements such as e.g. CO_2 recovery and avoiding of non-condensable gas in-leakage, which are generally not required for conventional FGD processes.

During the test campaigns, the influence of certain operation parameters on the removal efficiencies have been investigated and the impact of the oxyfuel conditions on the removal efficiencies, the system chemistry and the operation of the desulphurization plant have been studied. The purposes of the tests and evaluations are:

- Comprehensive evaluation of the FGD performance in terms of removal of important flue gas components including not only SO_2 but also other acidic gas components such as SO_3/H_2SO_4, NOx, HCl and HF;
- Evaluation of the impacts of operational variables on the FGD performance under oxyfuel combustion conditions;
- Evaluation of major mass and heat balance in terms of important flue gas components and processes including the new emission characteristics of the FGD system;
- Understanding the important characteristics of the FGD processes under the oxy-coal combustion conditions and comparison with the situation under air-firing conditions and
- Investigate the potentials for further optimisation and improvement.

2. The Oxyfuel pilot plant in Schwarze Pumpe

Vattenfall has constructed an experimental large-scale pilot test facility for detailed investigation of the oxyfuel firing process. The plant is located southeast of Berlin in Germany in the vicinity of the existing lignite fired power plant Schwarze Pumpe. The pilot plant consists of a single 30MWth, top-mounted PF burner and the subsequent flue gas cleaning equipment and CO_2 processing. The flue gas cleaning equipment consists of an ESP (Electrostatic Precipitator) for particle removal, wet FGD (Flue Gas Desulphurisation) for removal of SO_2 and other acidic components and the FGC (Flue Gas Condenser) for cooling of the flue gases and removal of water. In addition to these components, a CO_2 liquefaction plant is placed downstream of the FGC to produce liquid CO_2 from the cleaned flue gases during oxyfuel operation. A cryogenic ASU located at the site supplies the plant with the gaseous oxygen needed for the combustion. The oxyfuel plant is operated with a hot flue gas recycle, with the recycle located after the ESP. With this configuration, sulphur-rich recirculation is tested with special attention to effects on boiler, ESP and flue gas recycle ducts.

The plant has been in operation since September 2008 and has so far accumulated about 8 000 operating hours until August 2010, of which more than 5 000 hrs in oxyfuel mode. The plant has proven that it is possible to operate continuously in oxyfuel mode under stable conditions.

2.1. The FGD system at the pilot plant

The FGD is based on the wet scrubbing process, using limestone as absorbent. The task is washing SO_2^-, SO_3^-, HCl^-, HF^-, residual dust emissions and other impurities out of the flue gas. Special attention had to be paid to:

- Operation of a desulphurization plant with a practically nitrogen-free, CO2-enriched flue gas.
- SOx removal of > 99 % at very high inlet concentrations (8.000 – 11.500 mg/m³ in N. dry).
- Oxidation of the slurry without dilution of flue gas.

The flue gas cleaning process and its auxiliary systems have been designed according to subsequent specifications:

	Technical data
Maximum flue gas volume flow (Nm³/h, dry)	8.200
Inlet concentration SO_2 (Nm³/h, dry) Inlet concentration SO_3 (Nm³/h, dry)	11.500 50
Emission data (guaranteed): SO_2-degreed of separation (%) and outlet concentration SO_2 (Nm³/h, dry)	 99 100
Contents of flue gas: N_2 (Ma-%) O_2 (Ma-%)	 6,5 3,5

CO_2 (Ma-%)	72,5
H_2O (Ma-%)	14,1
SO_2 (Ma-%)	0,5

Table 1 Specification Requirements

In order to meet the requirements for high removal efficiency, operation in a CO2-rich atmosphere and avoiding of air in-leakage into the system, the FGD system at the pilot plant has been specially designed for the operation in the Oxyfuel process. Some design features include:

- Build-in tray system in the absorber to enhance the mass transfer and meet the required high removal efficiencies,
- Oxidation and degassing of the suspension in a separate, internally splitted tank,

3. Experimental investigations and results

In order to determine the general performance parameters of the FGD and to assess the influence of the oxyfuel operation conditions on the removal efficiencies, a series of tests has been performed at the pilot plant in oxyfuel combustion conditions as well as under air-firing conditions. During the tests, the main performance parameters have been determined by measurements in the flue gas as well as frequent liquid slurry sample analysis. A series of operation parameters have been varied and the influence on the main performance parameters has been investigated. The parameter variation tests include variation of the pH value, variation of the oxidation air flow, variation of the slurry recirculation rate as well as the number of spray levels in operation,.

The results show that a good performance of acidic gas removal has been achieved under oxyfuel combustion conditions. For SO_2 an average removal rate under normal oxyfuel operating conditions of > 99,5% could be observed with an average outlet concentration of <30 mg/Nm³ dry. The removal efficiency for SO_3 was measured between 45% and 65% with outlet concentrations between 4,5 and 25 mg/Nm³ dry. For HCl and HF a removal efficiency of >95% could be observed.

In Fig. 1 the result of a variation of the pH value is shown. The expected strong dependency of the SO_2 removal efficiency on the pH value as well as the behaviour of the carbonate buffer can be seen. The influence of the higher partial pressure of CO_2 on the limestone dissolution and the absorption of SO_2 as well as on the gypsum quality is limited in the tested FGD system. Overall a good gypsum quality with an average gypsum content of >95% was achieved under oxyfuel conditions which is comparable to the by product quality under air-firing conditions.

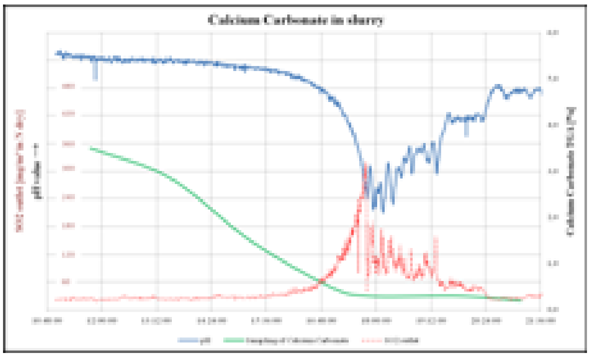

Fig. 1 Dependency of SO2 removal on pH value and slurry chemistry

No fundamental problems have been observed for the desulphurization processes under the oxyfuel combustion conditions. In the contribution, the results from the experiments will be shown and differences in the operation between oxyfuel and air-firing conditions will be analyzed.

[1] Anheden, M; Yan, J., De Smedt, G.: Denitrogention (or oxyfuel concepts). Oil & Gas Science and Technology - Rev. IFP, 60 (3): 485-495, 2005.