Nitrogenous emissions from the Delft Pressurized Fluidized Bed Combustor

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Abstract

Experimental and theoretical work on Pressurized Fluidized Bed Combustion (PFBC) of coal are main topics at the Laboratory for Thermal Power Engineering of the Delft University of Technology, The Netherlands. A sub pilot scale test rig is used operating at pressures up to 10 bar and a maximal thermal capacity of 1.6 MW.

At present, emphasis is laid on chemical reactions within the combustor which play a role in the formation and destruction of CO, CO₂, NO, N₂O, NO₂ and SO₂.

In the freeboard, downstream of the first cyclone and in the stack of the PFBC test rig gas concentrations have been measured using specially developed systems for flue gas sampling. Gas analysis has been performed using conventional gas analysis instruments and a Fourier Transform InfraRed (FT-IR) spectrometer.

The effect of ammonia injection in the exhaust of the combustor (SNCR) and staged combustion on the emissions was determined.

1. INTRODUCTION

Research into fuel conversion and related environmental aspects is carried out at the Laboratory for Thermal Power Engineering of the Delft University of Technology, The Netherlands. Experimental and theoretical work on Pressurized Fluidized Bed Combustion (PFBC) of coal using a sub pilot scale test rig operating at pressures up to 10 bar and a maximal thermal capacity of 1.6 MW, are among the main topics.

More recently work on environmental aspects like the influence of fuel-related and operation-dependent parameters upon the formation and reduction of noxious emissions has been carried out. At present, emphasis is laid on the chemical reactions within the combustor which play a role in the formation and destruction of CO, CO₂, NO, N₂O, NO₂ and SO₂.

2. EXPERIMENTS

2.1. Objectives

The objective of the experiments was to determine the influence of NO reduction techniques (SNCR and staged combustion) on the emission of nitrogenous species.

A new gas analysis system, based on the Fourier Transform InfraRed (FT-IR) spectrometry, was introduced and used for the analysis of nitrogenous species in the flue gas of the PFBC.

2.2. Experimental facility

Experimental research has been done using a semi-technical scale Pressurized Fluidized Bed Combustor. The main design data are given in Table 1. The fuel (coal), the bed material (i.e. sand and/or ash) and the additives for SO₂ reduction (dolomite or limestone) are supplied separately by double valve lock systems and fed together pneumatically into the bed through the air distributor plate. Part of the heat released in the bed is extracted by a vertical, water cooled, heat exchanger immersed in the bed. The freeboard zone is followed by one or two cyclones and, optional, a granular bed filter for fly ash precipitation. Figure 1 shows a scheme of the test rig. Different bituminous coals are used for the experiments. Properties of the coals are given in Table 3.

Table 1
Main design data of the PFBC test rig

Combustor height (m)	6.0
Bed diameter (m)	0.485
Bed height (m)	0.3 - 2.0
Freeboard height (m)	5.7 - 4.0
Operating pressure (MPa)	0.1 - 1.0
Fluidization velocity (ms ⁻¹)	0.8 - 1.2
Max. thermal cap. (MW)	1.6

Table 2
Technical data of the FT-IR equipment

<i>Spectrometer</i>	
Type	Mattson Polaris
Number of scans	64
Resolution (cm ⁻¹)	1
<i>Gas cell</i>	
Type	Infrared Analysis
Path length (m)	22
Temperature (°C)	35

Figure 1. The Delft PFBC test rig.

Table 3
Properties of the coals

Fuel	Kiveton	Westerholt	Bilsthorpe	Osterfeld
<i>dry (%)</i>				
Volatiles	31.5	38.4	31.7	22.8
Ash	15.1	4.7	19.3	29.0
Carbon	68.7	79.5	70.0	60.8
Hydrogen	5.1	5.1	5.4	3.7
Nitrogen	1.62	1.44	1.6	1.14
Oxygen	7.50	7.1	3.9	4.5
Sulphur	1.88	1.32	1.68	1.42

2.3. Gas composition determination

Gas sampling can be performed at different heights in the freeboard, downstream of the first cyclone and in the stack. Specially developed systems have been used for flue gas sampling: a nitrogen-quenched system in the freeboard and an air cooled system downstream of the first cyclone. In the stack an insulated, uncooled probe is installed. After sampling the particles in the gas are removed by cyclones and glass fibre filters. The gas is cooled to ambient temperature and transported to the gas analysis systems. Gas concentrations have been determined using a Fourier Transform InfraRed (FT-IR) spectrometer and 'conventional' gas analyzers (NDIR, NDUV, chemiluminescence, paramagnetism). Some technical data of the FT-IR equipment is given in Table 2.

The analysis of the flue gases reported here has been done at ambient temperature. This method has to be preferred for the measurement of components which are not influenced by the cooling and drying process, because of the major influence of the water vapour spectrum on the flue gas analysis.

In the experiments reported the FT-IR equipment is used for the analysis of CO, CO₂, NO, NO₂, N₂O, SO₂, H₂O, CH₄ and C₂H₄.

2.4 Selective Non Catalytic Reduction (SNCR)

The simplest way to remove NO from flue gases is through the injection of ammonia into the high temperature flue gases. This method, invented by Exxon [1], is known as the Thermal DeNO_x process. The NO formed during the combustion process is reduced by several non-catalytic gas phase reactions to molecular nitrogen.

The ammonia solution is evaporated, after which it is injected in the flue gas, upstream or downstream of the first cyclone, using a small amount of nitrogen.

2.5. Staged combustion

An other method to reduce the NO emissions is staged combustion. During the first stage of the combustion, with a primary air ratio less than 1, most fuel-N will be converted into N₂. The amount of CO and unburnt coal will be higher than at normal conditions. During the second stage of the combustion secondary air is injected and CO and unburnt char will be combusted. The N₂ formed during the first stage will not be oxidized to NO due to the low temperature. The combustion of CO and char leads to a normal combustion efficiency. The secondary air is injected in the centre of the freeboard at a location 2.5 m above the air distributor. The injection is in downward direction. During injection of secondary air the primary air amount (the air flow through the bed) and thus the primary

air ratio, is maintained constant.

3. EXPERIMENTAL RESULTS

3.1. Selective Non Catalytic Reduction

A number of experiments have been done at different injection temperatures, injection locations and amounts of NH_3 injected. Results are published elsewhere [2-4] and will only be summarized here: the effectiveness (the ratio between the number of NO moles removed and the number of NH_3 moles injected) increases with increasing injection temperature and decreases with increasing amount of NH_3 injected (figure 2). The NO reduction increases with increasing injection temperature and increasing amount of NH_3 injected. There is a substantial difference between the effectiveness and NO reduction for different coal types (figure 3). Measurements of the N_2O concentrations during the NH_3 injection experiments show no clear effect of NH_3 injection on the N_2O concentration.

Figure 2. NO reduction and effectiveness of NH_3 injection as a function of temperature (Kiveton Park, 8 bar, 0.8 ms^{-1}).

Figure 3. NO reduction and effectiveness of NH_3 injection at different type of coals (8 bar, 1.0 ms^{-1} , $T_{\text{bed}} = 800^\circ\text{C}$)

3.2. Staged combustion

A number of experiments have been done using different primary and secondary air ratios. During these experiments also more detailed measurements have been done in the freeboard. Figure 4 shows radial N_2O concentration profiles in the freeboard, measured at a probe in the lower part of the freeboard. The profiles are rather flat, showing a small deviation at the wall. These results show an increase of N_2O concentration with secondary air injection and hardly no change of the shape of the profile. NO concentration measurements give the same picture. Figure 5 shows the results of temperature measurements in the lowest part of the freeboard. This figure shows an increase of freeboard temperature with fluidization velocity and with the amount of secondary air. A decrease of temperature at the wall and hardly no change of the shape of the profile is shown. The centre line of the cylindrical combustor is at 243 mm. The increase of the fluidization velocity gives a shift of a part of the combustion from the bed to the freeboard. Secondary air injection causes combustion of unburnt char and CO in the freeboard.

Figure 4. Radial N_2O concentration profiles in the freeboard at different amounts of secondary air (Bilsthorpe, 4 bar, $T_{bed} = 810^\circ C$)

Figure 5. Radial temperature profiles in the freeboard at different fluidization velocity and different amounts of secondary air (Kiveton Park, 4 bar, $T_{bed} = 800^\circ C$).

Increasing the pressure seem to decrease the N_2O and NO emissions, but this effect is

not clear. A decrease of the primary air ratio gives a decrease of the NO emissions (Figure 6). Increasing the secondary air ratio gives an increase of the N₂O and NO emissions (Figure 6). In the freeboard a substantial reduction of the NO and N₂O concentrations takes place. The experiments show an influence of the coal type on the emissions, but this is not clear.

Figure 6. NO and N₂O concentrations at different primary and secondary air ratios (Westerholt, 4 bar, $T_{bed} = 800^{\circ}\text{C}$)

4. CONCLUSIONS

Selective Non Catalytic Reduction using NH₃ injection can reduce NO_x emissions to a low level. A negligible effect has been noticed on N₂O emissions.

Staged combustion using air staging is also an effective method to reduce NO_x emissions, it gives low NO with low CO emissions, but attention would be given to the N₂O emissions. The influence of coal type is not very clear. The decrease of the primary air ratio gives a decrease of the NO emissions. An increase of the secondary air ratio increases the NO and N₂O emissions. The freeboard plays an important role in NO and N₂O destruction.

Fourier Transform InfraRed spectrometry can successfully be used for the analysis of flue gases from a PFBC.

5. ACKNOWLEDGEMENTS

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6. REFERENCES

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Discussion

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Question: F. Kapteijn

1. What causes the further reduction of NO between the top of the freeboard and point 6 (after the first cyclone)? Is the freeboard sufficiently large?
2. What is the effect of pressure on the results?
3. Why does the N_2O concentration increase with increasing secondary air injection?

Answer

1. The results indicate that the process of NO reduction is going on after the top of the freeboard. Especially in the cyclone, with the intense mixing of particles and gases, NO reduction (homogenous (CO) and heterogenous (char)) takes place. So, for the chemistry the freeboard is, at staged combustion conditions, not sufficiently large.
2. The effect of the pressure is not very clear in these experiments. A careful conclusion from the results is that increasing pressure decreases NO and N_2O emissions.
3. At secondary air injection oxygen is added to the process. Thus, oxidation takes place of precursors of both NO and N_2O to NO and N_2O , respectively. So increasing secondary air injection increases NO and N_2O emissions.

Question: W. Prins

Referring to the SNCR: Is the NH_3 introduced by a single point injection? If so, how can you be sure that the mixing with flue gas will be sufficient to achieve the optimal result?

Answer

Yes, the NH_3 is injected by a single point injection. At the point of injection there is a very turbulent flue gas flow, so the mixing will be very good.

Question: M.J. Cooke

If SNCR were to be applied to a PFBC plant, could NH_3 slip be a problem?

Answer

In the near future we will do experiments to determine, to measure, the NH_3 slip at SNCR. We want to use the FT-IR spectrometer equipped with a heated ($150^\circ C$) gas cell. The results indicate decreasing efficiency with increasing NH_3 amount injected. This NH_3 can be lost by NH_3 slip or by oxidation to NO. At the moment we do not exactly know what happens: the experiments must clarify it.