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METHODS FOR REDUCING THE EMISSION OF NITROUS OXIDE FROM FLUIDIZED BED COMBUSTION

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Abstract - Two methods for the reduction of nitrous oxide emissions, afterburning and reversed air staging, are investigated in a 12 MW circulating fluidized bed boiler. With *afterburning* the N_2O emission can be reduced by 90% or more, using an amount of secondary fuel corresponding to 10% of the total energy input. With *reversed air staging* it is possible to reduce the emission of N_2O to one fourth (25 ppm), without significantly affecting the other emissions. With reversed air staging no secondary air is used in the combustor and an air-ratio of about unity is maintained throughout the combustion chamber. Air for final combustion is added in the cyclone outlet.

INTRODUCTION

Fluidized bed combustion technology has received a rapidly growing interest, and a large number of commercial boilers of this type have been put into operation during the last 20 years. Important reasons for this development are low emissions of nitric and sulphur oxides (NO and SO_2) and fuel flexibility. The low emission of nitric oxide is a consequence of low temperature and of reduction processes during combustion. The low sulphur dioxide emission is a result of limestone addition to the combustion chamber which then acts as a reactor for sulphur capture. Thus, low emissions of nitric and sulphur oxides are realized without equipment for flue gas cleaning downstream of the combustion chamber.

For more than five years, however, it has been known that fluidized bed boilers (FBBs) differ from conventional boilers by emitting considerable amounts of nitrous oxide (N_2O), owing to the low combustion temperature. N_2O is a greenhouse gas and is also believed to contribute to the ozone depletion in the stratosphere. In order to benefit from the low emissions of NO_x and SO_2 from FBBs, a solution is needed for the N_2O emissions.

It is well known that the emissions of NO_x, SO_2 and N_2O can be significantly affected by changes in operational parameters like bed temperature and air supply. The problem is, that, while a measure taken to decrease one of the emissions may prove successful, it has the opposite effect on one or two of the others. The situation can be summarised as follows:

Raised bed temperature: N_2O decreases, but NO increases and the sulphur capture efficiency is considerably reduced.

Lowered air-ratio: N_2O and NO decreases, but the sulphur capture efficiency is considerably reduced.

Lowered fraction of primary air (increased degree of air staging): NO decreases and N_2O decreases somewhat, but the sulphur capture efficiency is considerably reduced.

In conclusion, it is not possible to obtain a major reduction of the N_2O emission from changes in parameters of operation, because of the negative effects on other emissions.

However, this coupling of positive effects on N_2O to negative effects on other pollutants can be circumvented by addressing the conditions in the upper and the lower parts of the combustion chamber separately. This is possible because of a difference between the emissions with regard to the conditions in the upper and lower part: The conditions in the upper part are important for the N_2O emission, while the effect of the conditions in the lower part is small. The NO emission and the sulphur capture, on the other hand, are affected by the conditions both in the lower and in the upper part of the combustion chamber.

The present paper reviews two methods of N_2O reduction which selectively influence the conditions in the upper and lower part, one method which uses temperature as parameter and one which uses air supply:

1. Afterburning. This method involves extra fuel addition in the upper part of the combustion chamber (*i.e.* in the cyclone inlet) thereby increasing the temperature in the cyclone while the bed temperature can be maintained unchanged. Thereby a significant decrease in N_2O can be obtained without affecting the conditions for low NO and SO_2 emissions.

2. Reversed air staging. This method involves a reversal of the conditions normally used, supplying more oxygen to the bottom part and less to the upper part. Such a reversal of the conditions compared to normal air staging can be accomplished as follows: The combustor air-ratio is kept close to unity. No secondary air is used in the combustion chamber and all air is added in the bottom zone, except for some air which is supplied for final combustion downstream the cyclone, giving a total air-ratio of 1.2. The increased air-ratio of the bottom part makes this part more oxidising compared to normal air staging. The gradual consumption of oxygen with height, decreases the average oxygen concentration from the bottom and upwards, approaching very low oxygen concentrations in the top zone of the combustion chamber and the cyclone, since the combustor air-ratio is kept at about unity.

EXPERIMENTAL CONDITIONS

The 12 MW circulating FBB used for the experiments has the features of a commercial boiler, but was built for the purpose of research. The boiler is shown in Fig. 1. The height of the combustion chamber is 13.5 m and the square cross-section is about 2.5 m^2. The fuel used was bituminous coal. Further details on the boiler, the operating conditions, the properties of fuel and limestone and the system for data collection including gas analysis, are given in [1-5].

Afterburning. During the afterburning tests, the temperature in the cyclone and in the uncooled exit duct after the cyclone outlet was increased by fuel injection in three positions in the cyclone inlet (FI in Fig. 1). In some cases additional air was supplied in the cyclone outlet (r5). Five injection fuels were tested - liquid petroleum gas (LPG), fuel oil, pulverized wood, sawdust and pulverized coal - although the data presented below are for gas if not otherwise indicated.

Reversed air staging: The air supplied to the bottom bed corresponds to an air-ratio of about 1, *i.e.* stoichiometric conditions. Since no secondary air is added in the combustion chamber, this air-ratio (the *combustor air-ratio*) is maintained in the entire combustion chamber and the cyclone. Additional air is supplied in the cyclone outlet (r5) giving a *total air-ratio* of 1.2. The reversed air staging is compared to normal air staging with 60% primary air supplied to the bottom bed and 40% secondary air supplied at 2.2 m height (r2), *i.e.* with an air-ratio of about 0.7 in the bottom part and 1.2 above the secondary air supply.

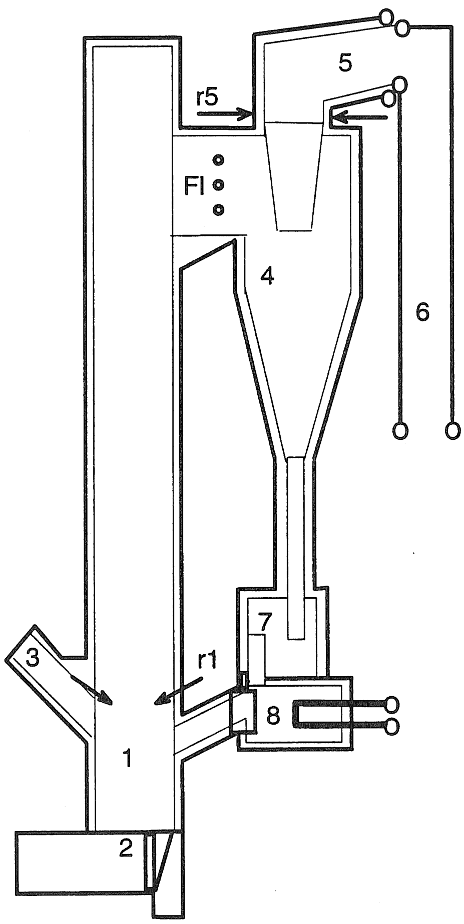

Fig. 1. Chalmers 12 MW boiler. 1. combustion chamber; 2. air plenum; 3. fuel feed chute; 4. cyclone; 5. exit duct; 6. convection cooling section; 7. particle seal; 8. particle cooler; → r1 and r5 secondary air nozzle inlets; ∘ FI fuel injection inlets.

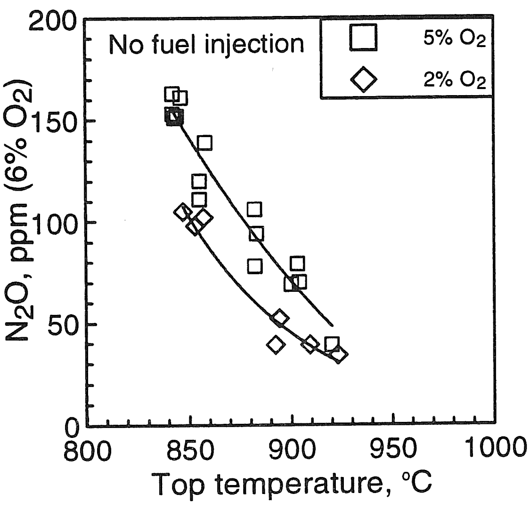

Fig. 2. N_2O emission without afterburning vs. combustion chamber temperature.

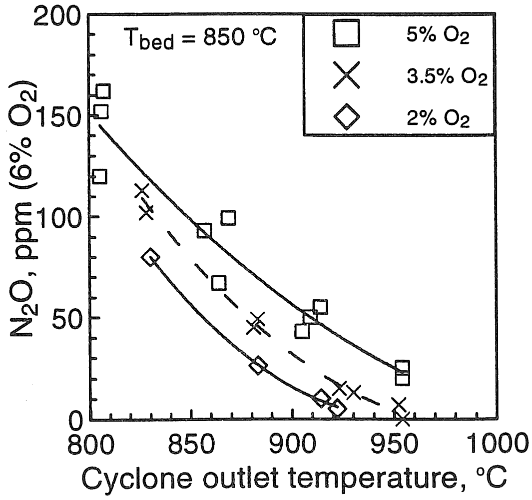

Fig. 3. N_2O emission vs. cyclone outlet temperature. Excess air-ratios (expressed as O_2) refer to values before fuel addition.

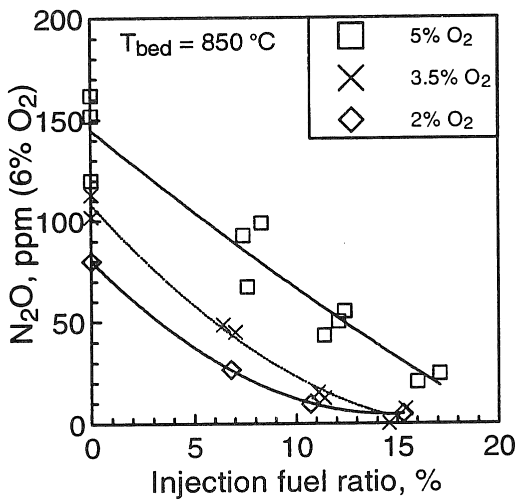

Fig. 4. N_2O emission vs. injection fuel ratio.

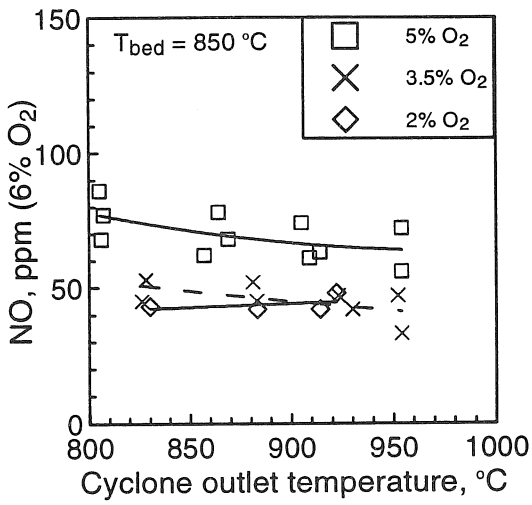

Fig. 5. NO emission vs. cyclone outlet temperature.

AFTERBURNING

The effect of air-ratio and temperature on N_2O without fuel injection is illustrated by Fig. 2. Here the temperature and the air-ratio have been changed in the entire combustion chamber, which causes a significant decrease of the N_2O emission, but also high emissions of NO and a very inefficient sulphur capture. With fuel injection in the cyclone inlet the temperature in the cyclone can be increased, while a constant bottom bed temperature is maintained. Thus, a similar decrease in N_2O, as in Fig. 2, is obtained without affecting the conditions in the combustion chamber, Fig. 3. The lines showing decreasing N_2O concentration as a consequence of an increasing temperature are the effect of a gradually increased rate of fuel injection. This is seen in Fig. 4 where the same data are shown versus the injection fuel ratio, *i.e.* energy in afterburning fuel divided by energy in primary fuel.

As seen in Fig. 5 there is no negative effect of fuel injection on the NO emission, instead the NO decreases slightly with increased temperature. Even though the temperature effect is restricted to the cyclone, a negative effect on NO would be expected; the reason why this is not the case is probably the

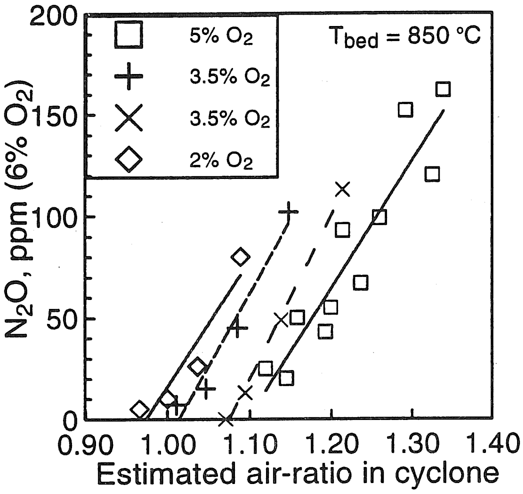

Fig. 6. N_2O emission vs. air-ratio in cyclone.
◇, + air addition in cyclone outlet.

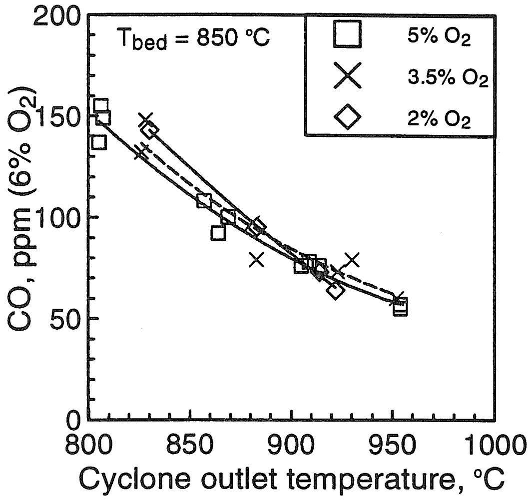

Fig. 7. CO emission vs. cyclone outlet temperature.

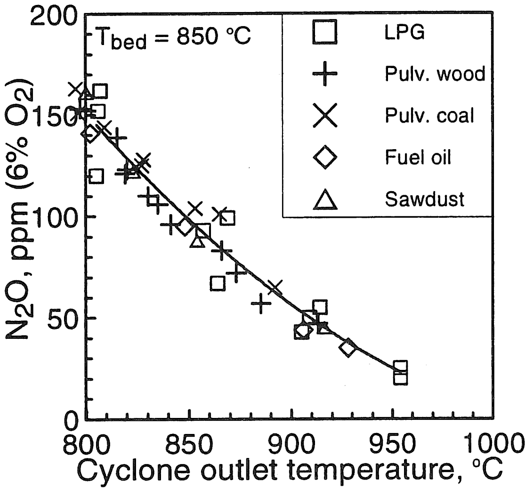

Fig. 8. N_2O emission vs. cyclone outlet temperature. Comparison between different afterburning fuels.

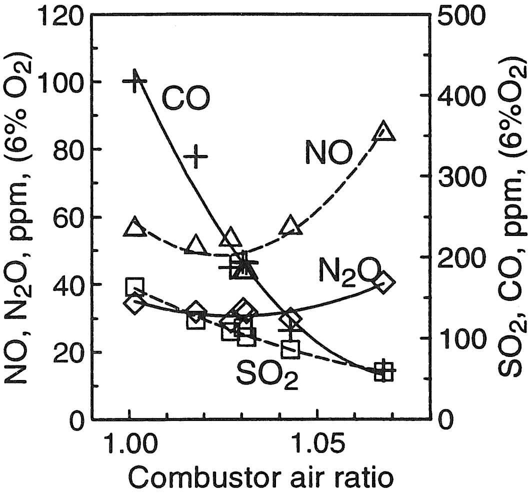

Fig. 9 Emissions vs. combustor air-ratio under reversed air staging.

simultaneous decrease of air-ratio in the cyclone, Fig. 6. The increased temperature in the cyclone also contributes to a decreased CO emission, Fig. 7.

Most of the experiments with afterburning were made on a sand bed, *i.e.* with no limestone addition. The test series performed with limestone addition showed no effect on the sulphur capture efficiency, [1]. These tests were only made for the higher air-ratio, *i.e.* for an initial oxygen flue gas concentration of 5%.

A comparison to other injection fuels, Fig. 8, shows that all the fuels tested cause a similar reduction in N_2O.

REVERSED AIR STAGING

With a bituminous coal of normal sulphur content, reversed air staging, if compared to normal air staging, has been shown to reduce the N_2O emission to one fourth (25 ppm) and NO to half (40 ppm) with a maintained sulphur capture efficiency of 90%, although the CO emission increases, [3-5]. This result is one example of what can be obtained with reversed air staging. For an optimization of the emissions three parameters are available:

i) combustor air-ratio: an increase gives lower CO and SO_2 and higher NO and N_2O,

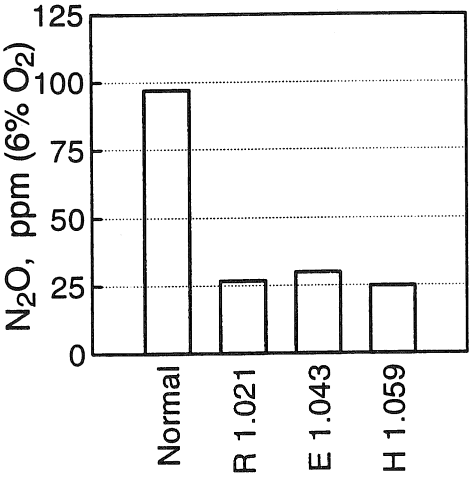

Fig. 10. Emission of N_2O for three cases of reversed air staging compared to normal air staging.

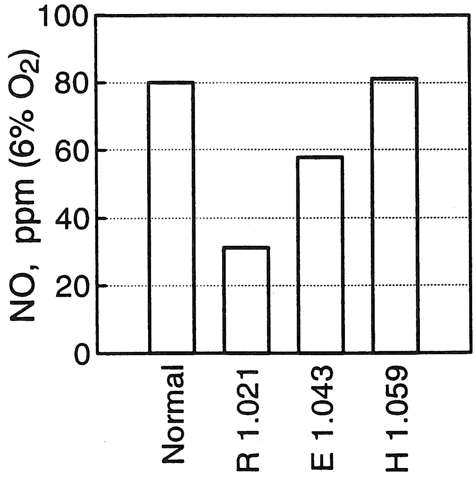

Fig. 11. Emission of NO for three cases of reversed air staging compared to normal air staging.

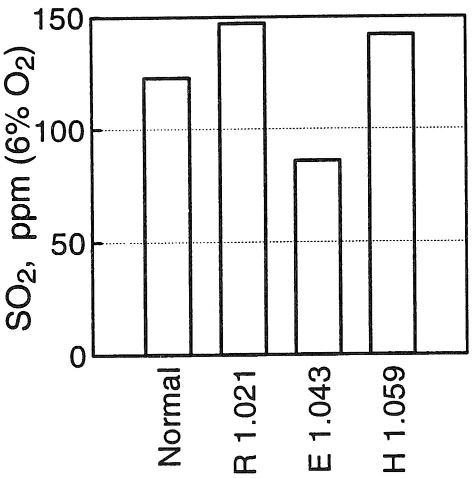

Fig. 12. Emission of SO_2 for three cases of reversed air staging compared to normal air staging.

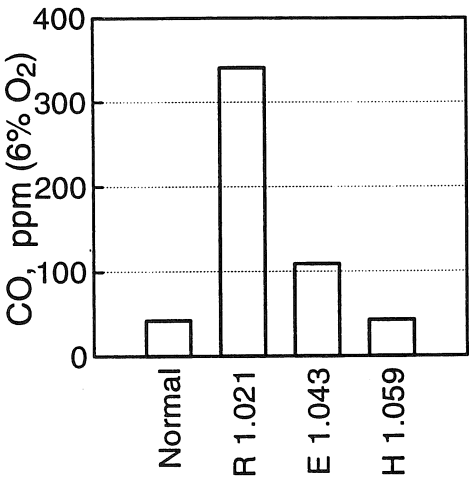

Fig. 13 Emission of CO for three cases of reversed air staging compared to normal air staging.

ii) temperature: an increase results in lower CO and N_2O and higher NO and SO_2,

iii) limestone addition: an increase gives lower SO_2, somewhat lower CO and N_2O and higher NO. Increases in the first two parameters can also be expected to increase the combustion efficiency which needs to be considered in an optimization. The effect of the first parameter, combustor air-ratio, is illustrated by Fig. 9.

Figures 10-13 show the emissions during normal air staging compared to reversed air staging. The reversed air staging cases are carried out with normal limestone addition (R), extra limestone addition at a somewhat higher combustor air-ratio (E), and higher bed temperature in combination with extra limestone and a high combustor air-ratio (H). The combustor air-ratios are indicated by the numbers in the x-axis labels. The three cases of reversed air staging in the figures were chosen to have approximately constant N_2O and SO_2 emissions. The cases show that the CO emission can be reduced from more than 300 ppm to the level achieved with normal air staging, below 50 ppm. The penalty of the decreased CO emission, however, is that the halving of NO is lost.

In conclusion, it is possible to reduce N_2O to one fourth with the emissions of NO, SO_2 and CO being unaffected. If a rise in CO is accepted, an additional reduction of the NO emission can be obtained. A further reduction of the emissions can be obtained by lowering the load, [5].

DISCUSSION AND CONCLUSIONS

Two methods for decreasing the N_2O emission have been presented. Both methods are based on the same principle, *i.e.* that the N_2O emission, as opposed to the emissions of NO and SO_2, is only affected by the conditions in the cyclone and the upper part of the combustion chamber. The insensitivity of N_2O to the conditions in the lower part is explained by the destruction of N_2O in the combustion chamber. The destruction has been verified by addition of N_2O to the bottom part of the Chalmers FBB; only a very small part of the added N_2O remained in the flue gas, [6]. Despite the destruction, gas measurements inside the combustion chamber show a marked N_2O increase with height owing to simultaneous formation, [7].

The two methods investigated both adapt the conditions in the upper part and/or the cyclone to N_2O reduction, while providing conditions in the lower part of the combustion chamber suitable for sulphur capture and low NO emissions.

Afterburning decreases the N_2O emission with up to about 90%, without significant effects on other emissions, except a decrease in CO. The drawback of the method are the extra costs involved in fuel injection, *e.g.* a more expensive fuel and arrangements for efficient afterburner construction.

Reversed air staging is able to reduce the N_2O emission to one fourth, without significant effects on the other emissions. A minor drawback of reversed air staging is a somewhat higher power consumption for the supply of air to the bottom part.

The combustion efficiency is important and, in the first available measurements, this was seen to be lower for reversed air staging. These results, however, were obtained for high CO emissions, 300-400 ppm, [3]. Recent investigations indicate that the combustion efficiency is correlated to the CO emission, and no significant difference in combustion efficiency between normal and reversed air staging was seen for lower CO emissions, *i.e.* 100-200 ppm CO. Considering the means available to reduce the CO emission, it should thus be possible to realize a good combustion efficiency.

The two methods have been tested in a boiler under a number of different conditions. Although the methods have been proved in this scale, further work would of course be needed to find the best design and operating conditions for a commercial application.

The present experience demonstrates the applicability of the methods. A comparison between the methods has to await a commercial implementation but afterburning can be expected to involve higher costs, while on the other hand a higher reduction of N_2O is possible. The two methods should also be possible to combine, which, hopefully, could result in even lower emissions.

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