Low N$_2$O, NO and SO$_2$ emissions from circulating fluidized bed boilers

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LOW N$_2$O, NO AND SO$_2$ EMISSIONS

FROM CIRCULATING FLUIDIZED BED BOILERS

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ABSTRACT

A new method, reversed staging, for decreasing N$_2$O without increasing the emission of the other pollutants, NO$_x$ and SO$_2$, was investigated in the 12 MW circulating fluidized bed boiler at Chalmers University of Technology. It was possible to reduce the emission of N$_2$O to one fourth (25 ppm), NO to half (about 40 ppm) compared to normal staging and normal temperature, without significantly affecting the sulphur capture efficiency (about 90%).

Air staging, which is normally used in circulating fluidized bed boilers means that only a part of the combustion air, primary air, is added to the bottom zone, resulting in a lower oxygen concentration in the bottom part, while the secondary air results in more oxidizing conditions in the upper part of the combustion chamber and the cyclone. The principal idea of *reversed staging* is to reverse the conditions in top and bottom, *i.e.* to decrease the oxygen concentration in the upper part and to increase it in the bottom part. Such a reversal is accomplished by adding air in the bottom corresponding to an air ratio of approximately unity. No secondary air is added to the combustion chamber which means that the oxygen concentration will be low in the upper part of the combustion chamber and the cyclone. Air for final combustion is added in the cyclone outlet.

NOMENCLATURE

K_c ratio of theoretical flue gas (including moisture) to theoretical air [-]

O_2 oxygen concentration in flue gas including moisture (=O2,w in Table 3), [%]

$O_{2,c}$ oxygen concentration in gas from cyclone, Eq. (5), [%]

λ_{tot} total air ratio, [-]

λ_c combustor air ratio, Eq. (6), [-]

INTRODUCTION

The problem

N$_2$O is a greenhouse gas and is also believed to contribute to the ozone depletion in the stratosphere. The discovery of considerable emissions of N$_2$O from fluidized bed combustion, compared to other types of boilers, shifts the image of fluidized bed combustion technology from "clean" (low NO$_x$ and SO$_2$) to "dirty" (N$_2$O unresolved).

The processes involved in formation and destruction of NO and N$_2$O are complex and not completely understood. The same also applies to the sulphur capture process, which involves release of sulphur from the combustibles, reaction with CaO to CaSO$_4$ and reductive decomposition of CaSO$_4$ (see *e.g.* Lyngfelt and Leckner, 1993a).

It is well known that the emissions of NO$_x$, SO$_2$ and N$_2$O can be significantly decreased or increased 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 summarized as in Fig. 1:

Increased bed temperature: N$_2$O decreases, but NO increases and the sulphur capture efficiency is considerably reduced.

Increased degree of air staging: NO decreases, N$_2$O decreases somewhat, but the sulphur capture efficiency is considerably reduced .

Air staging

By air staging is meant that a part of the combustion air is introduced in the form of secondary air at a later stage in the combustion process. The degree of air staging can be increased by lowering the primary air ratio (=total air ratio×primary air fraction), by raising the position of secondary air addition in the combustor, or both. These measures increase the presence of
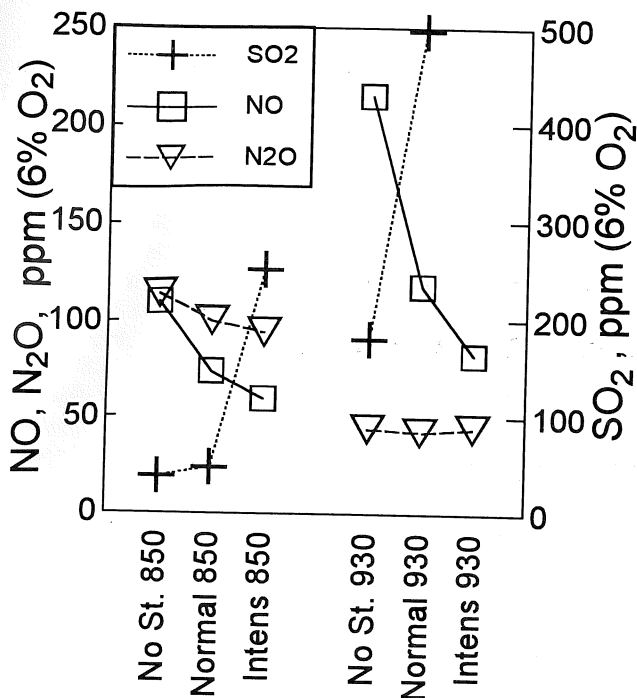
FIG. 1. EFFECT OF TEMPERATURE (850 AND 930°C) AND AIR STAGING (NO STAGING, NORMAL STAGING AND INTENSIFIED STAGING) ON THE EMISSION OF SO_2, NO AND N_2O.
DATA FROM LYNGFELT AND LECKNER (1993b).

zones with reducing conditions, which is assumed to be the most important effect of air staging as far as emissions are concerned. Another measure giving a similar effect is lowering the total air ratio.

A lowered primary air ratio means a reduced access to oxygen in the bottom part of the combustion chamber resulting in more reducing conditions, and thus affecting combustion and other chemical reactions. Furthermore, the concentration of combustible particles in the system increases and part of the combustion is moved upwards from the bottom zone. In addition, the changed gas velocity in the bottom part will affect the bed behaviour and particle movement. Thus, the total effect of reducing the primary air ratio involves changes throughout the combustion chamber and the final impact on the complex NO_x/N_2O and SO_2 chemistry is not fully understood. The general effect of increasing the presence of zones with reducing conditions is, however, well known: NO and N_2O emissions decrease while the SO_2 emission increases.

Local effect of reducing conditions on emissions

In order to improve the understanding of processes related to the emissions of NO_x, N_2O and SO_2 a comprehensive test series was performed at Chalmers 12 MW circulating fluidized bed boiler (FBB). The series involved measurements of gas components inside the combustion chamber for different cases of air staging in order to investigate the effect of air staging on gas concentration profiles in the combustion chamber. (These results will be reported separately.) The preliminary results of

the test series suggested a *difference*, in the effect of reducing conditions, between the various emissions depending on where the reducing conditions appear in the furnace. From this, the following questions evolved:

1) Is the relative effect of reducing/oxidizing conditions markedly different for the three types of emission in different parts of the furnace?

2) If so, could the reducing/oxidizing conditions in the various parts of the furnace be selectively adjusted to suit such a difference in order to give a simultaneous reduction of all emissions (or a reduction of the N_2O emission without increasing the others)?

Or more specifically:

1) Could more oxidizing conditions in the bottom part of the furnace in combination with more reducing conditions in the upper part have the effect of simultaneously decreasing the NO, N_2O and SO_2 emissions? (The idea is that the more reducing conditions in the top region have a greater effect on NO_x and N_2O compared to the more oxidizing bottom part, while the more oxidizing conditions in the bottom override the negative effect of more reducing conditions in the top for SO_2 capture.)

2) If so, is it possible to shift the conditions sufficiently both in top and bottom regions for such an improvement to take place?

These questions led to an additional test case, called reversed staging, which is the focus of the present paper.

Reversed staging

A circulating fluidized bed boiler is normally operated with a significant amount of secondary air and the bottom part of the combustion chamber is to a great extent under reducing conditions, as shown by zirconia cell measurements (Lyngfelt et al., 1993), while oxidizing conditions in the upper part are necessary to obtain acceptable sulphur capture and combustion. In addition to air staging, the gas flow pattern in the bottom bed, with considerable amounts of air by-passing the dense particle phase, contributes to make the bottom part more reducing.

In order to reverse the situation with oxidizing conditions in the upper part and reducing conditions in the lower part, *i.e.* to obtain more oxidizing conditions in the lower part and less oxidizing conditions in the upper part, the following strategy was used:

The combustor[1] air ratio was kept close to unity. No secondary air was used and all air was added in the bottom zone except for some air which was added for final combustion after the cyclone giving a total air ratio of 1.2.

The increased air ratio of the bottom part will make this part more oxidizing compared to normal staging. The gradual consumption of oxygen with height will give a decrease in 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.

This method of operating a boiler, *reversed staging*, requires that air can be added in the outlet of the cyclone and that there is a chamber after the cyclone outlet for final combustion.

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[1]By combustor or furnace is here meant the combustion chamber including the particle separator (cyclone), but not including the duct after the cyclone.

THEORY

Reducing conditions

Reducing conditions designate an understoichiometric gas mixture, *i.e.* the oxygen is not sufficient to burn the combustible gases present. This state can be measured by a zirconia cell probe which yields the equilibrium oxygen concentration. Under reducing conditions the equilibrium oxygen concentration is below 10^{-6} bar, normally 10^{-10} to 10^{-15} bar. Reducing conditions may appear locally in the vicinity of burning particles, in the bottom zone because of air staging, and are enhanced in the dense bottom bed because of by-passing of air in bubbles and jets. Measurements show rapid variations between oxidizing and reducing conditions, and a variation in the degree of air staging affects the *fraction of time* in which a location is under reducing conditions. For example, shifting from normal air staging to no air staging, *i.e.* from a primary air ratio of about 0.7 to 1.2, gave a decrease in the fraction of time under reducing conditions from about 0.8 to 0.1 at 0.65 m from the bottom (Lyngfelt et al., 1993).

The following will focus on the effect of oxygen concentration and the fraction of time under reducing conditions in various parts of the boiler.

Sulphur capture

In presence of O_2 the sulphur emitted from the fuel is oxidized to SO_2. The emission of SO_2 can be decreased by the addition of limestone, which after calcination and in presence of O_2 reacts with SO_2

$$SO_2 + CaO + \tfrac{1}{2}O_2 \rightarrow CaSO_4 \qquad (1)$$
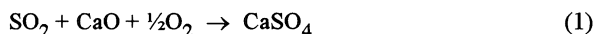
Under reducing conditions, reaction (1) may be reversed in presence of reducing gases such as CO and H_2

$$CaSO_4 + CO \rightarrow CaO + SO_2 + CO_2 \qquad (2)$$
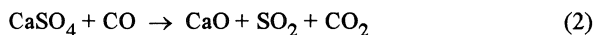
Alternatively $CaSO_4$ may first be reduced to CaS (*e.g.* in the lower part of the combustor) which could later be oxidized under SO_2 release (*e.g.* in the upper part).

The release of SO_2 takes place only when sorbent particles are exposed to reducing conditions; the oxygen concentration as such should not affect the sulphur capture. From the fundamental knowledge of the sulphur capture reactions it is difficult to draw any safe conclusions on the effect of reducing conditions on sulphur capture performance in the different parts of the combustion chamber. Experiments, however, clearly show that an increased fraction of time under reducing conditions in the bottom zone, *i.e.* increased air staging, is detrimental to sulphur capture performance. A decrease in the total air ratio is negative for sulphur capture performance, but if this should be attributed to changed conditions in the lower or upper part is not clear.

NO and N₂O

The reactions involved in N_2O and NO formation and destruction have been recently reviewed, for instance by Wójtowicz et al. (1993). Although a number of homogeneous and heterogeneous reaction mechanisms are known from laboratory measurements, the translation of this knowledge to the conditions of a circulating fluidized bed boiler still requires

further work. Some empirical knowledge obtained from measurements can, however, be used:

The N_2O concentration increases with height in the combustion chamber (Åmand and Leckner, 1993, 1994b). The formation of N_2O in the bottom zone is high, but its contribution to the emission of N_2O is low, because of the high rate of reduction along the gas path through the combustion chamber (Åmand and Leckner, 1994a). Consequently, the effect of staging on N_2O, *e.g.* Fig. 1, is small as long as it only affects the bottom zone. The response to changed conditions in the upper part of the combustion chamber is not sufficiently investigated, although some literature data are available. Support for the importance of the conditions in the upper part is given by Åmand and Leckner (1992): First, the N_2O emission decreased when the secondary air was moved upwards, second, the N_2O emission was markedly decreased when half of the secondary air was introduced at 5.5 m and the other half was added at the cyclone outlet, giving very low oxygen concentrations throughout the furnace. These results were, however, obtained with a sand bed and it is not known if they would remain the same in presence of a sorbent for sulphur capture. Another indication of the effect of the conditions in the upper part is the total air ratio. Assuming that the effect of total air ratio is important, this should be attributed to the conditions in the upper part of the combustion chamber, because of the moderate effect on N_2O of the conditions in the bottom part. The literature data regarding the effect of total air ratio are, however, uncertain due to the difficulties in keeping the top temperature constant. Åmand and Leckner (1992) showed a significant effect of air ratio on N_2O at constant top temperature, though again in the absence of sorbent.

For NO the situation is different, and the NO concentration decreases with height in the combustion chamber. The impact of staging is significant, *cf.* Fig. 1, indicating the importance of the bottom zone. Changes in total air ratio have a significant effect on NO, but to what extent this should be attributed to changes in the bottom zone or higher zones is not manifest, in view of the marked influence of the bottom zone. NO was not significantly affected by moving the position of secondary air addition upwards (Åmand and Leckner, 1992).

Summary

The effect of reducing conditions in the bottom zone is important for the emission of NO and SO_2, while small or moderate for N_2O. Available literature data suggest that the effect of changes in the upper part could be important for N_2O, while the situation is less clear for SO_2 and NO.

While the sulphur capture performance is affected by the fraction of time under reducing conditions, the emissions of NO and N_2O may be influenced also by the oxygen concentration as such.
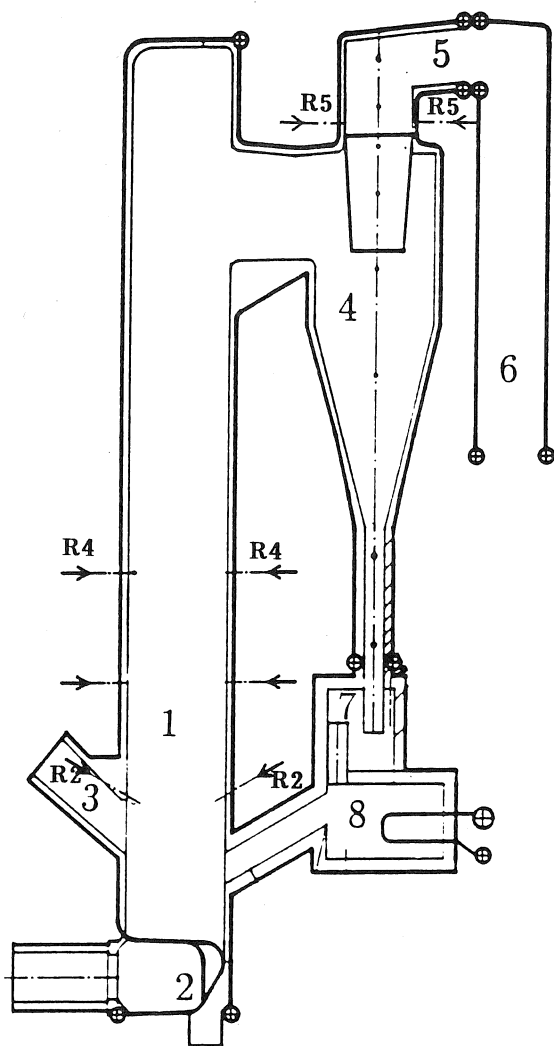
FIG. 2. THE CHALMERS 12 MW BOILER.
1, combustion chamber; 2, air plenum and start-up
combustion chamber; 3, fuel feed chute; 4, cyclone;
5, exit duct; 6, convection cooling section; 7, particle seal;
8, particle cooler; →, secondary air nozzle inlets,
R2 at 2.2 m, R4 at 5.5 m and R5 in cyclone outlet.

TABLE 1. FUEL AND LIMESTONE MASS FRACTIONS
(daf=dry, ash-free).

Fuel	Bituminous coal
size	<20 mm (50% <10 mm)
moisture	0.16
ash	0.08
volatiles, daf	0.40
carbon, daf	0.78
hydrogen, daf	0.055
nitrogen, daf	0.016
oxygen, daf	0.13 calculated
sulphur, daf	0.014
Sorbent	Ignaberga limestone
size	0.2-2 mm
$CaCO_3$	90%

TABLE 2. GAS ANALYSERS USED.

Gas	Range, ppm / %	Name, Type
SO_2 (b)	0-3000	Uras 3G, i.r.
SO_2 (a)	0-3000	Binos, vis./u.v.
CO	0-1000	Uras 3G, i.r.
NO(a)	0-250	Beckman 955, chemiluminescence
NO(b)	0-250	Beckman 955, chemiluminescence
N_2O	0-500	Spectran 647, non-dispersive i.r.
O_2(a)	0-10%	Magnos 7G, paramagnetic
O_2(b)	0-10%	Magnos 5T, paramagnetic
O_2-boiler	0-10%	Westinghouse 132/218, zircon. cell

Fuel is fed to the bottom zone of the combustion chamber through a fuel chute (3). Primary air is introduced through nozzles in the bottom plate (2) and secondary air can be injected through several nozzle registers located horizontally on both sides of the combustion chamber, as indicated by the arrows in Fig. 2. Entrained bed material is captured in the hot, refractory-lined cyclone (4) and returned to the combustion chamber through the return leg and particle seal (7). Combustion air can also be added in the cyclone outlet. After the cyclone the flue gas passes an uncooled duct (5).

Fig. 2 does not show the flue-gas recycling system, which supplies flue gas to the combustion chamber for fine tuning of the temperature in the boiler. Large, intentional changes in temperature can be made using the external, adjustable particle cooler (8).

EXPERIMENTAL CONDITIONS

The boiler

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 equipped for special measurements and has facilities that make it possible to vary parameters independently and in a wider range than in a commercial boiler. The boiler can also be operated under extreme conditions inappropriate to commercial boilers.

The boiler is shown in Fig. 2. The height of the combustion chamber is 13.5 m and the square cross-section is about 2.5 m².

The fuel and the limestone

The sorbent was Ignaberga limestone and the coal was a bituminous coal with a medium sulphur content. Limestone and fuel data are shown in Table 1.

Measurements

Regularly calibrated on-line gas analysers, see Table 2, were used for continuous monitoring of O_2, CO, SO_2, NO and N_2O in cold and dry gases. Except for the analyser called O_2-boiler, which was sampling in the convection pass, all the analysers were connected to the gas duct after the baghouse filter of the boiler.

TABLE 3. AVERAGE VALUES. Columns show:

Tbd	temperature in bed, °C	
Ttop	temperature in top of comb. chamber	
O2,w	%O_2-boiler (wet)	
O2a	%O_2 (dry) analyser a	
O2b	%O_2 (dry) analyser b	
SOa	ppm SO_2 normalised to 6% O_2	
SOb	ppm SO_2 normalised to 6% O_2	

NOa	ppm NO normalised to 6% O_2
NOb	ppm NO normalised to 6% O_2
N2O	ppm N_2O normalised to 6% O_2
CO	ppm CO normalised to 6% O_2
dPtt	total press. drop in comb. chamb., kPa
Airt	total air flow, kg/s
Prim	primary air flow, kg/s

Sec	total secondary air flow, kg/s
Rg4	secondary air flow at 5.5 m, kg/s
Rg5	air flow to cyclone outlet, kg/s
FGr	recirculated flue gas, kg/s
Tex	temperature after cyclone, °C
λc	combustor air ratio, Eq. (6)

	Tbd	Ttop	O2,w	O2a	O2b	SOa	SOb	NOa	NOb	N2O	CO	ΔPtt	Airt	Pri	Sec	Rg4	Rg5	FGr	Tex	λc
No stag.	851	859	3.46	3.97	3.85	68	66	139	138	125	30	6.0	3.54	3.01	0.00	0.00	0.00	0.21	822	1.212
Normal	851	859	3.47	3.99	3.83	123	133	80	85	97	42	6.1	3.54	1.74	1.25	0.00	0.00	0.98	832	1.213
Intens. low	852	868	3.46	3.89	3.77	317	301	71	71	94	58	6.0	3.54	1.27	1.67	1.65	0.00	1.00	853	1.212
Intens. cycl	852	860	3.46	3.77	3.64	385	370	45	46	18	142	6.9	3.54	1.61	1.36	0.68	0.69	0.87	779	1.010
Reversed	850	855	3.48	4.27	*	124	*	32	*	30	329	6.0	3.54	2.31	0.82	0.00	0.74	1.04	743	1.007
Rev. HB	851	855	3.47	4.13	3.78	153	129	35	40	23	410	8.6	3.54	2.37	0.76	0.00	0.70	0.73	741	1.003
Rev. FA	851	857	3.48	4.04	*	74	*	41	*	25	440	6.0	3.55	2.36	0.76	0.00	0.69	0.42	746	0.990
Rev EL	850	855	3.44	3.92	3.66	99	103	38	36	22	153	6.0	3.55	2.35	0.78	0.00	0.71	1.25	759	1.020

* b-analysers measuring in-situ

The emissions of SO_2, NO, N_2O and CO have been normalized to a flue gas with an oxygen concentration of 6%.

Before the gas was introduced to the N_2O analyser, the SO_2 in the flue gas was captured in a solution of carbonate, since the N_2O analyser is affected by high SO_2 concentrations.

Combustor air ratio

The *total* air ratio, λ_{tot}, is defined

$$\lambda_{tot} = 1 + K_c \frac{O_2}{21 - O_2} \qquad (3)$$

where O_2 is per cent oxygen in the flue gas (including moisture) as measured in the convection pass, and the correction factor K_c is the ratio of theoretical flue gas (including moisture) to theoretical air (*i.e.* moles of flue gas over moles of air at stoichiometric conditions). K_c is 1.07 for the fuel used.

By *combustor* air ratio is here meant the air ratio corresponding to the conditions in the flue gas exiting the cyclone, *i.e.* before the addition of final combustion air in the reversed staging case.

If flue gas is not recycled, the combustor air ratio may be calculated as

$$\lambda_{c,no\ recycle} = \lambda_{tot}(1 - x) \qquad (4)$$

where x is the fraction of total air which is introduced in the cyclone outlet.

Due to flue gas recirculation this definition of combustor air ratio is not suitable since it underestimates the air ratio in the boiler. A better definition is derived from an oxygen mass balance over the two flows that are mixed in the cyclone outlet: combustion air and flue gas from the cyclone. This yields an expression for oxygen concentration in the cyclone outlet and prior to air addition

$$O_{2,c} = \frac{O_2(1 + y) - 21x}{1 - x + y} \qquad (5)$$

where y is the ratio of flue gas recirculation to total air flow. From this expression the actual combustor air ratio is obtained

$$\lambda_c = 1 + K_c \frac{O_{2,c}}{21 - O_{2,c}} \qquad (6)$$

Cases studied

All test cases were run at constant load, *i.e.* the combustion air added was held constant at 3.54 kg/s and the total air ratio was kept constant at 1.2 (3.5% O_2,w) *cf.* Table 3. The bed temperature was 850°C, the total pressure drop 6 kPa and the limestone addition was constant at 165 kg/h corresponding to a molar Ca/S ratio of about 2.5.

In addition to the reference case and the reversed staging case, results from three other cases are included, giving a total of five cases of staging:

Reference case (normal staging): about 60% primary air and 40% secondary air (2.2 m above air distributor).

No staging: no secondary air, all air added in the bottom part. This means significantly more oxidising conditions in the bottom part compared to the reference case.

Intensified staging (low): about 40% primary air and 60% secondary air added at a higher level (5.5 m above air distributor). This means substantially more reducing conditions in the lower part.

Intensified staging (cyclone): about 60% primary air, 20% secondary air (at 5.5 m) and 20% air for final combustion in cyclone outlet. This means significantly more reducing conditions in the top of the boiler, and a prolonged primary zone compared to the reference case.

TABLE 4. TEST CASES

Per cent of total air in bottom, at 2.2 m and 5.5 m height and in cyclone outlet. (The sum is not 100% since some air is introduced via the particle cooler, the classifiers and the fuel chute.)

	Bott	2.2 m	5.5 m	Cyc. o.	Comment
Reference	49	35	-	-	normal staging
No staging	85	-	-	-	no secondary air
Intens. low.	36	-	47	-	more red. lower part
Intens. cycl.	45	-	19	19	more red. all over
Reversed	65	-	-	21	rev. cond. top & bot.
RevHB:	67	-	-	20	Rev. high bed
RevFA:	66	-	-	19	Rev. fly-ash
RevEL	66	-	-	20	Rev. extra lime

Reversed staging: no secondary air in the combustion chamber, but about 20% of the total air added after the cyclone for final combustion. The combustor air ratio, before addition of final combustion air, was kept at about unity. This means less oxidizing conditions in the upper part and more oxidizing conditions in the bottom part compared to the reference case.

In addition to these cases, three variations of the case of reversed staging are included.

RevHB: During this test period the discharge of bed-ash was not used, resulting in a higher pressure drop in the combustion chamber.

RevFA: Fly-ash (from a secondary cyclone) was recirculated to the combustion chamber.

RevEL: 25% extra limestone addition, and, most important, the combustor air ratio was optimized to give minimum emissions.

RESULTS

An overview of the test cases is given in Table 4. The emissions of SO_2, NO, N_2O and CO are shown in Figs. 3 to 6, and mean values are also shown in Table 3. The results compared to the reference case (normal staging) can be summarized as follows:

No staging: less reducing conditions in the bottom part result in a more efficient sulphur capture, but a significantly higher NO emission and a somewhat higher N_2O emission.

Intensified staging (low): more reducing conditions in the bottom part dramatically lower the sulphur capture efficiency, while the NO emissions are significantly decreased and the N_2O emissions are somewhat decreased.

Intensified staging (cyclone): more reducing conditions in the boiler as a whole have a similar but more pronounced effect as intensified bottom staging. The N_2O emission is considerably lower, however.

Reversed staging: here the N_2O emission is reduced by about three fourths, while the NO emission is halved and the SO_2 emission is not significantly affected. The higher CO emission in this case is treated below.

The variations of the reversed staging case, *RevHB*, *RevFa* and *RevEL* do not differ greatly from the Reversed staging case. The sulphur capture performance is, as expected, somewhat improved by the recirculation of fly-ash and by the addition of

extra limestone. An important difference between the cases is the small variation in combustor air ratio, (Eq. 6) which affects all the emissions, especially CO, see below.

Combustor air ratio

The reversed staging case was further explored by a variation in the fraction of air added in the cyclone outlet, see Fig. 7 and Table 5. This variation was made with 25% more limestone addition than in the other test cases. The total air ratio was kept constant, while the fraction of air added to the cyclone outlet was varied. The conditions are best characterized by the combustor air ratio, λ_c, given by Eq. (6), which includes the effect of flue gas recirculation. As can be seen, there is an optimum point at $\lambda_c \approx 1.02$. Below this point CO increases dramatically, while SO_2 increases slowly, N_2O does not decrease anymore and NO is close to its minimum. (Surprisingly enough, NO seems to go through a minimum point.)

It follows from these results that the high CO emissions in the cases Reversed, RevHB and RevFA can be explained by the combustor air ratio, which was 1 to 3% lower than the optimum point in these cases, cf. Table 3.

The value of $O_{2,c}$ in the optimum point is about 0.4%, corresponding to a λ_c of 1.02, which makes the optimum point slightly over-stoichiometric. This is, however, within the error margin, if the possible errors in the O_2 measurement and in x are taken into consideration, and λ_c in the optimum point can be said to be about unity.

Reproducibility

The reference case was run under stable conditions for about 5×24 h, the reversed staging case, including variations, for 3×24 h, and the other cases for at least $1\frac{1}{2}\times24$ h. Representative periods for the calculation of average values were, if possible, chosen when the so called b-analysers (Table 2) were not occupied by in-situ measurements. The averaging periods were 4 to 6 h, but for the case RevFA it was $2\frac{1}{2}$ h long and for RevEL and the data shown in Fig. 7 and Table 5 the periods were about one hour.

The reproducibility of the NO, N_2O and CO emissions was very high. The reproducibility of SO_2 was somewhat lower, probably as a result of variations in the fuel sulphur content. Even a variation in the sulphur capture performance of a few per cent affects the SO_2 emission significantly when the sulphur capture performance is as high as 90%.

From Table 3 it is seen that bed temperature, top temperature, air ratio (O_2), load (shown by total air and air ratio) and total pressure drop are the same in all cases. The test periods chosen were all run at stable operating conditions, with typical standard deviations of <0.1% for O_2 and 1 to 2°C for bed and top temperature.

DISCUSSION

The results clearly show that it is possible to separate the effects of reducing/oxidizing conditions on the emissions by producing these conditions selectively in the bottom and top parts of the combustor. A major decrease of the N_2O and NO emissions took place without increasing SO_2.
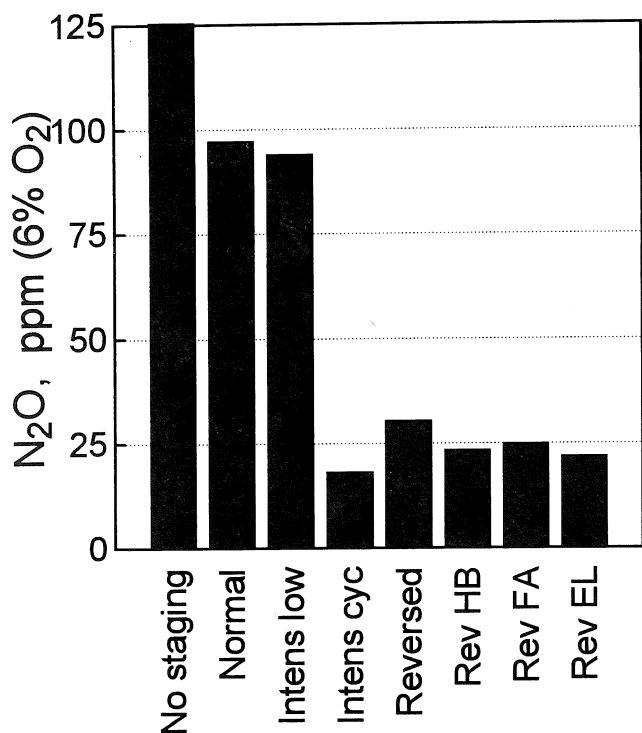
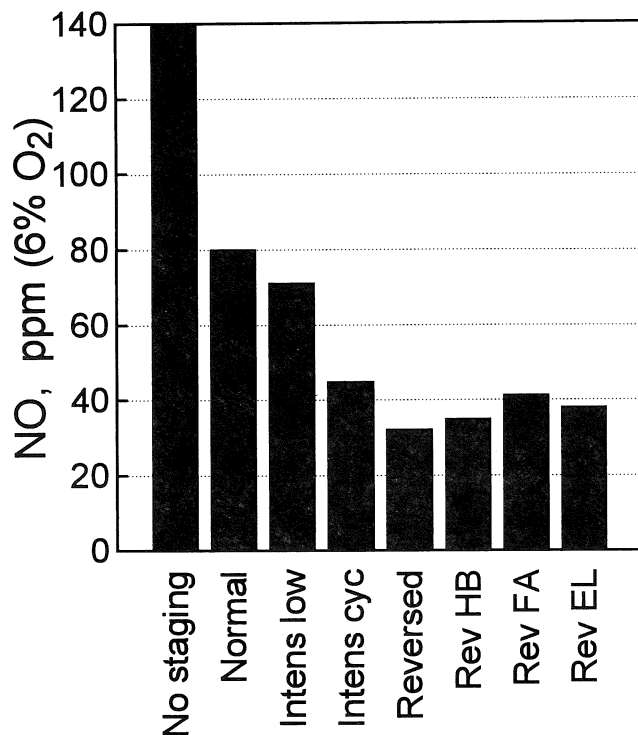FIG. 3. N$_2$O EMISSION IN THE CASES INVESTIGATED.

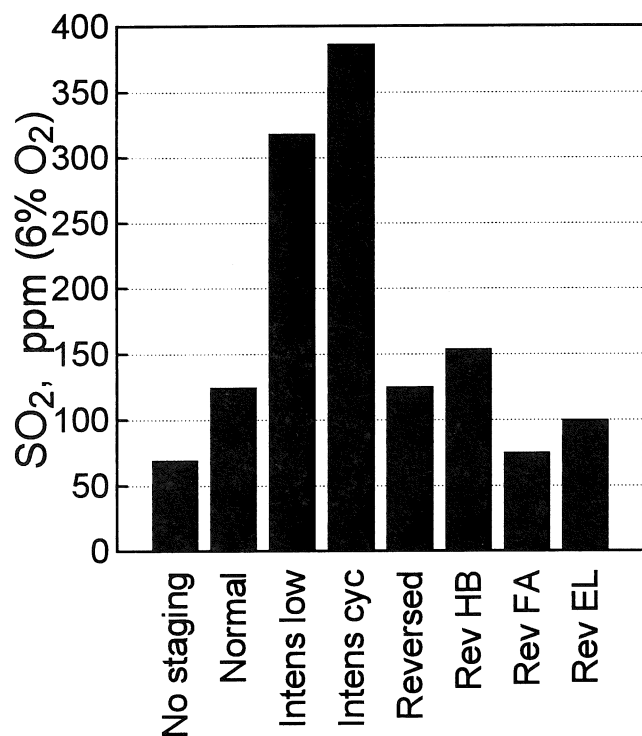
FIG. 5. NO EMISSION IN THE CASES INVESTIGATED.

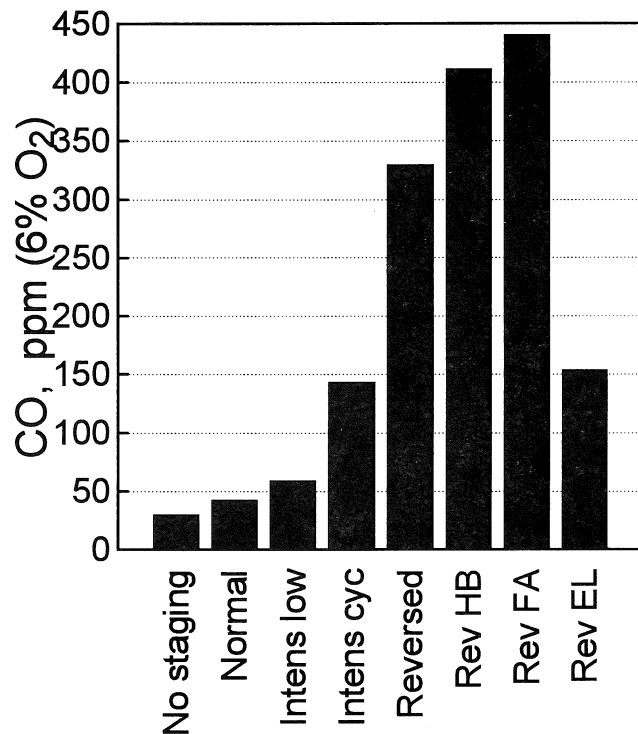FIG. 4. SO$_2$ EMISSION IN THE CASES INVESTIGATED.

FIG. 6. CO EMISSION IN THE CASES INVESTIGATED.

TABLE 5. VARIATION OF COMBUSTOR AIR RATIO FOR REVERSED STAGING CASE. *Cf.* Table 3.

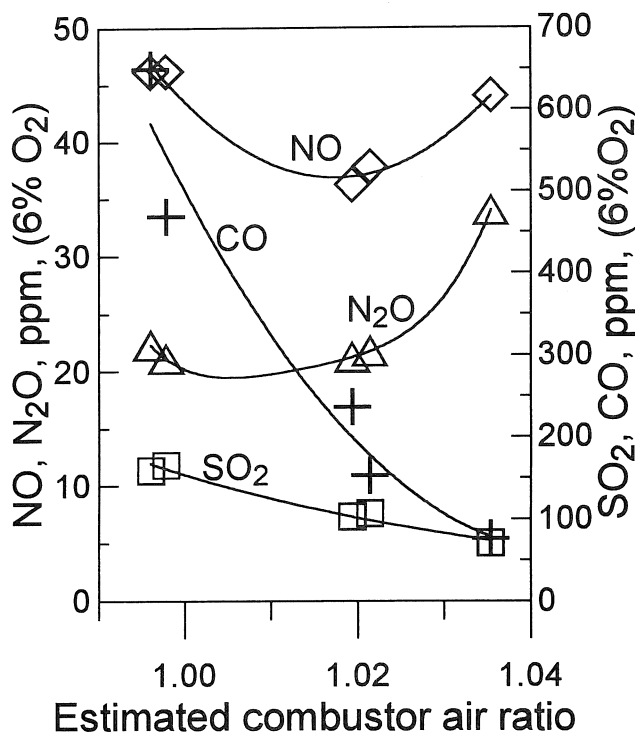| Tbd	Ttop	O2,w	O2a	SOa	SOb	NOa	NOb	N2O	CO	dPtt	Airt	Rg5	FGr	Tex	λc
849	853	3.49	4.04	70	66	44	43	34	76	6.0	3.54	0.659	1.21	762	1.035
851	856	3.38	3.92	106	103	38	36	22	151	5.9	3.55	0.689	1.23	764	1.021
849	853	3.51	4.05	103	101	36	35	21	238	6.0	3.55	0.731	1.26	754	1.019
851	854	3.42	3.95	165	174	47	47	21	470	5.9	3.54	0.787	1.24	738	0.998
848	851	3.56	4.07	159	171	46	50	22	651	6.1	3.55	0.823	1.22	726	0.996

FIG. 7. EMISSIONS VERSUS COMBUSTOR AIR RATIO,
EQ. (6), FOR THE REVERSED STAGING CASE.
THE TOTAL AIR RATIO WAS 1.2 AND THE LIMESTONE
ADDITION WAS 25% HIGHER THAN IN NORMAL
CASES.

the lower part. This takes place despite the marked effect on NO of changes in the bottom zone, and reveals that NO reduction in the upper part is significantly enhanced by the less oxidizing conditions.

Like NO, the sulphur capture performance is very sensitive to changes in the degree of staging. Less oxidizing conditions in the upper part of the boiler result in a dramatic decrease in sulphur capture performance, *cf.* the case of Intensified staging (cyclone), if not compensated by more oxidizing conditions in the bottom as in the Reversed staging case. A good sulphur capture performance is maintained when shifting from normal to reversed staging and this indicates the importance of the bottom zone for the sulphur capture process. Two explanations contribute to the importance of the bottom bed: *i)* the high concentration of sorbent in the bottom part and *ii)* that the major fraction of the sulphur is normally released from the fuel in the bottom zone.

Possible disadvantages with reversed staging.

An undesirable increase in the emission of CO was seen. This CO increase was significantly reduced by adjusting the fraction of air added in the cyclone exit, Fig. 7. Other improvements could be obtained by:

- **Preheated air to cyclone exit**. The exit duct temperature drops considerably when (cold) air is added to the cyclone exit, see Table 3. This is believed to contribute to the higher CO emission in the Reversed staging case. It is likely that CO can be significantly reduced without affecting the other emissions by using preheated air.
- **Improved air distribution**. In-situ measurements have shown that the oxygen concentration varies significantly in the horizontal plane (also in absence of secondary air). A better distribution of air in the bottom part would refine the conditions and probably further improve the results.

The loss of char (=fly-ash flow×fraction of char) increased with about 25% compared to the reference case resulting in a decrease in combustion efficiency of about 2%. This decrease would probably be less in a larger (higher) boiler with a more efficient cyclone. The combustion loss can be reduced by recirculation of fly-ash from a secondary (cold) cyclone. A combustor air ratio corresponding to the optimum point is expected to reduce the loss of combustibles, but this case was not run for a long enough period to verify the combustion efficiency under these conditions.

It is not known whether the lower oxygen concentration in the upper part could have effects on the tube wall material.

The increased air flow to the bottom zone results in a higher power consumption.

The dramatic decrease in the N_2O emissions with reversed staging, indicates the important role of the reactions in the upper part of the combustion chamber. The high reduction rate of the bottom zone preventing the major fraction of the N_2O formed in the lower part of the combustion chamber from passing through the boiler emphasizes the contribution from the upper part. This interpretation is consistent with the moderate effect of changes of the conditions in the bottom zone (cf. cases No Staging, Normal and Intensified low). It is not known to which extent the low N_2O emission at reversed staging should be attributed to a reduced N_2O formation or to an increased reduction of N_2O.

Also for NO the influence of less oxidizing conditions in the upper part overrides the effect of more oxidizing conditions in

CONCLUSIONS

By reversed staging is meant that the oxygen is decreased in the upper part and increased in the lower part. With reversed staging it was possible to reduce the N_2O emission to one fourth (25 ppm) and the NO emission to half (40 ppm) without affecting the sulphur capture performance (90%). The combustion efficiency was somewhat decreased. It is not known if the lower oxygen concentration in the upper part of the boiler could have a negative effect on the tube wall material.

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