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Citation for the published paper:

Åmand, L. ; Leckner, B. ; Dam-Johansen, K. (1993) "Influence of SO₂ on the NO/ N₂O chemistry in fluidized bed combustion 1. Full-scale experiments". Fuel, vol. 72(4), pp. 557-564.

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Influence of SO₂ on the NO/N₂O chemistry in fluidized bed combustion

1. Full-scale experiments

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(Received 10 February 1992; revised 21 July 1992)

The lime used for sulfur capture in circulating fluidized bed (CFB) boilers interferes with the NO/N₂O chemistry. This has been explained as an effect of the surface of the lime. However, homogeneous reactions involving SO₂ could also be important. Large-scale experiments were carried out in a CFB boiler with the aim of separating the effect of lime from that of SO₂. This was achieved by comparing a test involving removal of SO₂ by lime addition, while burning a high sulfur coal, with a test where SO₂ was added to the combustion air for a low sulfur coal in a quantity corresponding to the emission of SO₂ from the high sulfur coal. The tests show that the SO₂ itself influences the NO/N₂O chemistry independent of the catalytic effect of the lime surface. Carbon monoxide is believed to be an important intermediary species in the reactions, but laboratory investigations are needed to interpret the results further. The connection between NO reduction and N₂O formation is further studied by supply of NO to the primary air duct.

(Keywords: fluidized beds; combustion; sulfur)

Fluidized bed combustion is a convenient way of burning fuels of different ranks, ranging from wood to anthracite. Due to the low combustion temperature (800–900°C), the emission of nitrogen oxides (NO_x) is low compared to other combustion technologies such as pulverized coal combustion. By adding limestone to the bed, sulfur can also be captured, and very low emissions of SO₂ can be achieved. However, addition of lime to the fluidized bed causes increased emission of NO. This has been shown by tests in large-scale circulating fluidized bed (CFB) boilers^{1–5}, in pilot-scale CFB boilers⁶ and in laboratory test units^{7–10}. The increase of NO emission due to lime addition is critical, since a CFB boiler may then need additional NO reduction equipment, such as ammonia addition to the hot cyclone (selective non-catalytic reduction process), in order to fulfil stringent NO regulations at the same time as a proper sulfur reduction performance is achieved.

The increase of NO has been explained as a catalytic effect of the lime on the oxidation of ammonia species to NO. Experimental evidence of this reaction in the laboratory is available^{11–16}, but nevertheless, the proposed reaction may only partially account for the increase of the NO during lime addition. To demonstrate this, a test programme was carried out in the 12 MW CFB research boiler at Chalmers University of Technology. The programme focused both on the homogeneous effect of SO₂ on the NO/N₂O chemistry in the combustion chamber, and on the effect of free CaO surfaces on the oxidation of CO. Carbon monoxide is

an important NO-reducing species, which together with the char surface is responsible for the low emission of NO from CFB boilers^{17–19}. Nitrous oxide was also studied in the test programme, and results regarding this species are discussed in the light of previous publications^{17,20,21}.

The full-scale experiments have the advantage of providing a realistic environment for the features tested but, with a few exceptions, only measurements in the flows before and after the boiler were made. The detailed processes in the combustor or close to particles were not assessed. In order to support the boiler measurements, particular features were investigated in laboratory installations; results of these tests are presented in part 2²².

EXPERIMENTAL AND RESULTS

Apparatus

The 12 MW CFB research plant is a commercial boiler with the combustion chamber made up of membrane tube walls (*Figure 1*). The combustion chamber is 13.5 m in height and the square cross-section is about 2.9 m². Fuel was fed to the bottom of the combustion chamber through a fuel chute (1). Primary air was introduced through nozzles in the bottom plate (2) and secondary air could be injected through several registers located along the combustion chamber, as indicated by the arrows in *Figure 1*. In the present tests only the lowest secondary air level (3) was used, located 2.2 m above the

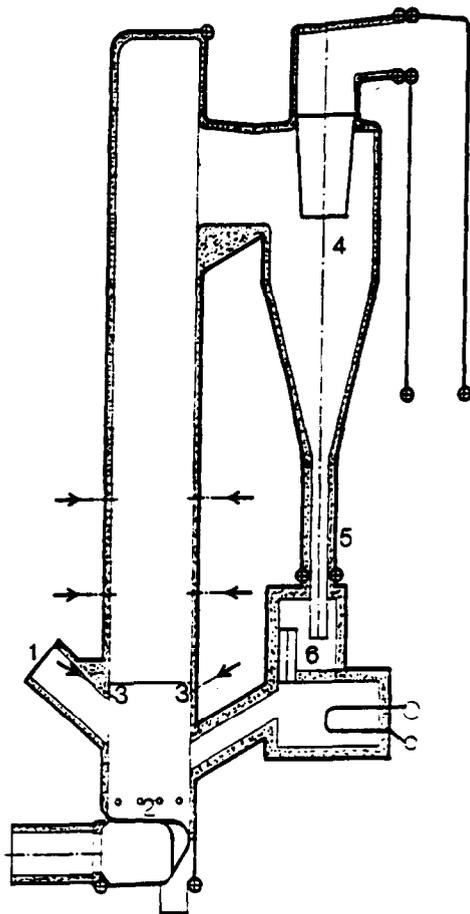


Figure 1 The 12 MW CFB boiler: (1) fuel feed chute; (2) bottom plate; (3) secondary air inlet at 2.2 m; (4) cyclone; (5) particle return leg; (6) particle seal

bottom plate. The lower part of the combustion chamber was refractory lined. In order to operate the boiler at the desired temperatures (800–900°C), two of the four sides of the combustion chamber were also refractory lined. Entrained bed material was captured in the refractory-lined cyclone (4) and returned to the combustion chamber through the return leg (5) and particle seal (6). The equipment for secondary fly-ash recirculation (not shown in Figure 1) consisted of a secondary cyclone located before the bag-house filter and a special fly-ash bin which maintained a stable recycling flow of fly ash. The secondary fly-ash recirculation system was used in all tests of the present programme. The flue-gas recirculation system (not shown in Figure 1) supplied flue gas to the bottom of the combustion chamber through the bottom plate in order to control the temperature of the boiler.

The research plant was equipped with on-line gas analysers for continuous monitoring of O₂, CO, SO₂, NO and N₂O on cold and dry gases. The N₂O instrument (an infra-red analyser, Perkin Elmer, Spectran 647) was cross-sensitive towards water, SO₂ and CH₄. Water was removed in a refrigerator located ahead of the gas analysers; SO₂ was only important at concentrations higher than 500 ppm, and then the N₂O signal was compensated as described by Hulgaard *et al.*²³. Methane (CH₄) was not present in the exit flue gas at normal operating conditions. As a check, some samples of dry flue gas were collected in Tedlar bags for subsequent analyses for N₂O and CH₄ on two different gas

chromatographs. No CH₄ was found and there was no difference in the N₂O level between the gas chromatograph and the corrected signal from the N₂O analyser. The CO analyser (an infra-red analyser, Hartmann & Brown, Uras 3K), was cross-sensitive towards N₂O; this was compensated by a small correction of the CO signal.

A data acquisition system was used for collection of data from the boiler.

Materials

Two high-volatile bituminous coals from Poland were used. The fuel characteristics are listed in Table 1. The fuels have similar properties apart from the sulfur content, which is the parameter of interest in this study. Furthermore, the high-sulfur coal contains a larger fraction of fines than the low-sulfur coal.

A porous limestone (Ignaberga) was used, as in Refs 1 and 3.

General methods

A reference case was defined, corresponding to a bed temperature at the bottom of the combustion chamber of 850°C, a total excess air ratio of 1.2, and a primary air stoichiometry of 0.75. In the reference case, the temperatures at the top of the combustion chamber and at the outlet of the hot cyclone were 860°C and 810°C, respectively. The boiler load was kept constant during the tests, with a fluidizing velocity of about 6 m s⁻¹ at the top of the combustion chamber. The recycling of fly ash from the secondary cyclone was held at a ratio of recycled flow of fly ash to fuel feed rate of 0.5. The operating conditions of the tests are given in Table 2.

Table 1 Characteristics of the fuels

Type	High-sulfur coal	Low-sulfur coal
Mean size (mm)	2.5	14.4
<1 mm (%)	40.0	4.5
Volatiles (% daf)	39.5	35.7
Proximate analysis (% as delivered)		
Combustibles	82.6	85.5
Ash	7.6	7.1
Moisture	9.8	7.4
Ultimate analysis (% daf)		
C	84.7	83.8
H	5.9	4.9
O	5.1	9.1
S	2.7	0.8
N	1.6	1.4
Lower heating value (MJ kg ⁻¹ daf)	33.9	31.6
Ash analysis (% on dry ash)		
CaO	6.3	7.5
MgO	1.3	4.2
Fe ₂ O ₃	18.1	9.0
Na ₂ O	1.1	1.3
K ₂ O	1.6	2.2
Al ₂ O ₃	22.9	23.0
SiO ₂	43.3	44.9
P ₂ O ₅	0.5	0.8
TiO ₂	0.9	0.9
BaO	0.1	0.2
SO ₃	5.0	6.4

Table 2 Operating conditions

Parameter	Excess lime	SO ₂ -sand bed	SO ₂ -lime bed	NO-lime bed	High-low S coal
Fuel sulfur content	High	Low	High	Low	Low, high
Bed material	Sand, lime	Sand	Lime	Lime	Sand, lime
Load (MW)	8.9 ± 0.2	9.0 ± 0.2	8.9 ± 0.1	8.9 ± 0.2	9.0 ± 0.1
Temperature (°C)					
Bottom of bed	848 ± 2	850 ± 2	847 ± 2	854 ± 5	850 ± 2
Top of furnace	859 ± 3	863 ± 2	861 ± 3	868 ± 6	863 ± 2
After hot cyclone	805 ± 5	813 ± 1	806 ± 1	800 ± 1	812 ± 4
Excess air ratio	1.21 ± 0.01	1.21 ± 0.01	1.21 ± 0.01	1.21 ± 0.01	1.21 ± 0.01
Primary air stoichiometry	0.74 ± 0.01	0.73 ± 0.01	0.73 ± 0.01	0.73 ± 0.01	0.74 ± 0.01
Recirculation flow of fly ash/fuel flow	0.67	0.54	0.54	0.54	0.54
Differential pressure, reactor (kPa)	7.1 ± 1.0	5.5 ± 0.1	5.0 ± 0.1	4.3 ± 0.4	5.4 ± 0.2
Differential pressure, <2 m (kPa)	3.3 ± 0.7	2.7 ± 0.1	2.2 ± 0.1	1.7 ± 0.2	2.4 ± 0.3
Fluidization velocity, top (m s ⁻¹)	6.3 ± 0.1	6.3 ± 0.2	6.1 ± 0.1	6.3 ± 0.1	6.5 ± 0.3

When the boiler was run at steady state conditions with the high-sulfur coal and a new bed of pure silica sand, supply of excess lime was started, corresponding to a molar ratio Ca/S of 4.3. The transient was then followed for 24 h, during which the lime flow was lowered in two steps and the changes in emissions of SO₂, NO, N₂O and CO were followed.

In order to study the influence of SO₂ independent of the lime, SO₂ was introduced with the primary air during operation with the low-sulfur coal and a bed consisting of ash and silica sand. The SO₂ concentration was raised to the same level as that of the high-sulfur coal without lime supply. For comparison, this test was repeated for the high-sulfur coal with a Ca/S molar ratio of 1.6.

Finally, to understand more about the N₂O formation, a step response test with NO addition to the primary air was also carried out, using the low-sulfur coal.

Step response test with excess lime

Figures 2a and b show the transient response of the concentrations of SO₂, NO, N₂O and CO when the supply of excess lime was started. The boiler was operated with the low-sulfur coal overnight, then with high-sulfur coal. After operation with the high-sulfur coal under constant conditions for 2 h, the supply of excess lime was started with a feed rate corresponding to a molar ratio of Ca/S of 4.3. Due to the high feed rate of lime, a large decrease in the SO₂ concentration in the stack was recorded and a sulfur retention of 98% was achieved 3.5 h from the start. At the same time a rapid increase of NO from 45 to 70 ppm could be seen, followed by a gradual increase up to 281 ppm. At time, $t = 6.15$ h the lime flow was lowered to a Ca/S molar ratio of 2.1. This had only a minor effect on the SO₂ concentration during the 2.5 h of operation before the lime flow was lowered again. On the other hand, the effect on NO was dramatic. After a rapid drop in NO from 280 to 175 ppm, a shift in the rate of decline could be seen (Figure 2a) at $t = 7.0$ h. From this point, the NO gradually decreased to 120 ppm. At $t = 8.5$ h the lime flow was lowered again to a Ca/S molar ratio of 1.15. This time, a clear increase of the SO₂ concentration could be seen. For NO there was a decrease similar to that shown before, first a rapid decrease followed by a shift in the rate of decline at $t = 9.3$ h. NO and N₂O behaved in opposite ways during the entire test: if NO increased then N₂O decreased and vice versa. Also, the shift in the rate of increase of NO at $t = 1.76$ h and in the rate of decline at $t = 7.0$ and 9.3 h were also seen for N₂O.

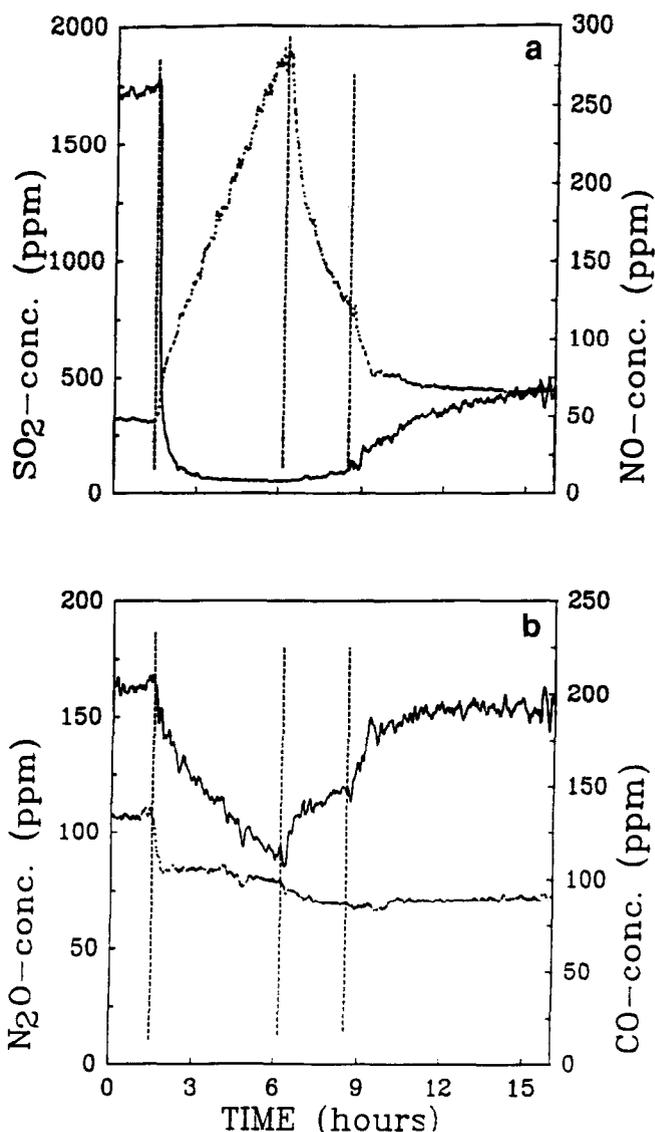


Figure 2 Step response test with excess supply of lime: (a) SO₂ (—) and NO (···) and (b) N₂O (—) and CO (···). See Table 2 for operation conditions. The Ca/S ratios were:

Ca/S	t (h)
0	0–1.41
4.3	1.41–6.15
2.1	6.15–8.5
1.15	8.5–24

The CO emission decreased when the lime supply started and continued to decrease even after the first lowering of the lime flow; it did not increase again until the second lowering of the lime flow.

After $t = 16$ h the lime flow was kept constant for 8 h, during which only a small change of the SO₂ and NO concentrations was recorded: the SO₂ increased from 440 to 510 ppm, while the decrease of NO was hardly noticeable.

Step response test with addition of SO₂ to a sand bed

Figures 3a and b show the result when SO₂ was supplied to the primary air. From $t = 0-20$ min and from $t = 55-80$ min the gas concentrations were kept at the reference level. At $t = 20$ min concentrated SO₂ equal to about 1400 ± 100 ppm in the total air was injected into the primary air duct. This supply of SO₂ caused the exit concentration of SO₂ to increase from 500 to 1880 ppm. At the same time the NO concentration decreased from 65 to only 50 ppm and, like the step response test with

the lime, an opposite trend of N₂O could be seen (Figure 3b). The N₂O increased from 130 to 140 ppm. Finally, a clear difference in the CO level could be seen, increasing from 155 to 205 ppm.

Step response test with addition of SO₂ to a lime bed

This test was similar to that on the sand bed, but with some differences. The bed contained lime, which was continuously supplied at a rate corresponding to a molar ratio Ca/S = 1.6. Two different amounts of SO₂ were supplied one after the other, and a cooled suction probe was located at the centre of the combustion chamber, 3.8 m above the bottom plate. The results are shown in Figures 4a to c. The initial concentrations of NO and CO were different from those of the sand bed test, owing to the lime supply. For NO, the reference level was 90 ppm, which decreased to 60 ppm, a decrease twice that in the sand bed. On the other hand, CO only increased from 90 to 110 ppm, which is 30 ppm less than in the sand bed test. Figure 4c shows that changes in the exit value of CO are followed by a corresponding change in the CO concentration at the 3.8 m level in the combustion chamber.

Step response test with addition of NO to a lime bed

In order to learn more about the connection between NO reduction and simultaneous formation of N₂O, a separate test was carried out with NO supply to the primary air duct; the result is shown in Figure 5. The equipment for gas supply was designed for condensed gases such as SO₂ or NH₃ and not for a compressed gas such as NO. This deficiency of the supply equipment led to an uneven flow of NO during the test. However, this had the advantage that the peaks of both the NO concentrations shown in Figure 5 could be followed, since these peaks are related to the uneven flow of NO supplied. The mean supply of NO between $t = 18$ and 22 min corresponded to 410 ppm in the total air. Of this 410 ppm NO, 30% was found at the 2.5 m level, if one takes the staged air supply into account, and 17% was left for the increase of the NO emission from the stack. This means that 83% of the NO supplied had been reduced before the flue gases left the plant. At the same time N₂O increased from 136 to 158 ppm. If two molecules of NO were consumed for every N₂O molecule emitted, 13% of the NO reduced was found as N₂O. It is also interesting to note that every change in the flow of NO added was followed by the N₂O signal.

DISCUSSION

The increase of NO observed during lime addition can be understood as an effect of the change of the SO₂ concentration (the 'homogeneous' effect) and of the catalytic surface of the lime (the 'heterogeneous' effect).

Homogeneous effect of SO₂

The results shown in Figures 3 and 4, where SO₂ was supplied to the primary air duct, prove that the SO₂ concentration in the combustion chamber influences the NO/N₂O chemistry, independent of the lime. This homogeneous effect will be studied by analysing the lime transient of Figure 2. Shortly after the start of the highest lime flow, at $t = 1.76$ h, a shift in the inclination of the

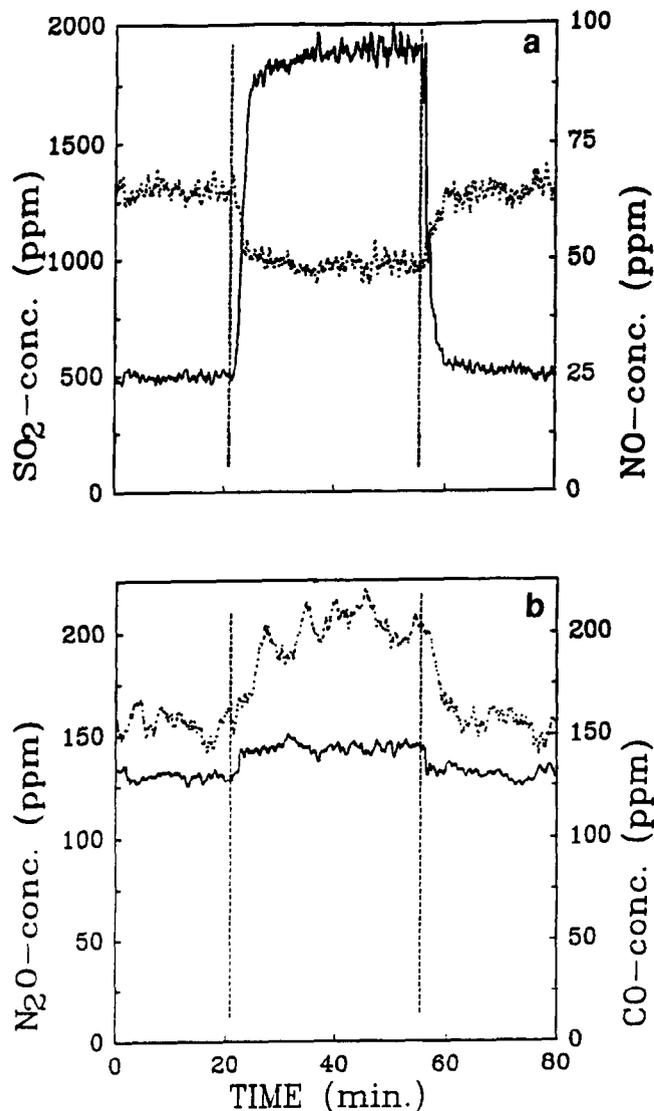


Figure 3 Supply of concentrated SO₂ to the primary air duct of a sand bed: SO₂ supply = 1470 ppm related to total air at $t = 20-55$ min. (a) SO₂ (—) and NO (···); (b) N₂O (—) and CO (···). See Table 2 for operating conditions

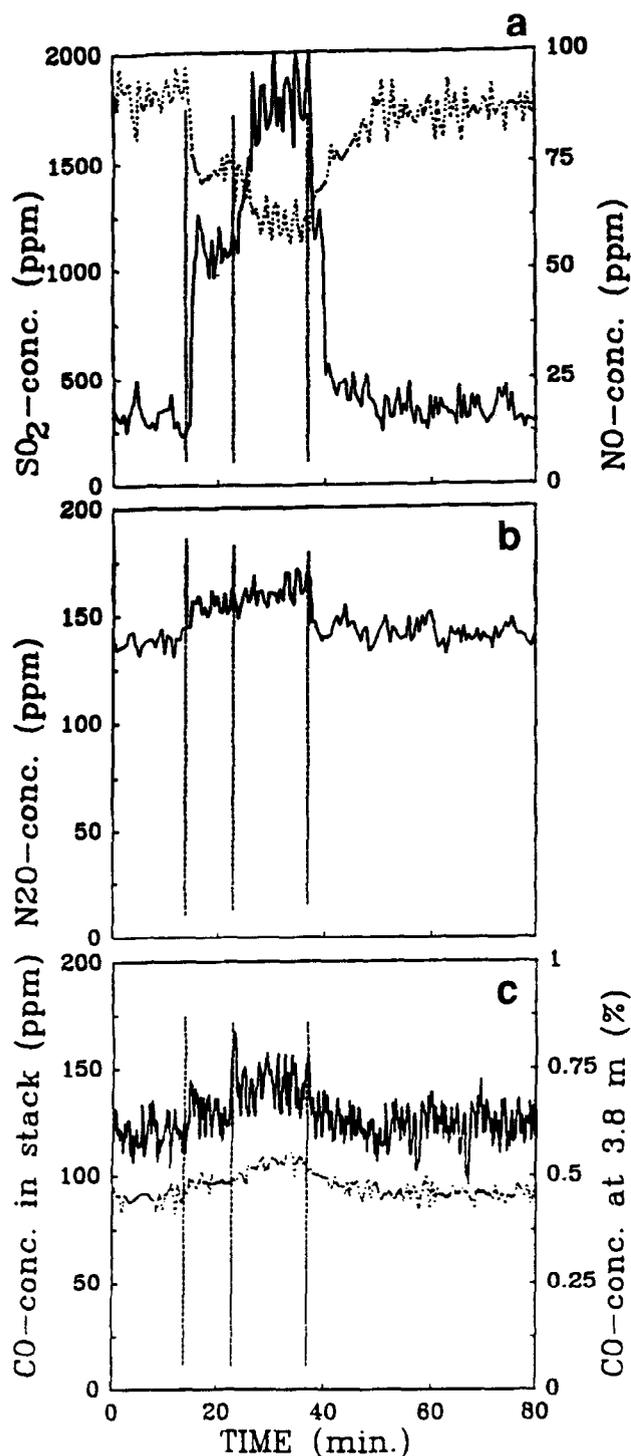


Figure 4 Supply of concentrated SO₂ to the primary air duct of a lime bed: (a) SO₂ (—) and NO (· · ·); (b) N₂O; (c) CO at 3.8 m (—) and CO in stack (· · ·). SO₂ supply = 1300 ppm related to total air at $t = 13-23$ min; 2200 ppm at $t = 23-37$ min. See Table 2 for operating conditions

NO curve is clearly seen. The greatest decrease of SO₂ is observed where the increase of NO is highest. During this part of the lime transient NO is mostly influenced by the SO₂ concentration, since lime has not yet significantly accumulated in the bed, and this is therefore called the homogeneous part of the transient. The influence of SO₂ becomes more evident if the data from Figure 2 are plotted as concentration of NO versus SO₂, as shown in Figure 6 for the part of the lime transient with the highest lime flow ($t < 6.15$ h). In Figure 6 the

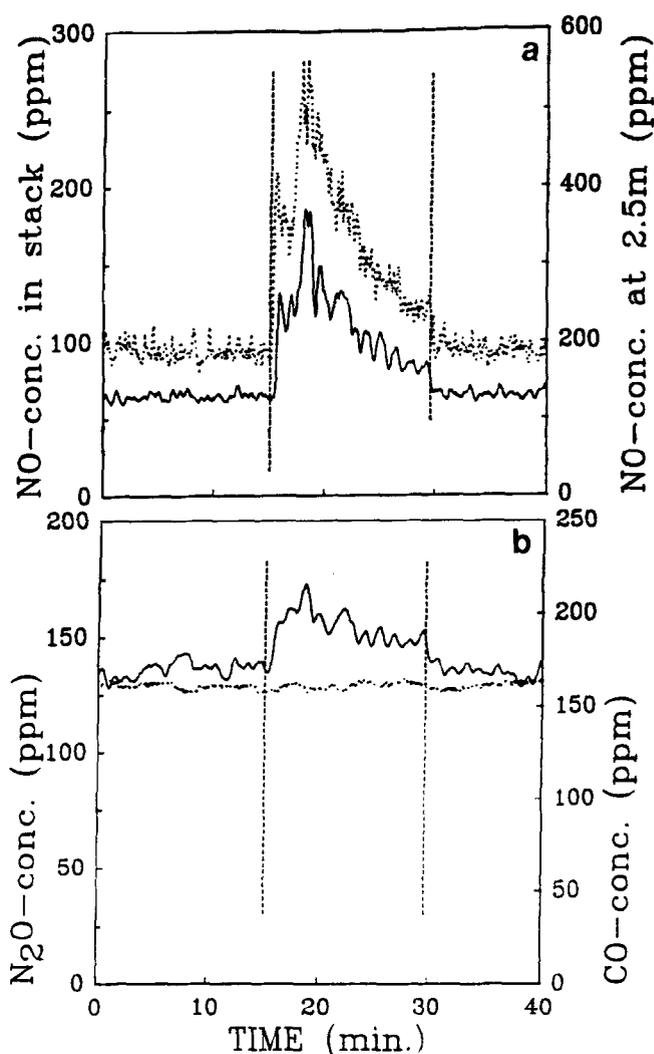


Figure 5 Supply of concentrated NO to the primary air duct at $t = 15-30$ min: (a) NO in stack (—) and NO at 2.5 m (· · ·); (b) N₂O (—) and CO (· · ·). NO supply = 410 ppm related to total air at $t = 18-22$ min. See Table 2 for operating conditions

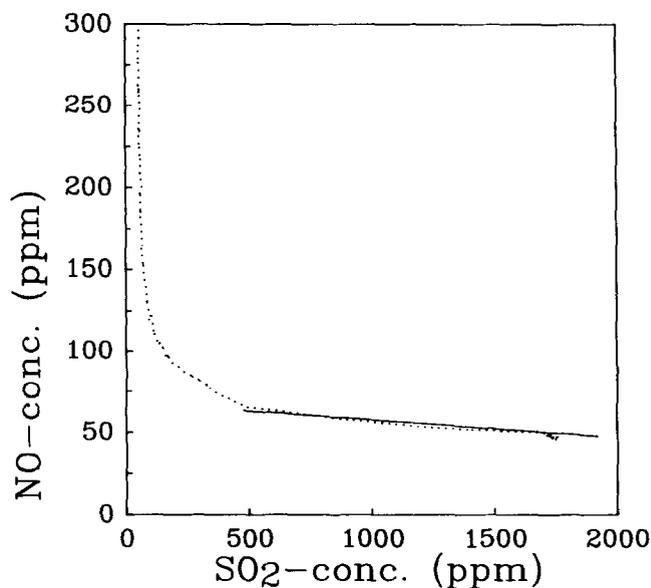


Figure 6 Exit concentration of NO as a function of SO₂ concentration for lime transient test (Figure 2) with Ca/S = 4.3 (· · · · ·) and test with SO₂ supply to sand bed (Figure 3) (—). Lines represent best fit of measured values

shift in the inclination of the SO₂-NO relationship at 500 ppm is clearly seen, and the part of the curve for SO₂ > 500 ppm represents the homogeneous effect. For comparison, the result from the test with SO₂ supply to the sand bed (Figure 3) is also shown. The two results coincide, supporting the assumption of a homogeneous relationship between SO₂ and NO since no lime was supplied during the test with the sand bed.

Heterogeneous effect of the lime

Figure 6 and Figure 2 at t = 1.76 h show that after the homogeneous part of the curve, NO increased while the SO₂ concentration was almost constant. The increase of the NO concentration should consequently depend on something other than the SO₂ concentration. Because of the excess supply of lime (molar ratio Ca/S = 4.3) and the high sulfur retention (98%), lime continuously accumulated in the bed. This period of the lime transient is therefore named the heterogeneous part, the increment in the NO concentration being interpreted as a consequence of the increased availability of CaO surface.

The highly catalytic CaO surface is exposed to the combustion gases with only minor protection from CaSO₄. The uncovered CaO surface is called the 'free' CaO surface and the amount can be estimated during the period of excess lime supply, knowing the lime efficiency, the lime supplied, and the lime leaving the boiler. This quantity of free CaO surface, estimated from the data of Figure 2, together with the NO concentration is shown in Figure 7. Apart from the homogeneous part in the very beginning, a linear relationship between the accumulation of free CaO surfaces in the bed and the increase of the NO concentration in the stack is observed.

Influence on N₂O

Based on measurements and literature data, Åmand et al.²¹ have given an outline of the formation and destruction of N₂O in CFB boilers in a simplified situation where no lime was added for sulfur capture. The results from the NO addition test given in Figure 5 are related to the important question discussed in Ref.

21: whether N₂O is produced when NO is reduced on char surfaces. Thus it should be considered whether the changes in N₂O, due to changes of SO₂ concentration and different amounts of free CaO surface, are caused indirectly, by changes of the NO reduction on the char surfaces, or directly for example by the CaO surface, on the formation and/or the reduction of N₂O.

Results from the NO addition test (Figure 5) were plotted as the N₂O formation versus the reduction of the NO added in the form of average values during the period between t = 18 and t = 22 min, and yielded a straight line, as seen in Figure 8. This shows that when 340 ppm of NO is reduced in the combustion chamber, 22 ppm of N₂O is produced. When NO and N₂O of the heterogeneous part of the lime transient in Figure 2 are plotted in the same way in Figure 8, a steeper decline of N₂O as a function of NO is obtained. Thus, there is an influence of the free CaO surface on the decrease of N₂O. This result is not surprising, since there are four independent laboratory investigations^{16,24-26} all showing that CaO is an effective catalyst for the decomposition of N₂O compared to silica sand. Lastly, when results from the SO₂ supply tests, as well as the homogeneous part of the lime transient, are drawn in Figure 8 in the same way, the decline differs from those of the two other cases. The reason for this is unclear at present.

Tentative explanation

Heterogeneous oxidation or decomposition of NH₃ on CaO surfaces is the general explanation for the increased formation of NO during lime addition^{7,11-16}. The CaO surface may favour the oxidation of CO as well, and this may be the reason for the reduction of CO concentration during the lime accumulation period of Figure 2. Since CO contributes to the reduction of NO on char surfaces^{27,28} this might be just as important as oxidation of NH₃ for the observed increase in the NO concentration.

The injection of SO₂ and the corresponding decrease of NO in the sand bed of Figure 3 are accompanied by

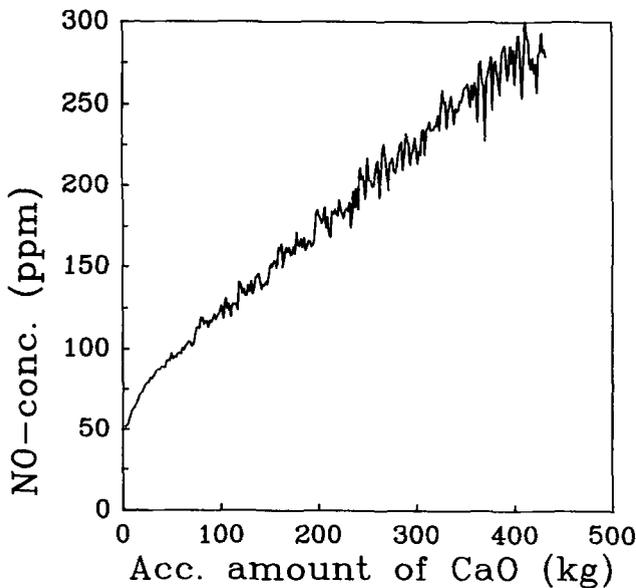


Figure 7 Exit concentration of NO as a function of the accumulated amount of free CaO surface in the combustion chamber during the lime transient test with Ca/S = 4.3

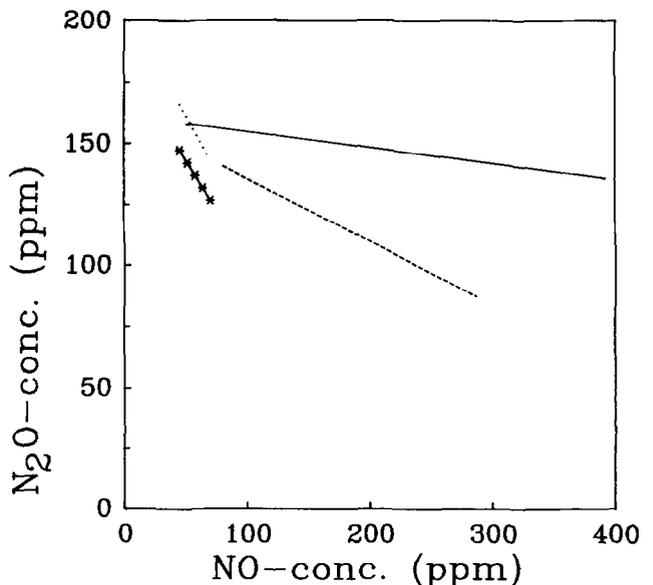
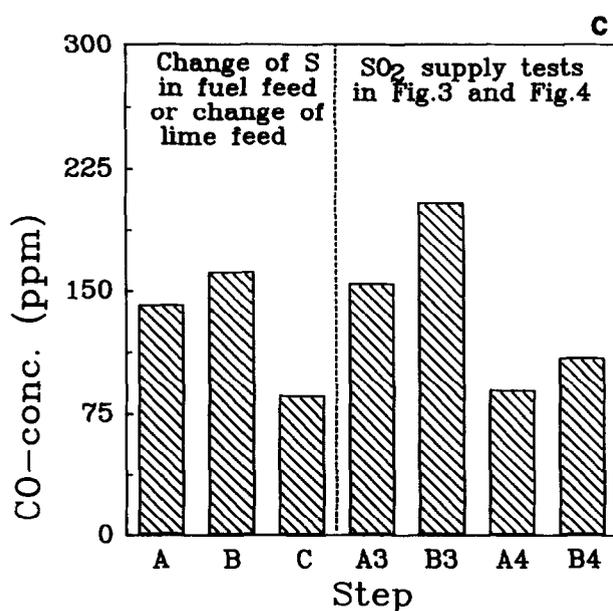
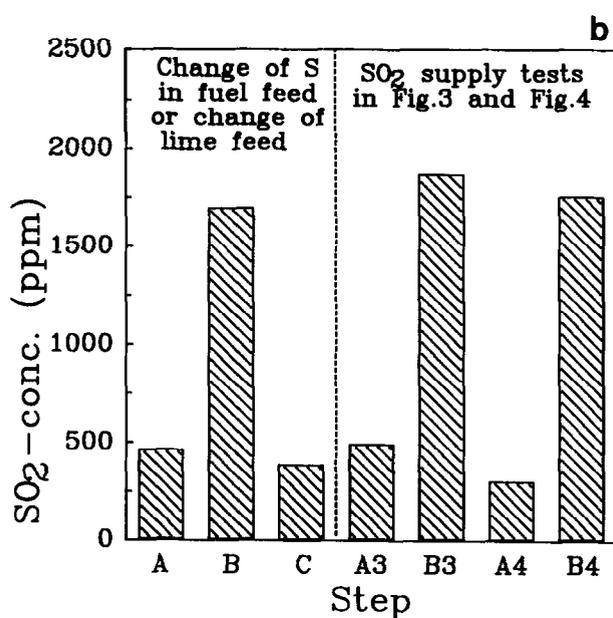
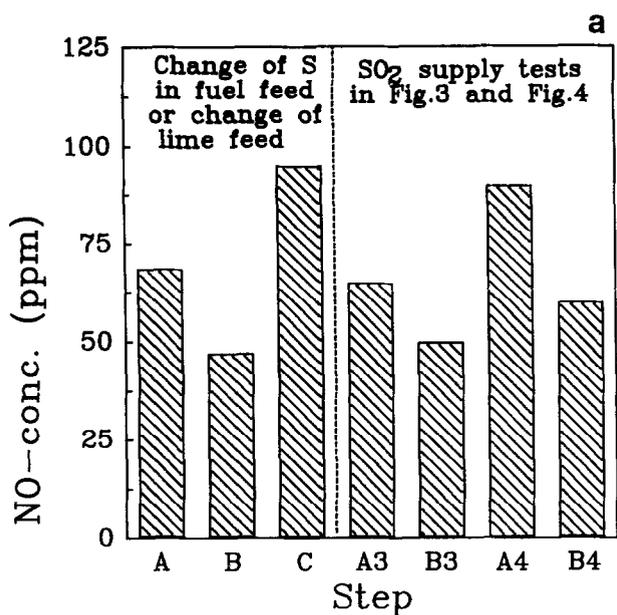


Figure 8 Exit concentration of N₂O as a function of NO concentration: —, NO supply to a bed of silica sand; ---, lime transient, heterogeneous part, Ca/S = 4.3; ·····, lime transient, homogeneous part, Ca/S = 4.3; ***, SO₂ supply to a bed of sand. Lines represent best fit of measured values



an increase of CO. It is therefore tempting to believe that there is a connection between SO₂-CO-NO, but it is not known at present how the CO oxidation is influenced by SO₂.

The interpretation of the results is based almost entirely on the gas composition measured at the exit of the boiler. The detailed processes in the combustion chamber are not assessed. The interaction between CO and CaO, and CO and SO₂ also requires further study, and this has been carried out in the complementary laboratory tests presented in part 2²².

Further evidence

The influence of SO₂ was demonstrated by addition of SO₂ gas to the primary air. A similar result can be obtained if the SO₂ concentration is changed by the same amount by switching between low- and high-sulfur coals or by limestone addition. This is illustrated in Figure 9, where the results from Figures 3 and 4 are compared with a test in which the low sulfur (A) and the high sulfur (B) cases are produced by switching between low- and high-sulfur coals.

Figure 9 confirms that the change in NO concentration achieved by switching between the coals is the same as that produced by addition of SO₂. Since no lime was present in the bed during steps A and B (Figure 9), these steps are most similar in magnitude to the data from the sand bed (Figure 3) and steps A3 and B3 (Figure 9). When limestone was added in step C and steady state conditions were attained, this step was most similar in magnitude to the data from Figure 4, where limestone also was added. This test is represented in Figure 9 as steps A4 and B4. The large difference in NO between step C, when lime was added, and step B, when no lime addition took place, depends on three additive effects: (1) the SO₂ effect being studied; (2) an influence of the lime; (3) an influence of the dilution of the char by lime in the recirculation ash flow, being run with a constant flow rate. Also, the results regarding N₂O from the low- and high-sulfur coals were similar to those of the SO₂ gas addition tests.

CONCLUSIONS

The tests carried out in the 12 MW CFB boiler lead to the following conclusions.

1. A change of concentration of SO₂ in the combustion chamber influences both the NO and the N₂O emissions, independent of the catalytic influence of the lime supplied for the sulfur capture process.
2. The influence of the concentration of SO₂ is the same, regardless of whether the SO₂ concentration in the combustion chamber is caused by SO₂ injection into the combustion air, the sulfur content in the fuel, or limestone addition to the combustion chamber.

Figure 9 Influence of sulfur content in fuel and lime supply on the emission of (a) NO, (b) SO₂ and (c) CO. Step A, low-sulfur coal, no lime supply; step B, high-sulfur coal, no lime supply; step C, high-sulfur coal, Ca/S = 1.8; step A3, step A from Figure 3, low-sulfur coal, sand bed, no SO₂ supply; step B3, step B from Figure 3, low-sulfur coal, sand bed, SO₂ supply 1400 ± 100 ppm; step A4, step A from Figure 4, high-sulfur coal, Ca/S = 1.6, no SO₂ supply; step B4, step B from Figure 4, high-sulfur coal, Ca/S = 1.6, SO₂ supply 2200 ± 100 ppm

3. Changes of SO₂ concentration and of the amount of free CaO surface are followed by changes in the concentration of CO, and this may influence the NO emission.

ACKNOWLEDGEMENT

This work was financially supported by the Swedish National Energy Administration.

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