REDUCTION OF N_2O EMISSIONS FROM FLUIDIZED BED COMBUSTION WITH AFTERBURNING

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ABSTRACT

One way to reduce N_2O emissions is to raise the temperature of the combustion gases. This can be done in the combustion chamber during conventional operation of a boiler, but also through injection of a secondary fuel, preferably in a separate combustion chamber. This paper reports investigations made at the 12 MW circulating fluidized bed boiler at Chalmers University of Technology, where in the absence of a separate combustion chamber, secondary fuel was injected into the primary cyclone. While the gas temperature was increased in the cyclone normal operation conditions were maintained in the combustion chamber.

A number of injection fuels were used, including Liquid Petroleum Gas, pulverized coal and pulverized wood. The reduction of N_2O was shown to be independent of the fuel used and only a function of the temperature achieved. Almost all N_2O could be removed by a temperature increase of about 150°C under otherwise normal conditions in the fluidized bed combustor. The other emissions were not increased by the fuel injection. On the contrary, the NO and CO emissions were reduced. Sulphur capture was not affected by the temperature rise in the cyclone. The behaviour of the cyclone as a separate combustion chamber and the burning characteristics of the different injection fuels are discussed.

INTRODUCTION

N_2O emission from fluidized bed combustion is known to decrease when the temperature of the combustion gases increases. This offers a possibility to reduce the N_2O emission. The simplest way is to increase the bed temperature of the combustor. In this case, however, the decrease in N_2O emission is accompanied by an increase in NO emission, a reduced sulphur capture and possible bed-material sintering problems. In order to avoid these drawbacks, the temperature of the combustion gases can be increased by combustion of a secondary fuel, preferably injected into a separate afterburning chamber. Earlier investigations of this technology, using Liquid Petroleum Gas (LPG) as injection fuel, showed that an N_2O

reduction ratio of 90% or more is possible (Leckner & Gustavsson 1991, Gustavsson & Leckner 1992). In the present report, results from experiments using other injection fuels are shown. A general characterization of afterburning as an N_2O abatement technique is also made.

EXPERIMENTAL

The investigation consisted of full-scale experiments in the 12 MW circulating fluidized bed (CFB) boiler at Chalmers University of Technology. The boiler is extensively equipped with measuring systems for gas composition, temperature, and gas and solid flows. A detailed description of the boiler has been given elsewhere (Gustavsson & Leckner 1992). The boiler has no separate combustion chamber for afterburning of a fuel to raise the flue gas temperature. In the absence of such an arrangement, fuel injection was made in the entrance of the primary cyclone and the cyclone was used as a combustion chamber for afterburning, Fig.1. The characteristics of the injection fuels are given in Table 1.

Table 1 Characteristics of the injection fuels

	Pulverized coal	Pulverized wood	Fuel oil
Moisture, %	2.2	6.2	0
Ash, % (dry basis)	9.3	0.3	0
Carbon, % (amf)	83.1	50.7	87
Hydrogen, % (amf)	5.4	6.3	13
Oxygen, % (amf)	8.8	42.3	0
Nitrogen, % (amf)	1.4	0.3	< 0.1
Sulphur, % (amf)	1.3	0.03	< 0.01
Net calorific value, MJ/kg (amf)	33.0	19.0	42.8

<u>Liquified Petroleum Gas (LPG)</u>: Ethane < 2.0 vol-%, Propane > 95.0 vol-%, Butane < 5.5 vol-%, Net calorific value: 46 MJ/kg, density 2.02 $kg/m^3{}_n$.

In general, no air was added at the location of the secondary fuel injection; instead, the oxygen content of the flue gases leaving the combustion chamber was used for combustion of the injection fuel. In certain cases, when the boiler was operating at a low excess-air ratio, additional air was introduced downstream of the cyclone outlet in order to achieve burn-out.

Normally, the tests were run on a "clean" sand bed, i.e. no limestone was added for desulphurization, but a few tests were made with limestone addition. The primary fuel was a low-sulphur bituminous coal with a nitrogen content of 1.5%(amf). Tests were run at bed temperatures of 850°C and 900°C and at O_2 contents in the flue gases leaving the bed of approximately 5%, 3.5% and 2%. The boiler load was 8 MW_{th}. In each case, three tests were run at different injection fuel ratios (IFR). (IFR = power of injection fuel divided by power of primary fuel.)

RESULTS

In Fig.2, the N_2O emission as a function of cyclone outlet temperature is shown for afterburning with LPG. In this case, the bed temperature, which is principally the same as the cyclone inlet temperature, was 850°C. As fuel was injected into the cyclone inlet duct, the cyclone outlet temperature rose. This was accompanied by a steep decrease in N_2O emission, so that at a cyclone temperature of 950°C only a very small N_2O emission remained. At lower excess-air ratios this low N_2O value was achieved at lower cyclone outlet temperatures. Typically, an LPG injection giving a cyclone outlet temperature of 940°C resulted in N_2O emissions decreasing from about 150 ppm without injection of fuel to below 30 ppm when the boiler was operated at 5% O_2, and below 10 ppm when operating at 3.5% or 2% O_2. An increase in cyclone outlet temperature of 100°C required an IFR of about 12% and an IFR of about 17% was needed to achieve the above-mentioned N_2O reductions.

The combustion of the injection fuel consumes oxygen, as shown in Fig.3. In the cases with $O_2 = 3.5$ and 2%, additional air was normally introduced after the cyclone.

In Figs 4 and 5 N_2O emissions are shown for cases with pulverized wood as injection fuel. The results are almost identical to those of LPG in terms of N_2O levels and temperature limits. This means that the N_2O levels are only a function of the temperature to which the combustion gases are raised. In Fig.6 this is verified by a comparison between all injection fuels for a bed temperature of 850°C and 5% O_2 in the flue gases. At a given cyclone outlet temperature, the N_2O emissions were the same irrespective of the fuel which was used to achieve that temperature.

Differences between injection fuels occur, however, in the amount of fuel needed to achieve a given cyclone exit temperature, and thereby a given N_2O reduction. This is shown in Figs 7 and 8, where N_2O reduction is shown as a function of injection fuel ratio for different fuels. LPG, fuel oil and pulverized wood seem to be almost equal in this respect. Pulverized coal, on the other hand, gave a lower N_2O reduction for a given injection fuel ratio. This can be attributed to incomplete combustion, which is further discussed below. The efficiency of combustion of the injection fuel is much influenced by the experimental conditions for afterburning prevailing in the cyclone of the CFB used. In a system designed and optimized for afterburning, the efficiency of combustion may be improved also for pulverized coal as an injection fuel.

Possible drawbacks of afterburning are increased NO levels, increased CO levels, and influence on sulphur capture. In Figs 9 and 10, NO emissions are shown for cases with pulverized wood as injection fuel. Instead of increasing emissions with temperature, a slightly decreasing trend was observed. In the case with 900°C bed temperature and 2% O_2, the reduction was quite significant. The results are analogous for other injection fuels. Afterburning thus has a positive influence on the NO emission. The picture is also positive for CO emission, which can be seen from Figs.11 and 12: afterburning contributes to lower CO emissions, provided that additional air is introduced after the cyclone in cases with low excess-air ratios in the bed.

The combination of afterburning and limestone addition to the bed could have effects on either N_2O reduction or SO_2 removal. A few experiments were therefore run with a high-sulphur coal (1.5% S) and limestone addition. In Fig.13 a comparison is made for N_2O emissions with afterburning with and without limestone addition, in the former case both with low-sulphur and high-sulphur coals. No effect on N_2O reduction can be seen; the correlation between cyclone outlet temperature and N_2O emission is the same in all cases.

Figure 14 shows the other aspect of the combination, i.e. the possible influence of afterburning on sulphur capture from temperature rise and reduction of the oxygen content. The experiments do not show any increase in SO_2 emission when the gas temperature was raised to about 1000°C. (The different levels of the curves of Fig.14 depend on the sulphur content of the fuel and the rate of limestone addition, $Ca/S = 1.5$).

DISCUSSION

Afterburning through fuel injection has been shown to be a possible way of N_2O reduction in a CFB combustor. A number of injection fuels, ranging from LPG to pulverized coal, gave the same reduction for a given rise in gas temperature. The nitrogen content in these fuels ranges from almost zero to 1.5% N. It is therefore shown that the contribution of N_2O formation from afterburning is negligible. Instead, the destruction of N_2O seems to be the dominant mechanism. N_2O is known to principally decompose according to the reactions:

$$N_2O + H \rightarrow N_2 + OH \qquad \text{(R1)}$$

$$N_2O + OH \rightarrow N_2 + HO_2 \qquad \text{(R2)}$$

$$N_2O + M \rightarrow N_2 + O + M \qquad \text{(R3)}$$

Of these, (R3) is the thermal decomposition. Bearing in mind the various volatiles contents of the injection fuels, they should give rise to different H and OH concentrations, which in turn should give different N_2O reductions. This is, however, not the case, which may have two explanations. Either the H and OH radical concentrations were high enough not to be rate-limiting for any of the injection fuels, or the thermal decomposition was the dominant N_2O removal channel. To elucidate this question, further investigations are obviously needed.

The fuels have different N_2O reduction efficiencies. As can be seen from Figs 7 and 8, LPG, fuel oil and pulverized wood show principally the same efficiency. Pulverized coal, on the other hand, has a lower efficiency. This can be understood if the heat balance of the cyclone is analyzed. The heating value of the injection fuel is not totally transferred to the combustion gases. Instead there are a number of possible losses:

I Transfer to the cooling tubes of the cyclone. (The cyclone is water-cooled, but protected with refractory.)

II Heating of recirculating bed material.

III Unburnt fuel carried away with flue gases.

IV Unburnt fuel recirculated back to the CFB combustor.

With the exception of loss number III, these losses are not thermal losses for the boiler, they are recovered by the boiler, but the heat does not become available for N_2O reduction. Also, all the losses mentioned are closely related to the plant in which the experiments were conducted. In a plant designed and optimized for afterburning, the losses may be eliminated or considerably reduced.

Figures 15 and 16 describe the result of a heat balance over the cyclone for the cases with LPG and pulverized coal as injection fuels respectively. The figures show the ideal temperature rise for an adiabatic combustor with complete combustion. Also the temperature drop due to the cooling of the cyclone is seen, counteracted to some extent by the normal CO combustion in the cyclone. Though the data representing the various losses (II, III and IV) are rather scattered, it is obvious that the heating of recirculated particles (II) takes a significant part of the heat release from the injection fuel in both cases. In the pulverized coal case, the losses in unburnt fuel carried away with the flue gases (III) and in unburnt fuel recirculated back to the combustor (IV) are also considerable. The last loss is presumably present for all injection fuel ratios as long as pulverized coal is used. On the other hand, the losses of unburnt carried away with the flue gases may not be a simple consequence of the fuel type; the higher injection fuel ratios compared to the LPG cases also contribute to the losses.

The analysis shown in Figs 15 and 16 should be taken as qualitative information, due to the scattered data. What is shown, however, is that there are a number of losses in the cyclone which influence the availability for N_2O reduction of heat release through fuel injection. Without losses, the figures show that an IFR of only 10% is sufficient to raise the gas temperature enough to remove practically all N_2O.

CONCLUSIONS

The investigation of afterburning through fuel injection has given the following results:

1 Injection of a secondary fuel upstream of the cyclone can provide at least 90% reduction of the N_2O emitted without fuel injection.

2 A higher N_2O reduction ratio was achieved at low excess air than at high excess air. Higher bed temperatures also made N_2O reduction by afterburning more efficient.

3 The resulting N_2O emission was only a function of the gas temperature. The type of fuel used to reach a given temperature was irrelevant.

4 Afterburning decreased the NO and CO emissions. At high bed temperature and low excess-air ratio, reburning-like effects were seen, resulting in significant NO reduction.

5 Sulphur capture was not affected by afterburning for temperatures up to 1000°C. Conversely, lime addition did not affect N_2O reduction by afterburning.

6 Heat was lost from the afterburning fuel through the cyclone walls, with recirculating bed material and, in the case of pulverized coal, as unburnt in the flue gases and unburnt going back to the combustor. This is a consequence of the design of the present experimental plant. In a plant optimized for afterburning these losses could be reduced, and an injection fuel ratio of 10% or lower would remove practically all N_2O, irrespective of type of injection fuel.

ACKNOWLEDGEMENT

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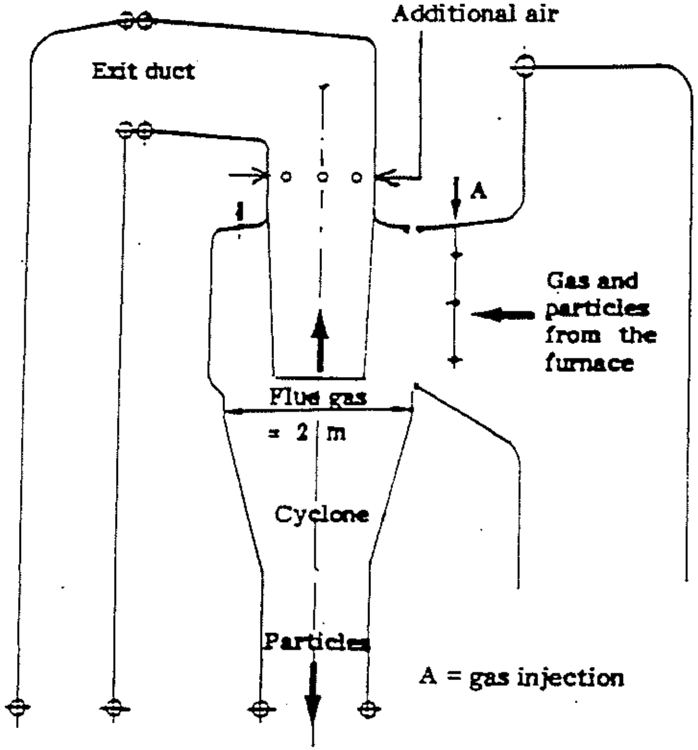

Fig.1 The cyclone part of the 12 MW CFB at Chalmers University of Technology.

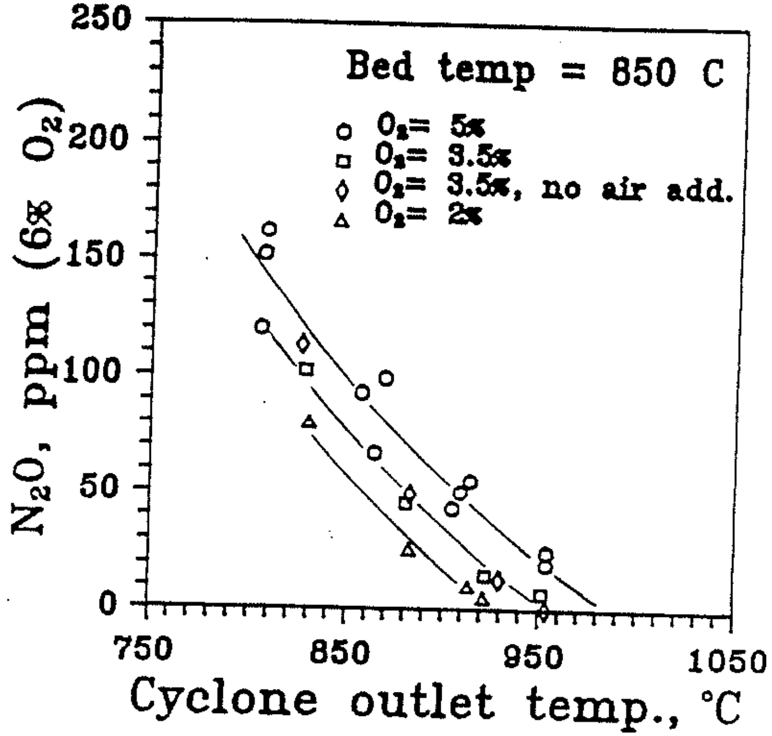

Fig.2 N_2O emissions *vs* cyclone outlet temperature in afterburning with LPG.

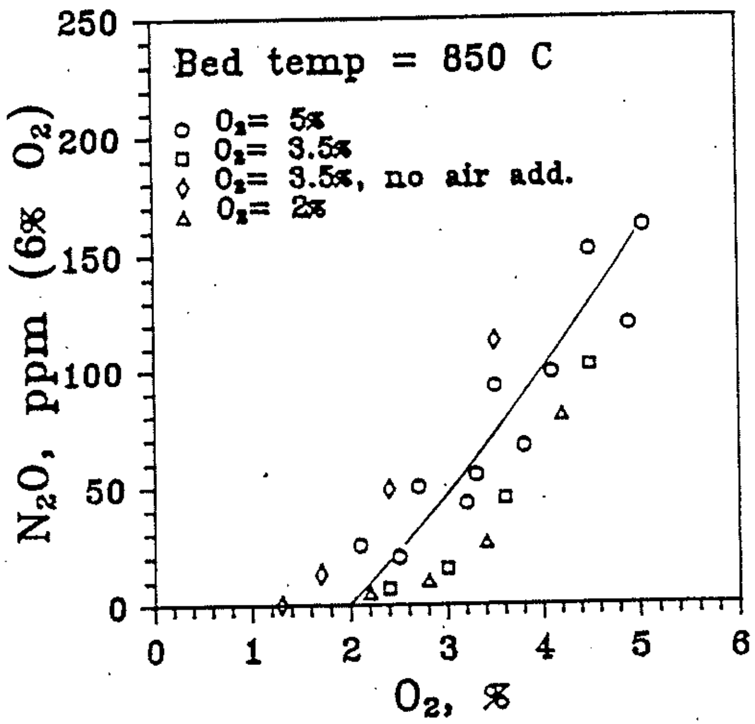

Fig.3 N_2O emissions *vs* oxygen content in flue gases for different excess-air ratios (before afterburning). In cases with $O_2 = 3.5\%$ and 2% respectively, additional air normally had to be introduced after the cyclone.

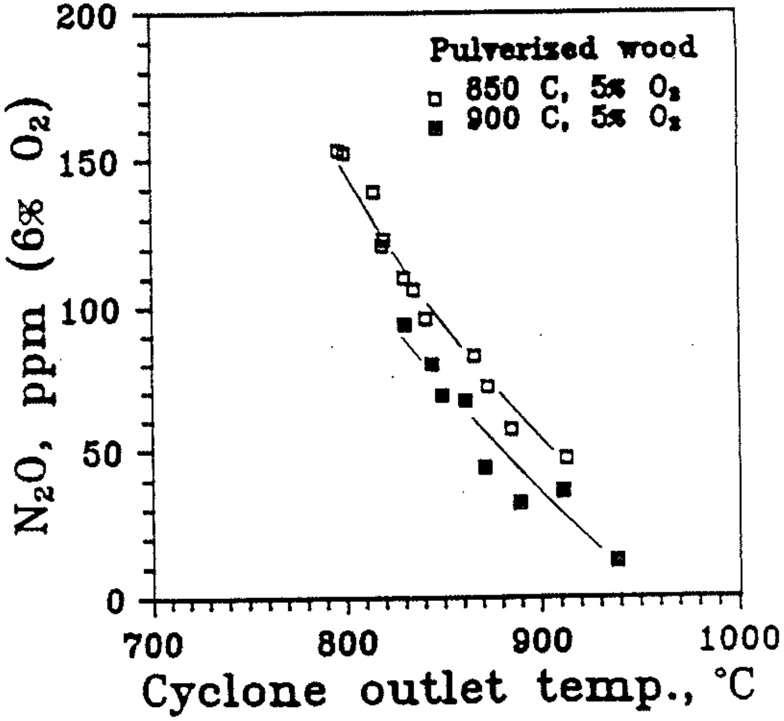

Fig 4 N_2O emissions *vs* cyclone outlet temperature in afterburning with pulverized wood. $O_2 = 5\%$ before afterburning

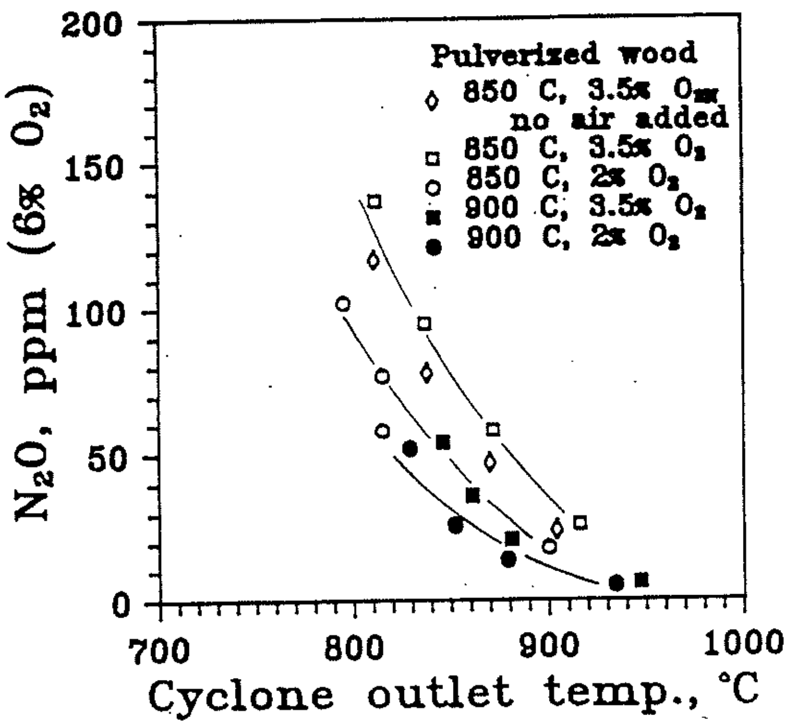

Fig.5 N_2O emissions *vs* cyclone outlet temperature in afterburning with pulverized wood. $O_2 = 3.5\%$ and 2% respectively before afterburning.

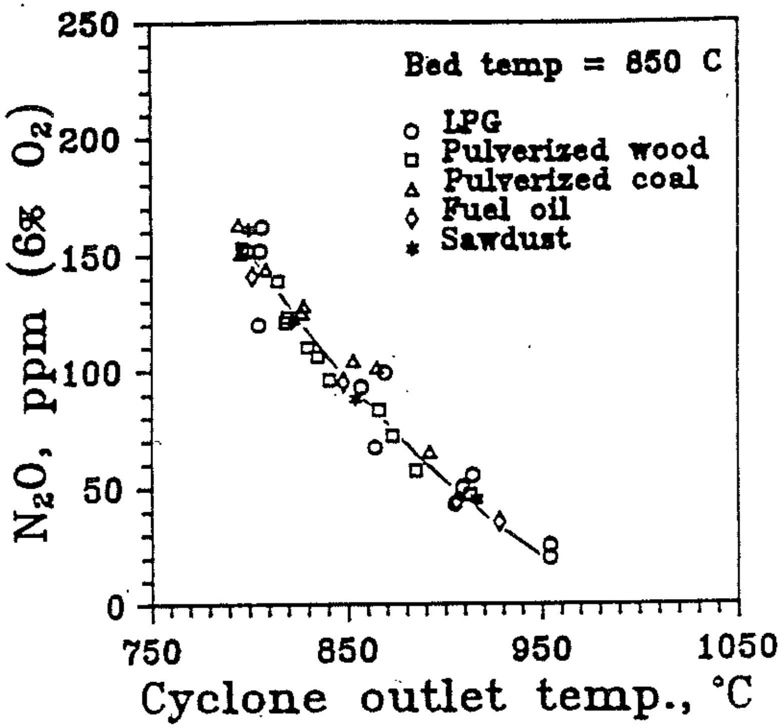

Fig.6 N_2O emissions *vs* cyclone outlet temperature. Comparison between different injection fuels. $O_2 = 5\%$ before afterburning.

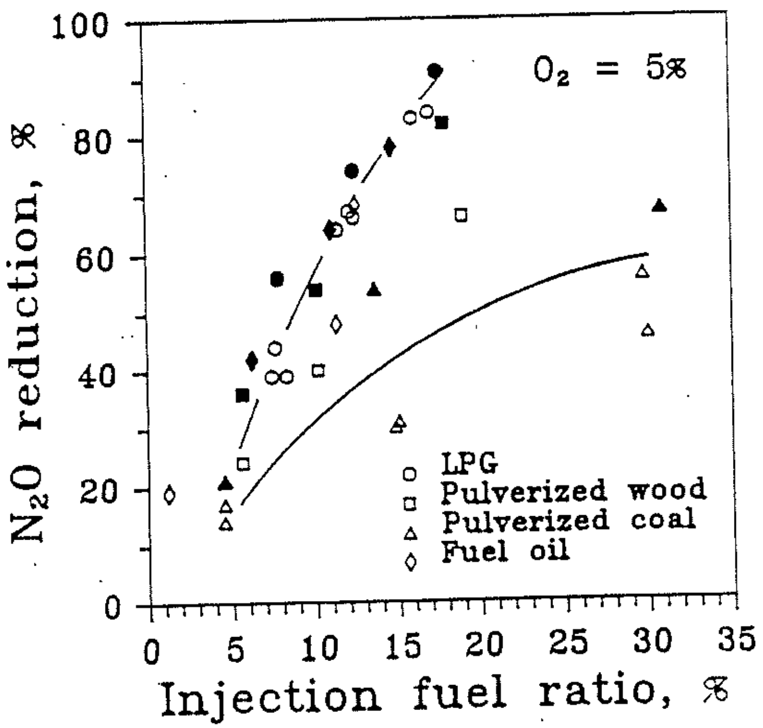

Fig.7 N_2O reduction (as compared to reference cases without fuel injection) *vs* injection fuel ratio.
Open symbols: bed temperature 850°C;
Filled symbols: bed temperature 900°C

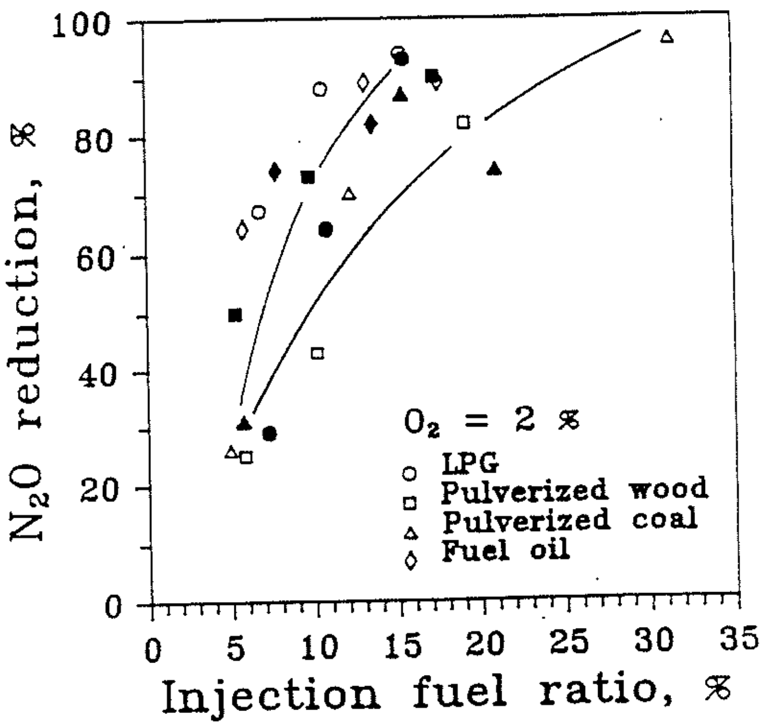

Fig.8 N_2O reduction (as compared to reference cases without fuel injection) *vs* injection fuel ratio.
Open symbols: bed temperature 850°C;
Filled symbols: bed temperature 900°C.

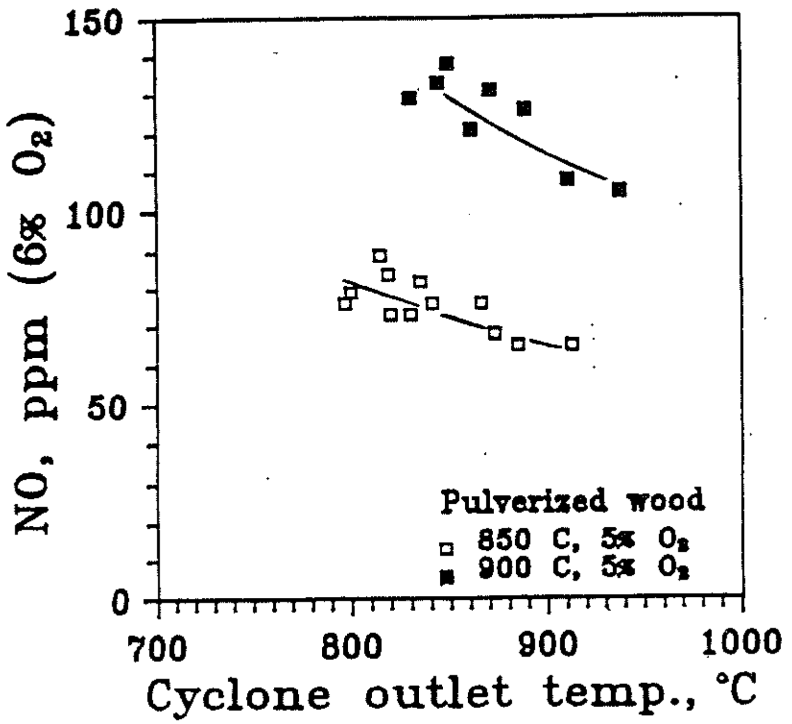

Fig.9 NO emission *vs* cyclone outlet temperature in afterburning with pulverized wood. $O_2 = 5\%$ before afterburning.

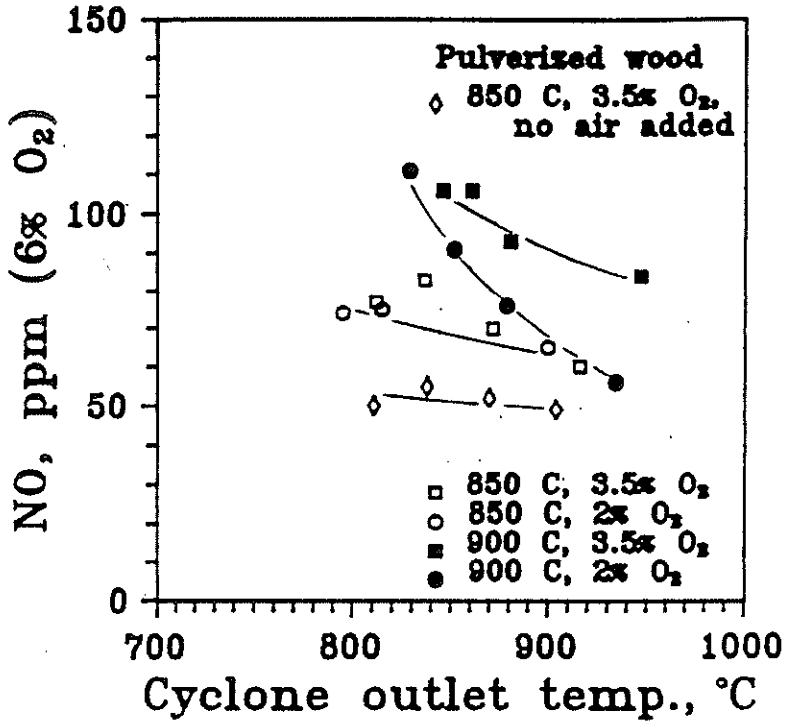

Fig.10 NO emission *vs* cyclone outlet temperature in afterburning with pulverized wood. $O_2 = 3.5\%$ or 2% before afterburning.

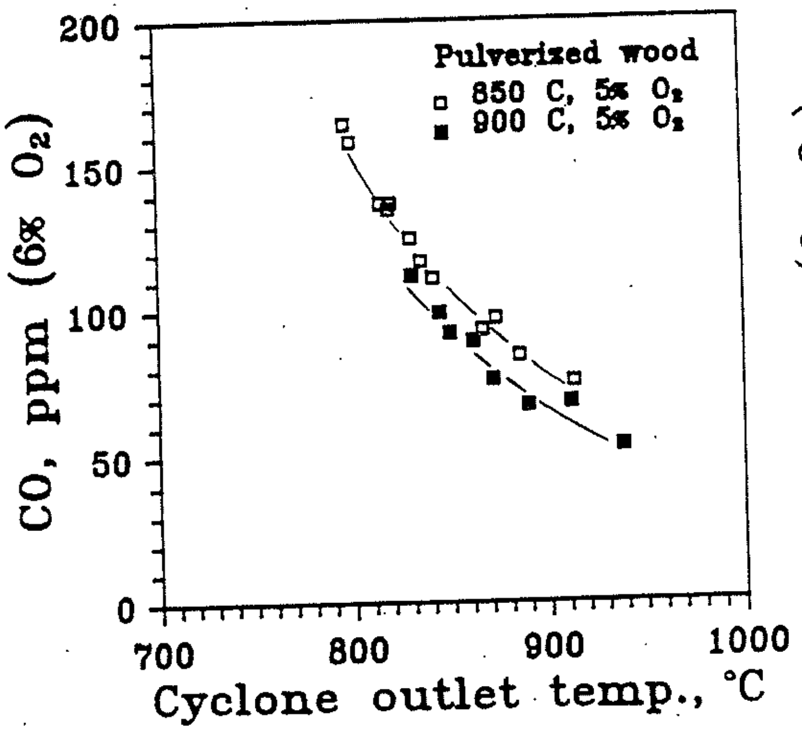

Fig.11 CO emission *vs* cyclone outlet temperature in afterburning with pulverized wood. $O_2 = 5\%$ before afterburning.

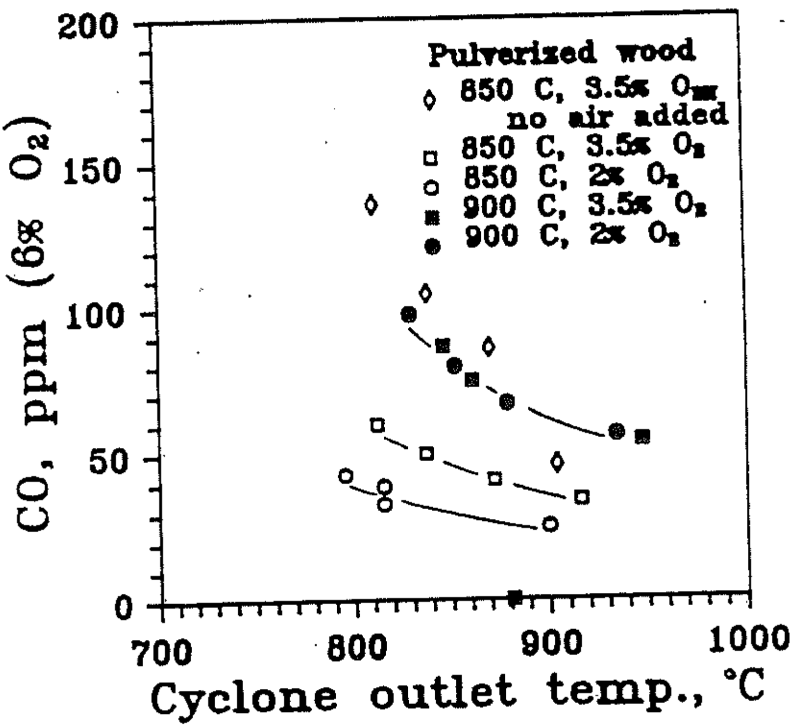

Fig.12 CO emission *vs* cyclone outlet temperature in afterburning with pulverized wood. $O_2 = 3.5\%$ or 2% before afterburning.

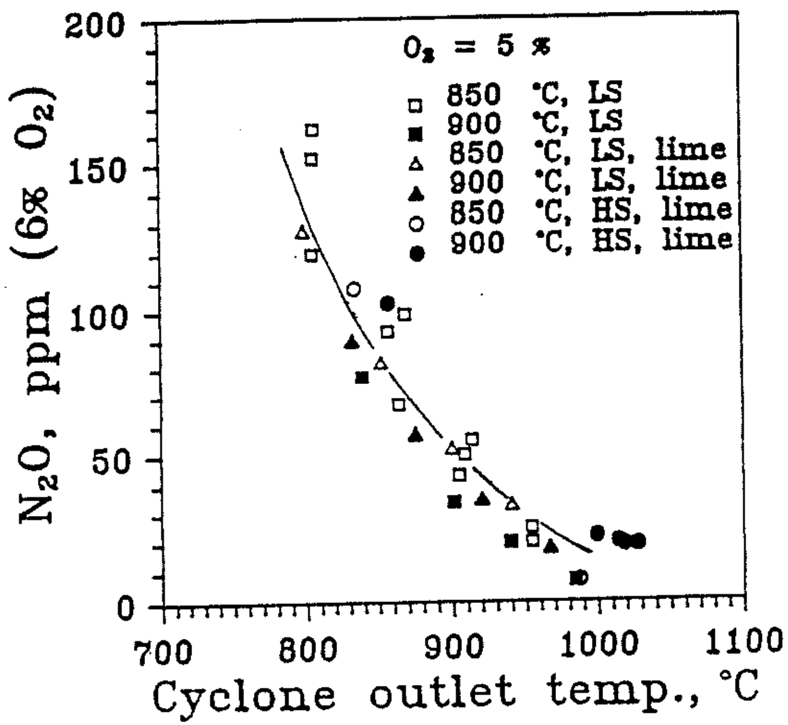

Fig.13 N_2O emissions *vs* cyclone outlet temperature in afterburning. Comparison between cases with and without limestone addition. LS = low-sulphur coal, HS = high-sulphur coal.

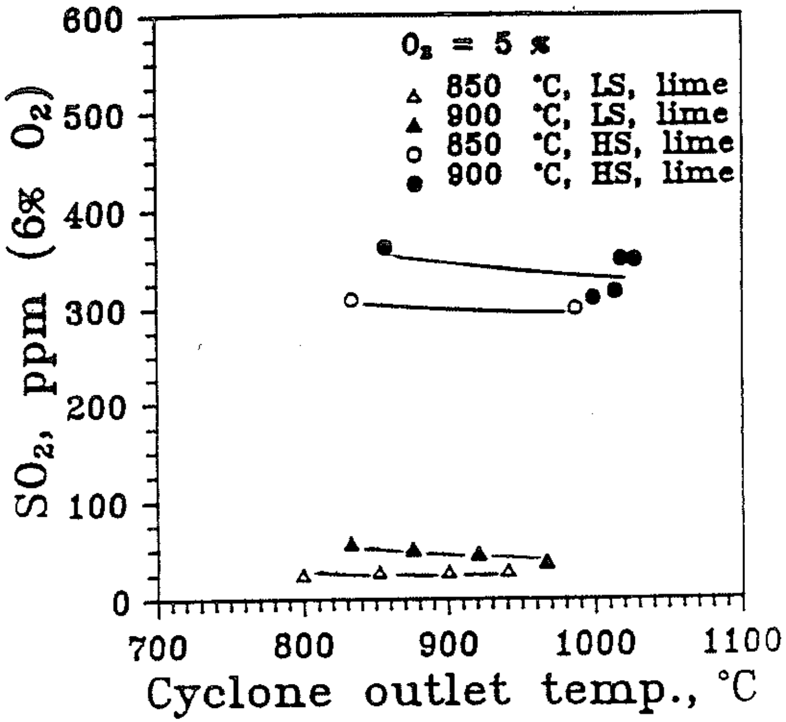

Fig.14 SO_2 emissions *vs* cyclone outlet temperature in afterburning. Limestone addition for sulphur capture. LS = low.sulphur coal, HS = high-sulphur coal.

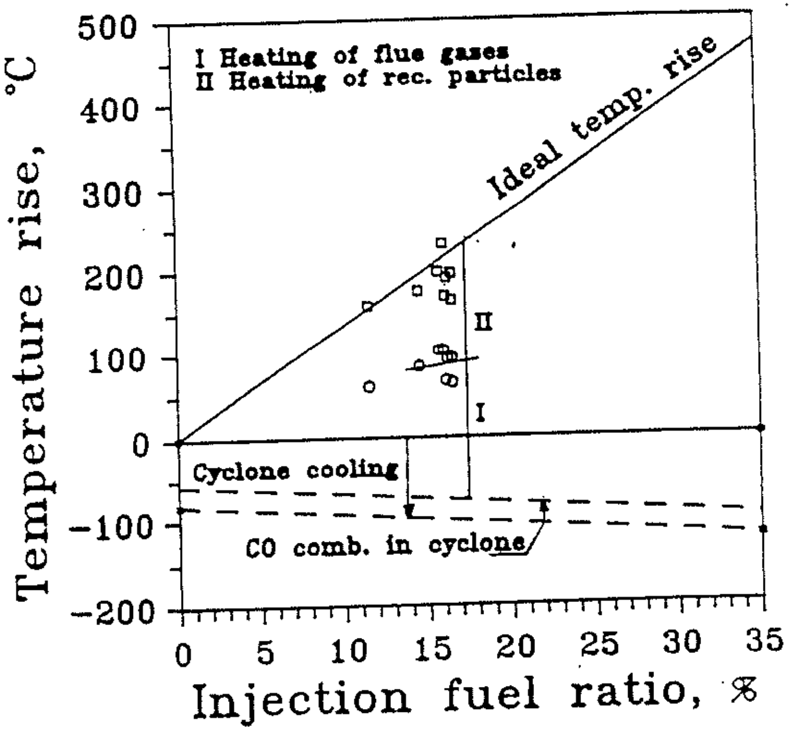

Fig.15 Heat balance of the cyclone.
Injection fuel: Liquified Petroleum Gas.

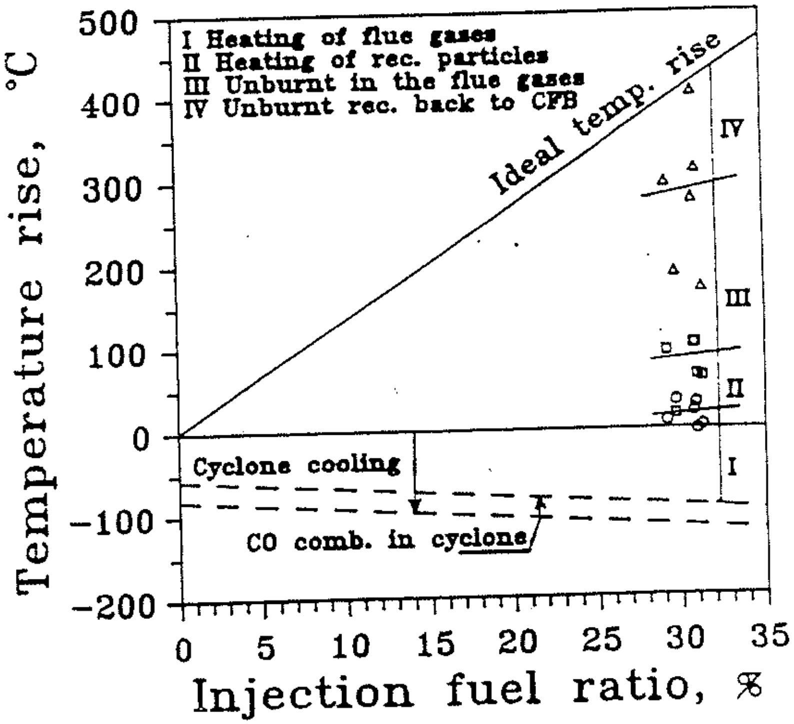

Fig.16 Heat balance of the cyclone.
Injection fuel: pulverized coal.