EMISSION CONTROL USING FLUIDIZED BED COMBUSTION

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INTRODUCTION

Developments on the energy scene have focussed attention on the relative abundance and low cost of coal as an energy source. Coal was the main energy source at the turn of the century but gave way to oil and gas because of the low and decreasing cost of the latter two sources, and because of the serious pollution problems associated with the combustion of coal. Whilst low sulphur coals can still be used in a technically simple way, there are increasing restrictions on sulphur emissions especially in the light of the growing concern with the effects of acid rain. It is estimated that by the turn of the century, 50% of power stations in the USA will have flue-gas desulphurization. Such equipment can add 30% to the cost of the power-station with the commensurate increase in energy costs. With high-sulphur coals, even flue-gas desulphurization becomes inadequate. The other main pollutants associated with combustion processes are nitrogen oxides for which no flue gas processing system has yet been developed.

South African coals generally have a high ash content, but low sulphur content (below 1%S) and can be burnt acceptably$^{2)}$. However, a growing coal export market has resulted in coal being beneficiated, the discard coal having a high ash content (up to 60%) and a high S content (up to 13%). Such coal cannot be burnt acceptably using current practice and has had to be discarded. The fluidized bed combustor is however a type of equipment in which such discard coals could be used with acceptable SO_2 and NO_x emission levels.

FLUIDIZED BED COMBUSTION PRINCIPLES

The basic operating principle of a fluidized bed combustor involves feeding coal to a chamber filled with a solid medium (typically silica sand plus limestone or dolomite). Air passes in an upward direction through the chamber; the bed material then expands to take on fluidlike properties.

Gas/solid intimate contact is enhanced leading to high average volumetric energy release rates. This, coupled with the low operating temperatures (typically 750–950°C) produces significant savings in equipment and operating costs. Other beneficial aspects of FBC's include their flexibility in coping with fuels of variable rank and quality, the reduction in the vaporisation of potentially harmful elements, and the elimination of slag fouling and ash clinkering. FBC's also afford an easy method for SO_2 retention by the in-situ addition of limestone or dolomite. The SO_2 liberated during coal combustion reacts with the calcined limestone according to the following reaction to yield an inert solid product:

$$CaO + SO_2 + \tfrac{1}{2}O_2 \rightarrow CaSO_4$$

In the temperature range encountered in FBC's, the contribution to NO_x levels from oxidation of atmospheric nitrogen is generally less than 10%.$^{3)}$ Final NO_x concentration levels are the result of competing reactions between oxidation of nitrogenous volatiles (exemplified by ammonia), direct reduction of NO_x with char, and coal ash catalysed reduction of NO_x by CO.

$$\begin{array}{lllllll} 2C & + & 2NO_x & \rightarrow & 2CO & + & N_2 \\ 2CO & + & 2NO_x & \rightarrow & N_2 & + & CO_2 \\ NH_3 & + & O_2 & \rightarrow & NO_x & & \\ NH_3 & + & NO_x & \rightarrow & N_2 & & \end{array}$$

Low NO_x levels can be achieved in FBC's by operating the unit as a partial gasifier. Primary air (less than the stoichiometric amount) is introduced in the lower region of the bed, where increased char availability enhances the heterogenous NO_x reduction. High combustion efficiencies are achieved by injecting the balance of combustion air to the solids disengaging area (the freeboard) above the bed.

AIR BORNE POLLUTANTS FROM COAL COMBUSTION

SO_2: Sulphur dioxide is the most widely discussed pollutant from coal combustion mainly as a result of the large quantity of emissions and the expense of conventional SO_2 control methods (e.g. wet scubbing techniques). Once discharged from the combustor vessel, the SO_2 may retain its chemical identity or it may be oxidised to sulphurous acid, a primary component of acid rain.

NO_x: The major ecological effects of NO_x include the addition of a component to acid rain damage; leaching of toxic metals from soil surfaces, and possible damage to nitrogen fixing bacteria from increased soil acidity.$^{4)}$ In addition, NO_2, which is the main component of NO_x after atmospheric oxidation, is a precursor to certain photochemical oxidants such as ozone and the peroxyacyl nitrates.

Particulates: Particulate removal from coal fired equipment is commonly effected by electrostatic precipitation or by other techniques, including the use of cyclones (density separation). The presence of particulates in the atmosphere can result in decreased visibility as well as adversely affecting land/air chemical balances because of the presence of trace elements and hydrocarbons which are absorbed on particulate surfaces.

EXPERIMENTAL WORK

In order to determine the effect of fluidized bed combustion on South African coals and discard coals, a series of tests were conducted. The first tests were carried out on a 300 mm diameter refractory lined fluidized bed combustor.

The encouraging finding from this apparatus led to the construction of a 1000 mm diameter water-cooled combustor, details of which are given in Ref 1. This combustor was capable of a heat output of 2 MW. For better control of operating conditions, it was subsequently decided to build a smaller, more flexible combustor which is described below.

EXPERIMENTAL EQUIPMENT

The fluidized bed combustor consists of a 100 mm diameter steel tube, 1.5 m in height, enclosed by a 300 mm diameter refractory lined pipe providing an annular space between refractory and combustor tube. A flow diagram of the test facility is given in Figure 1. A high pressure fan provides air to the nozzle-type distributor plate at the base of the combustor tube. A second fan, used to control bed temperature, provides cooling air, which is channelled through the annular gap. Throughout all tests, the bed material was initially heated by a 3 kW electric element strapped to the outside of the combustor tube. The combustion gases pass through medium and high efficiency cyclones before exhausting to atmosphere. The exhaust ducting is maintained under a slight negative pressure by an induced draught fan. Coal and limestone/dolomite are pneumatically fed to the combustor vessel. The bed height is maintained by intermittent removal of ash and spent sorbent material through a central discharge port. Secondary combustion air is dispersed at a point in the freeboard region 300 mm above bed level, by a gas sparger suspended in the combustion chamber.

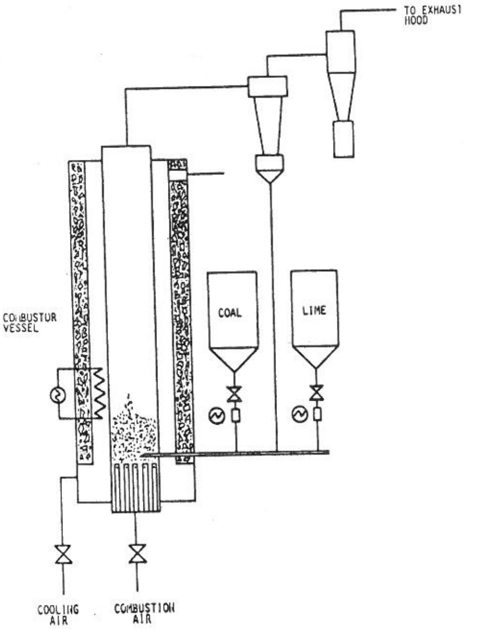

FIG. 1: Combustor tube layout

All gas analyses were performed on flue gas samples extracted from the cyclone inlet line. Sample preparation included particulate removal and two stage drying. Continuous monitoring of the flue gas oxygen concentration was performed with the aid of a polarographic analyser; CO, CO_2, SO_2 and hydrocarbon gases were monitored with non dispersive infra red analysers, and NO, NO_2 with a chemi-luminescence analyser, all of which were supplied by Beckman Instruments, USA.

EXPERIMENTAL PROCEDURE

Emission levels from a wide range of South African coals have been investigated. The comparison of limestone/dolomite quality was undertaken with high ash discard coals whose properties are outlind in Table 1. The purity of the carbonate materials is given in Table 2. The staged combustion tests were performed with a duff coal (Table 3), and Table 4 gives the proximate analyses for the two coal samples used to ascertain the effect of bed temperature and gas velocity on emission levels.

TABLE 1: SO_2 Emission coal samples analyses

	Vols	Fixed C	Ash	S	GCV (MJ/Kg)
Coal 1	24,6	28,6	46,0	13,5	14,4
Coal 2	21,9	22,1	55,0	2,9	11,7
Coal 3	–	–	–	1,3	–

TABLE 2: Carbonate stone purity

	L1	L2	L3	L4
$CaCO_3$	94,0	92,0	51,8	51,0
$MgCO_3$	–	–	38,5	40,3

TABLE 3: Staged combustion coal sample

Ash	C	H	N	S
14,3	67,7	3,8	1,6	0,6

TABLE 4: T. effect coal analyses

	Vols	Fixed C	Ash	S	N
Coal 1	22,3	60,0	15,2	0,7	1,9
Coal 2	26,8	55,0	14,6	0,6	1,7

All coal samples were prescreened to minus 2 mm before being fed to the combustor vessel. Silica sand, in the size range 0.85 – 1,4 mm was used as an inert bed medium. This gave use to a minimum fluidization velocity of 0.43 – 0.45 ms^{-1}.

In the staged combustion experiments, the level of primary fluidizing air flow to the combustor was maintained at between 90 – 100% of the stoichiometric limit (as measured by the oxygen level in the flue gases and the sharp rise in hydrocarbon gas concentration at this point). The balance of combustion air was added to the freeboard.

Combustion efficiencies were determined from the carbon content of the elutriated material.

RESULTS AND DISCUSSION

Effect of Temperature

Figures 2 and 3 show the effect of combustor temperature on SO_2 and NO_x emission levels respectively, for the range of gas superficial velocities investigated.

For both bed heights, corresponding to slumped measurements of 92 mm and 184 mm, a minimum in SO_2 emissions is observed between 1120 K and 1140 K, although the effect is less pronounced for the deeper bed. The highest emission levels are attained at the highest gas velocities, which are directly related to combustion efficiency.

NO_x emissions, however, tend to level off at about 1220–1270 K although this behaviour is less obvious for the deeper bed. This data is well substantiated in the literature. The fall off in emissions at higher temperatures is attributed to the contribution of char reduction of NO_x which becomes effective at temperatures above 1020 K.

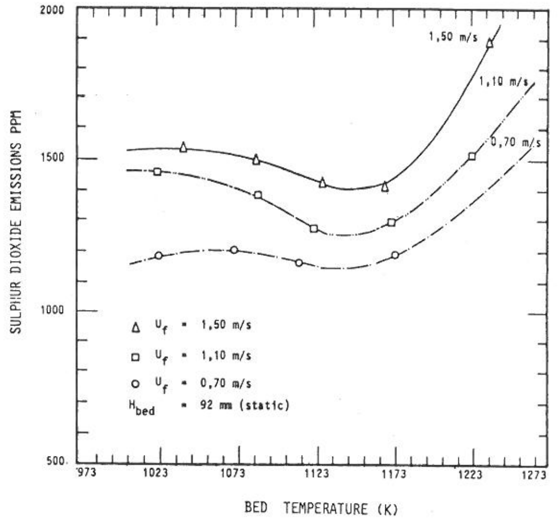

FIGURE 2A) : EFFECT OF TEMPERATURE ON SO_2 EMISSIONS FOR A BED HEIGHT OF 92 MM

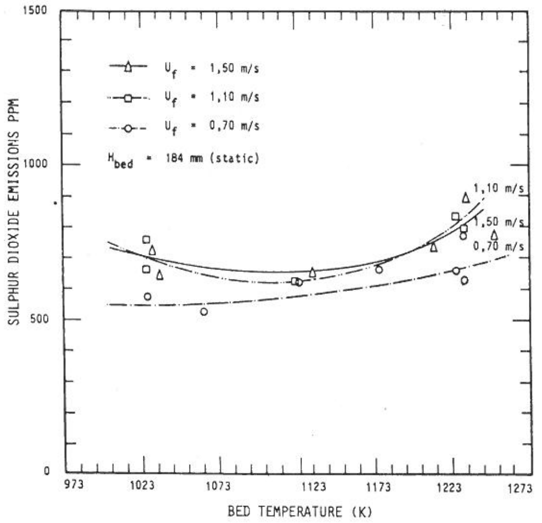

FIGURE 2B) : EFFECT OF TEMPERATURE ON SO_2 EMISSIONS FOR A BED HEIGHT OF 184 MM

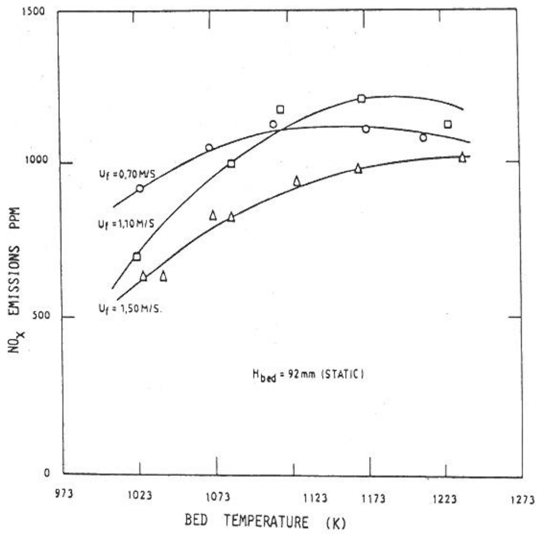

FIGURE 3A) : EFFECT OF TEMPERATURE ON NO_x EMISSIONS FOR A BED HEIGHT OF 92 MM

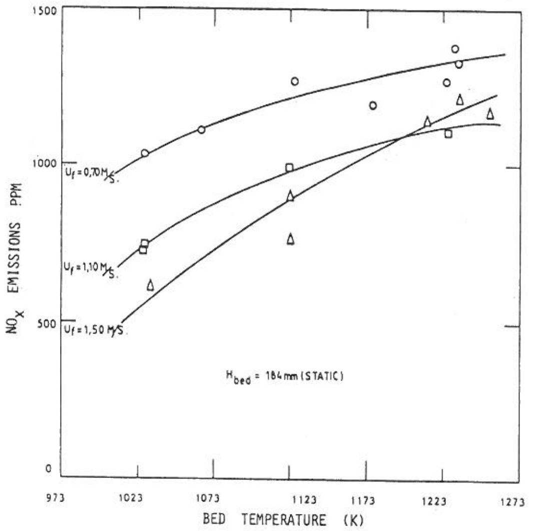

FIGURE 3B) : EFFECT OF TEMPERATURE ON NO_x EMISSIONS FOR A BED HEIGHT OF 184 MM

Control of SO_2 Emissions

A comparison of the potential of four locally obtained dolomites and limestones for SO_2 retention is given in Figure 4. Throughout all the tests, the combustor temperature was held constant at 1123 K and the flue gas oxygen concentration at 4% (V/V). Only one size fraction (0.85–1.4 mm) of sorbent stone was used. The superficial gas velocity was fixed at 1.0 ms^{-1} and the bed operated at a height corresponding to a slumped measurement of approximately 200 mm.

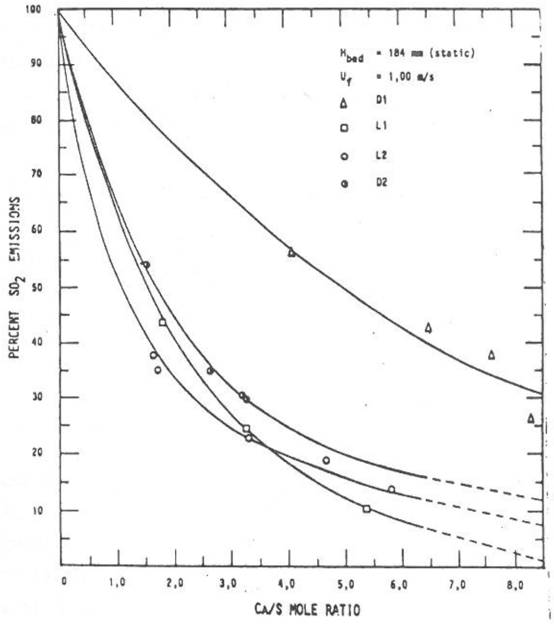

FIG. 4: Comparison of limestones/dolomites for SO_2 retention

When compared at an equivalent Ca/S mole ratio, both limestones are seen to effect a greater reduction in SO_2 emissions than either of the dolomites. The relatively high Ca/S values required to achieve an 80–90% reduction in SO_2 levels are linked to the large sorbent particle size, which was utilised in these experiments to prevent sorbent elutriation from the bed.

It is generally accepted that, for a given size fraction of sorbent, and all other things being equal, dolomitic stones are usually more reactive than limestones, a fact attributed to their much higher ratio of pore volume to CaO content. In dolomites, the inertness of the MgO component in the sulphation reaction prevents a complete blocking of the porous network, enabling continued access to the reaction surface.

Exceptions to this general trend have, however, been observed by other authors. It is therefore felt that a prediction of sorbent behaviour must consider the porosity and pore size distribution of the material. The influence of bed operation conditions on SO_2 retention is highlighted for both limestone samples in Figures 5 and 6. From Figure 5 (limestone 1) it can be seen that, for any given Ca/S molar ratio, a significantly greater reduction in SO_2 emissions is obtained with the deeper fluidized be,d equivalent to a static height of 184 mm. However, the effect of gas phase velocity is more pronounced with the shallow bed (static height 94 mm), the greatest reduction resulting from the highest velocity.

These findings were not noticeable in Figure 6 (limestone 2).

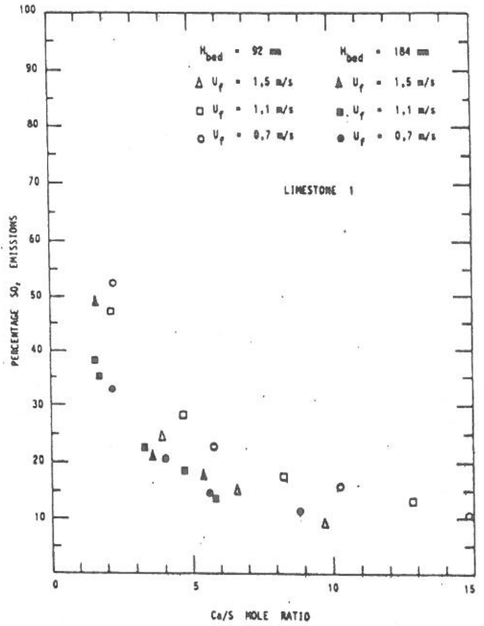

FIG. 5: Hydrodynamic effects on sulphur retention for limestone 1

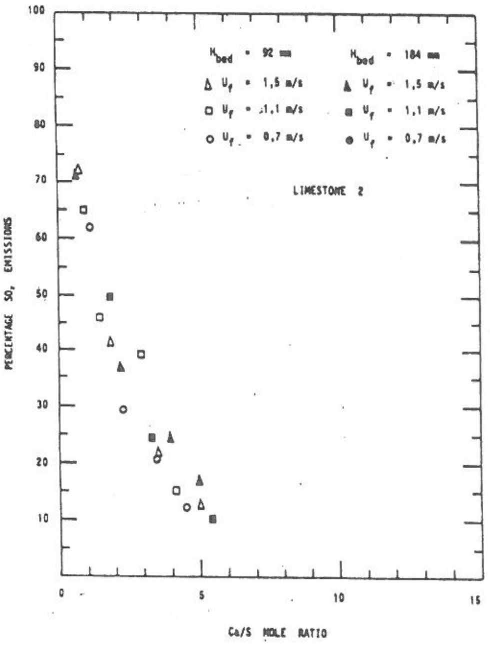

FIG 6: Hydrodynamic effects on sulphur retention for limestone 2

Staged Combustion

The effect of changes in excess air on flue gas emission levels of NO_x, during staged combustion, is shown in Figure 7, for a bed temperature of 1170 K and a constant slumped bed height of 184 mm. The emission index, E (kmol NO_x/kg coal) is used to represent NO_x levels because it is independent of dilution effects.

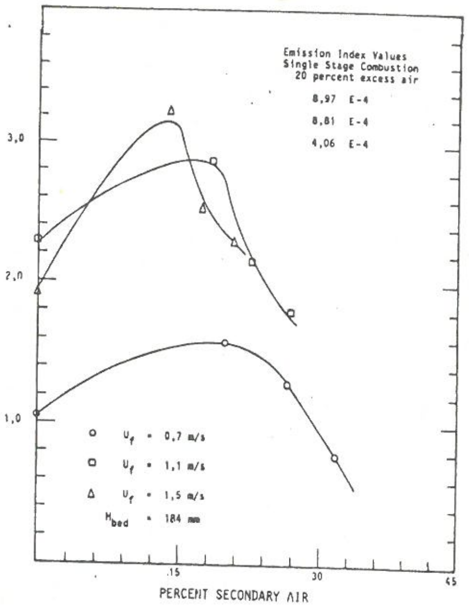

FIG. 7: Effect of excess air on NO_x emissions during staged combustion

The value of the emission index, which is proportional to NO_x concentration, is seen to remain constant or increase slightly with excess air up to a flue gas O_2 concentration of about 4%, and thereafter fall away.

Figure 8 shows the effect of increasing excess air, during the secondary combustion stage, on combustion efficiency. The combustion efficiency increases slightly with excess air to about 20%, then reaches a limiting value and begins to fall. Although CO burnout is more complete in the freeboard during staged combustion, increased char elutriation occurs causing the fall off in combustion efficiency.

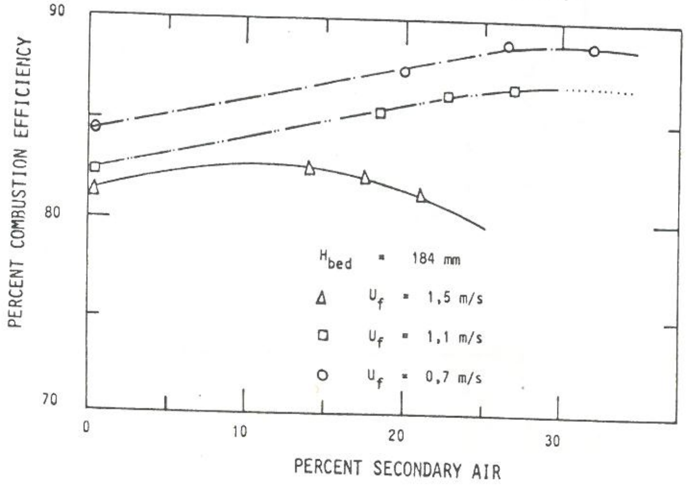

FIG. 8: Combustion efficiencies during staged combustion

It is proposed that, under the conditions tested in these experiments, most of the NO_x is generated *in* the bed and is reduced there to N_2. An increase in the amount of excess air in the secondary combustion stage serves mainly to increase char elutriation, resulting in a drop in combustion efficiency, and a concomitant fall off in NO_x emissions.

The other important trend evident from Figure 7 is that, for a given flue gas oxygen concentration, the lowest NO_x levels correspond to the lowest bed superficial gas velocity. This differs from the trend in Figure 3 for single stage combustion, where the inverse occurs. In single stage combustion, low gas velocities or deeper beds result in higher in-bed conversion of char or fuel nitrogen derivatives to NO_x, due to longer volatile residence times and a correspondingly increased level of available oxygen. In contrast however, during substoichiometric bed operation, longer gas residence times result in an increased reduction of NO_x.

A comparison of NO_x emission indices at similar operating conditions between Figures 3 and 5 does substantiate the claim that two-stage combustion leads to a reduction in NO_x levels.

CONCLUSIONS

High sulphur, high ash discard coals have been successfully burnt in a small scale atmospheric fluidized bed combustor, and exhibit the same temperature dependent SO_2 evolution pattern as good quality coal. SO_2 emissions are reduced by the addition of limestone or dolomite although relatively high Ca/S mole ratios are required to effect a significant reduction. Of the four sorbent materials tested, the limestones were found to be the more reactive.

A significant decrease in NO_x emissions is attainted by staged combustion. The freeboard gas velocity has been found to play an important role in determining the final emission levels and overall combustion efficiency.

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