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Coal Combustion Characteristics in a Pressurized Fluidized Bed

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Abstract–The characteristics of emission and heat transfer coefficient in a pressurized fluidized bed combustor are investigated. The pressure of the combustor is fixed at 6 atm. and the combustion temperatures are set to 850, 900, and 950 °C. The gas velocities are 0.9, 1.1, and 1.3 m/s and the excess air ratios are 5, 10, and 20%. The desulfurization experiment is performed with limestone and dolomite and Ca/S mole ratios are 1, 2, and 4. The coal used in the experiment is Cumnock coal from Australia. All experiments are executed at 2 m bed height. In this study, the combustion efficiency is higher than 99.8% through the experiments. The heat transfer coefficient affected by gas velocity, bed temperature and coal feed rate is between 550-800 W/m^2 °C, which is higher than those of AFBC and CFBC. CO concentration with increasing freeboard temperature decreases from 100 ppm to 20 ppm. NO_x concentration in flue gas is in the range of 5-130 ppm and increases with increasing excess air ratio. N_2O concentration in flue gas decreases from 90 to 10 ppm when the bed temperature increases from 850 to 950 °C.

Key words: Pressurized Fluidized Bed Combustion, Heat Transfer Coefficient, Flue Gas Emissions

INTRODUCTION

Coal-fueled pressurized fluidized bed combustion combined-cycle technology is one of the next generation power generating technologies which can increase power generation efficiency and comply with the emission regulations. Among various PFBC-CC technologies, PBFBC, PCFBC, and 2nd generation PFBC are representative technologies at the demonstration stage [Moritomi, 1994; McClung et al., 1997].

PFBC-CC generates electricity with a steam turbine operated by steam generated from heat of combustion and gas turbine operated by gas of high-temperature and high pressure.

PFB minimizes installation and operation cost of facility with smaller equipment, convenient manufacture, and simple operation. PFB becomes smaller by installing reactor excluding mechanical devices such as a fluidized combustor into a pressure vessel. PFBC is one of the clean coal combustion technologies [Miller, 1994]. The operation temperature of PFBC is as low as that of atmospheric fluidized bed combustion causing low NO_x emission. The pollution control is easy since in-situ desulfurization is possible [Kim and Park, 1987; Shun et al., 1996]. Heat generation per unit area is high [McDonald and Anderson, 1993].

In 1995, Korea Institute of Energy Research (KIER) started research and development on PFBC. In Korea, research and development (R&D) for PFBC began as a part of Clean Energy Program funded by the Ministry of Trade, Industry and Energy, started on November in 1994. In KIER the conceptual design of the PFBC was performed with Southeast University in China in 1995. In 1996 detailed design and construction of PFBC, 0.17 m bottom I.D.×2 m-high tapered bed and 0.25 m I.D.×3 m-high freeboard, were performed. The design parameters are as follows: 6 atmosphere operating pressure, 950 °C temperature, and maximum output of 0.14 MWt. In this research, the combustion characteristics of coal and emission of flue gases are considered experimentally. The target of the R&D is to understand the PFBC characteristics, to study engineering parameters, to increase the potential for technology development, and to identify the expected problems when the PFBC technologies are imported. This work concentrates on the characteristics of combustion, heat transfer, and flue gas emission of Australia coal of Cumnock.

To date, total operation of 900 hours has been performed. A continuous operation of 100 hours is planned to identify the effect of limestone on sulfur capture and the problems during long run operation. Data of coal combustion characteristics, heat transfer, and emission of flue gases will be accumulated and analyzed under various process variables such as temperature, superficial gas velocity, Ca/S ratio with sorbent species and excess air ratio. In this experiment, the combustion characteristics and the effects of operation conditions on SO_x, CO, NO_x, N_2O emission are investigated.

EXPERIMENTAL

Fig. 1 shows a schematic diagram of the PFBC facility in KIER. The system basically consists of coal and limestone feeders, a combustor within the pressure vessel, two cyclones and a bag filter. Compressed air passed through mass flow controller goes into the combustor through the distributor. The bed consisted of two parts: a combustion section (0.17 m bottom diameter, 0.25 m top diameter, tapered, 2 m height) and a free-board section (0.25 I.D.×3 m height), both covered with refractory of alumina and with insulator of ceramic wool.

Coal and limestone are fed into the combustor at 0.2 m above

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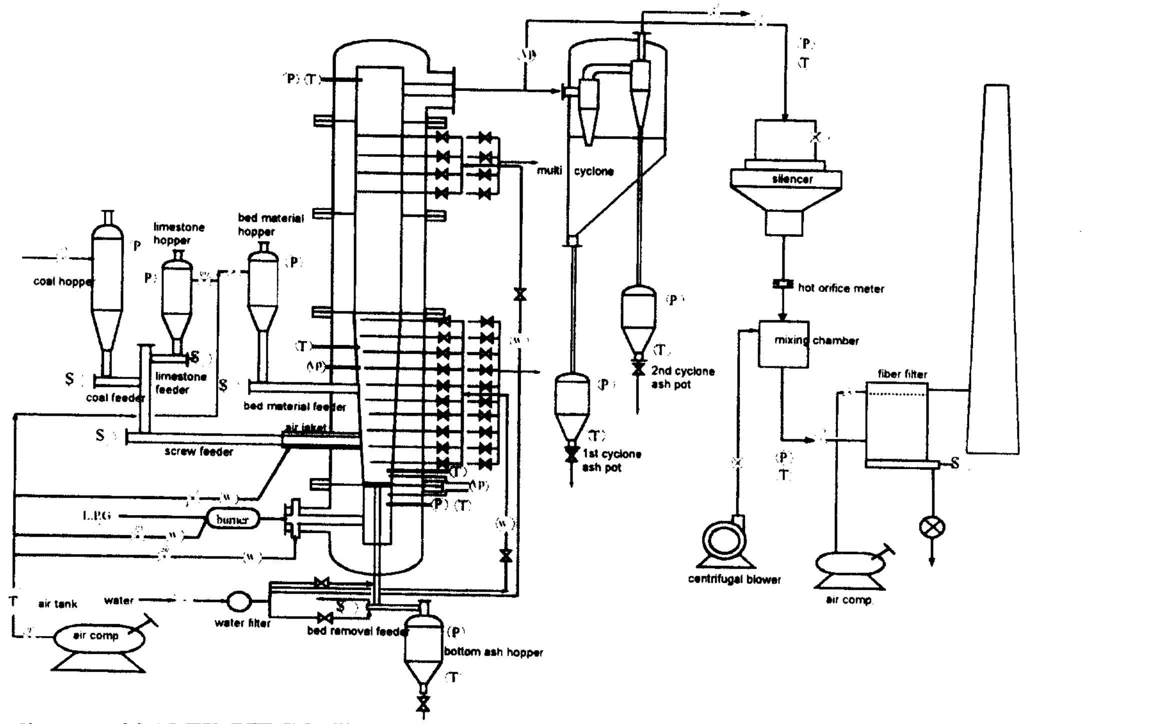

Fig. 1. Schematic diagram of 0.1 MWt PFBC facility.

the distributor by a screw feeder. Flue gas exits the top of freeboard and enters into two cyclones. The flue gas enters into silencer for reducing the pressure and exits to the atmosphere through bag filter followed by a stack.

Bed material is fed into the combustor in the pressurized vessel at atmospheric pressure. The bed is preheated by combustion of LPG. The coal is fed into the bed at 450 °C and the bed temperature increases abruptly owing to the combustion of coal. At 650 °C preheating by LPG is stopped and the combustion proceeds only by the coal combustion. From this point the temperature is stabilized to the desired one for experiment with a supply of air and coal and the vessel is pressurized. Bed material is fed to set 2 m bed height for effective heat removal.

For measurement of the heat transfer coefficient and bed temperature control by heat extraction, ten coaxial annular heat exchange tubes are installed in the dense bed. Cooling water mass flow, in and out water temperature from heat exchange tube, and bed temperature are measured for calculating heat transfer coefficient. The relation between amount of heat extraction, Q, log mean temperature between cooling water temperature and bed temperature, ΔT_l, overall heat transfer coefficient, U_o, and heat surface area, A, is as follows:

$$Q = U_o A \Delta T_l \tag{1}$$

The overall heat transfer coefficient can be expressed from the individual heat transfer coefficients and the resistance of the tube wall in the following equation.

$$U_o = \frac{1}{\frac{D_o}{D_i h_i} + \frac{x_w D_o}{k_m D_i} + \frac{1}{h_o}} \tag{2}$$

where D and h are heat exchange tube diameter and heat transfer coefficient, x_w and k_m are tube thickness and heat conductivity of heat exchange tube, and subscript i and o denote inside and outside, respectively. The internal heat transfer coefficient of heat exchange tube is calculated by the equations depending on laminar or turbulent flow [McCabe and Smith, 1976].

Operating variables are temperature, superficial gas velocity, excess air ratio and Ca/S molar ratio. Coal feed rate is determined with operating conditions. Coal properties of Cumnock and operation conditions are listed in Table 1 and Table 2. Flue gas concentrations such as O_2, CO_2, CO, SO_2 and NO_x are analyzed with gas analyzing systems.

Combustion efficiency can be calculated by the following equation

$$\eta(\%) = \left(1 - \frac{H_b F_b + H_f F_f + H_g F_g}{H_c F_c}\right) \times 100 \tag{3}$$

where H is heating value, F is feed rate, and subscripts c, b, f,

Table 1. Properties of Cumnock coal

Proximate analysis	Moisture	4.97%
	VM	31.63%
	Ash	11.12%
	FC	52.28%
Elemental analysis	Carbon	70.17%
	Hydrogen	4.79%
	Nitrogen	1.68%
	Sulfur	0.31%
	Oxygen	6.96%
Mean particle diameter (mm)		0.3
Calorific heating value (MJ/kg)		25.450

Table 2. Operation conditions

Superficial gas velocity (m/sec)	0.9, 1.1, 1.3
Pressure (atm)	6
Bed temperature (℃)	850, 900, 950
Excess air ratio (%)	5, 10, 20
Ca/S ratio	0, 1, 2, 4
Bed height (m)	2
Sorbent	Limestone dolomite

and g denote coal feed, bed ash, fly ash and CO in flue gas, respectively.

Technical, elemental, and calorimetric analyses are performed with MAC-400, Proximate analyzer (LECO. Co., USA), CHN-1,000 Elemental Analyzer, and ARR1261 Calorimeter (PARR Instrument Co., USA), respectively.

RESULTS AND DISCUSSION

In this research combustion efficiencies are over 99.8% at various operation parameters. In PFBC high combustion efficiency can be obtained without recirculation of bed material. It is presumed that increase of pressure results in increase of partial pressure of oxygen and, in turn, in increase of reaction rate. Fig. 2 represents temperature change curve of 10 hrs of PFB experiment after 12 hrs of preheating on the previous day. Temperature with time increases abruptly from initial ignition stage owing to the combustion of coal and LPG. The supply of LPG is shut off and combustion of coal maintains the temperature of FB. It is seen that the temperatures of FB maintain 950, 900, 850 ℃ depending on coal feed rate, excess air ratio, and by varying heat transfer tube cooling water flow rate.

1. Temperature History

Fig. 2 shows temperature profile with operation time in PFBC. Bed temperature is overlapped with 5 temperature profiles. Three profiles represent freeboard temperatures with different heights. The profile under 100 ℃ represents the temperature of air box.

2. CO Emission

Figs. 3 and 4 show CO concentrations in terms of bed temperature and freeboard temperature. CO is a toxic gas and regulated less than 250 ppm. In fact this level of CO concentration means reduction of combustion efficiency and affects economical and thermal efficiency of boiler [Anthony and Preto, 1995].

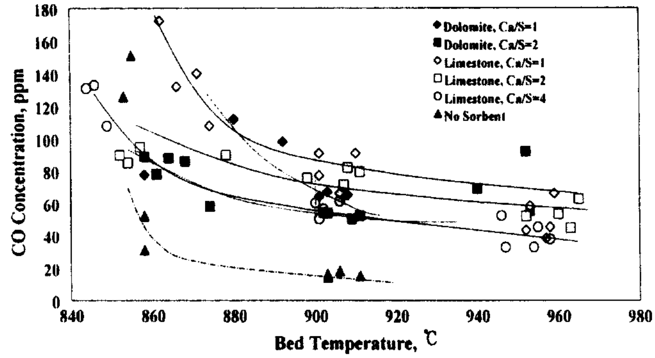

Fig. 3. Effect of bed temperature on CO emission.

In Fig. 3 it is seen that CO concentration decreases from 170 ppm to 40 ppm as the temperature of FB increases from 840 to 960 ℃. In nearby 850 ℃, the amount of CO emitted without sorbents ranges from 30 to 150 ppm. In case of sorbent injection, with the increase of Ca/S ratio, CO concentration is decreased. It is known that sorbents weakly catalyze CO oxidation in FBC systems [Lyngfet and Leckner, 1989]. Lyngfet and Leckner [1989] showed that $CaSO_4$ reacted to produce the lime, SO_2 and CO_2.

It is assumed that the sorbent reduced the CO concentration to the range of 80-130 ppm, while CO concentration is in the range of 30-150 ppm at 850 ℃ without sorbent. CO concentration is little affected by Ca/S mole ratio and the kind of sorbents around 950 ℃. There are many parameters affecting CO concentration due to the side reaction between sorbent, bed material and flue gases as well as incomplete combustion [Anthony and Preto, 1995]. With the temperature, it makes analysis complex even though reaction rate increases with temperature of bed and CO concentration decreases [Wallman and Carlsson, 1991].

Fig. 4 shows that CO concentration decreases from 100 to 40 ppm as temperature of freeboard increases from 650 to 780 ℃. When the dolomite is fed, CO concentration does not change much. There is no tendency with the kinds of sorbent and Ca/S ratio. The emission of CO and the fraction of unburned carbon are affected by the freeboard temperature and bed temperature. The lower freeboard temperature gives higher CO emission because of low reaction rate. In case of 950 ℃ of bed

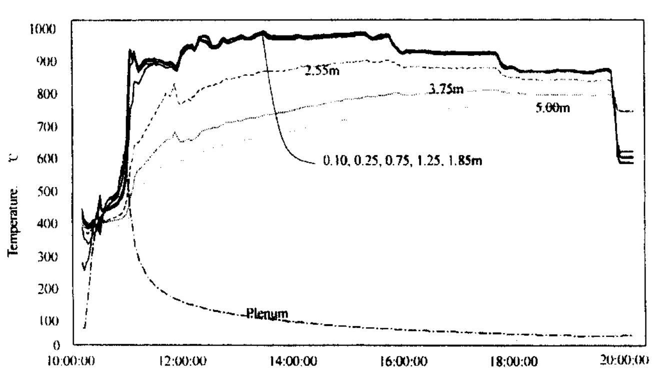

Fig. 2. Temperature profile with operation time.

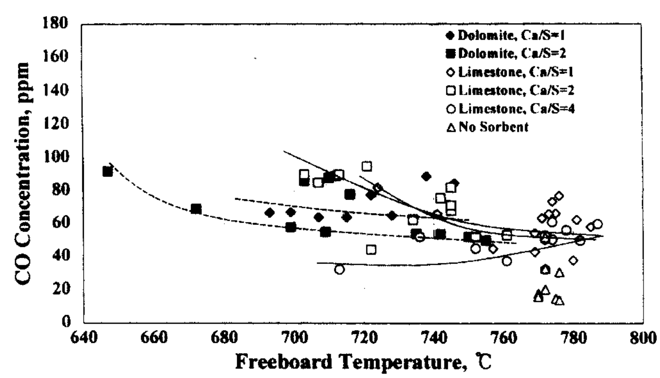

Fig. 4. Effect of freeboard temperature on CO emission.

temperature, most of CO concentration is lower than 60 ppm even though freeboard temperature is higher than 750 °C. It can be explained that the emission of CO is decreased by reaction with NO_x, gypsum produced from sorbents with SO_2 and complete combustion. At low temperature, concentrations of NO_x and gypsum are lower compared to those of high temperature, at which reaction rate is high and concentrations of NO_x and gypsum are high, especially at high pressure [Sarofim, 1994].

3. Heat Transfer Coefficient

The heat transfer coefficient is affected by various kinds of operation parameters. Heat transfer is induced from convection and radiation of particle and gas. Convection is mainly affected by bubbles which have cloud and wake. The heat transfer coefficients obtained are located in between 550 and 800 W/m^2 °C. These values are much higher than 200-500 W/m^2 °C of AFBC and CFBC [Grace, 1986]. The effect of the gas velocity on heat transfer is shown in Fig. 5. The heat transfer coefficient increases with fluidizing velocity, reaches a maximum value and decreases with higher fluidizing velocity. This is explained by the competing effects of decreasing particle residence time at the tube surface due to enhanced particle mixing caused by rising bubbles and decreasing solid hold-up adjacent to the tube surface when the fluidized velocity increases [Devaru and Kolar, 1995].

4. NO_x Emissions

NO_x emissions in terms of excess air ratio and coal feed rate are shown in Fig. 6 and Fig. 7. The effect of excess air ratio on NO_x emission is shown in Fig. 6.

The values of NO_x emissions are between 10 and 120 ppm.

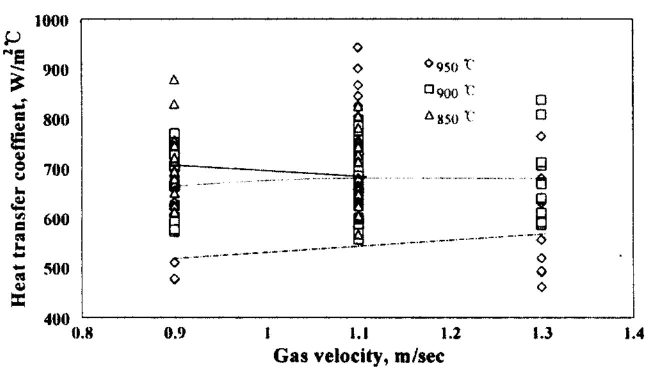

Fig. 5. Effect of superficial gas velocity on heat transfer coefficient at 400 mm.

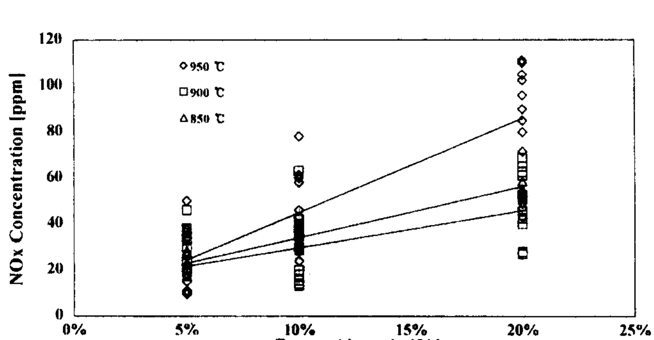

Fig. 6. Effect of excess air ratio on NO_x emissions.

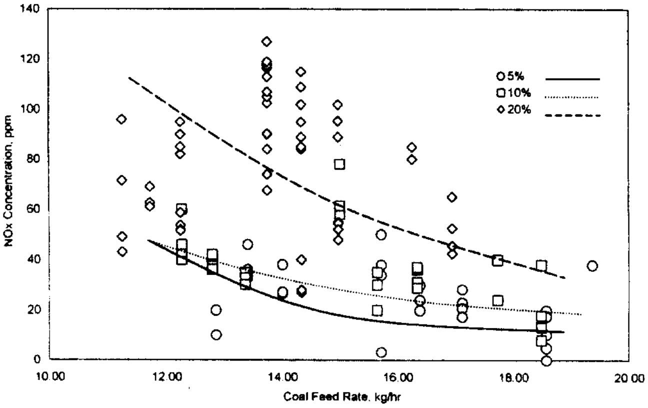

Fig. 7. Effect of coal feed rate on NO_x emissions.

These values are much lower than those of other investigators, 100-200 ppm [Wedel et al., 1993; Moritomi, 1994]. This can be explained by the relatively long gas residence time in the dense bed, where NO_x is reduced with CO and char. McDonald and Anderson [1993] explained that the NO_x increases with decreasing load in demonstration scale, because bed height decreases for maintaining appropriate heat transfer surface area with load. In Tomuro's experiment [Tomuro, 1995], changing bed height from 2 to 4 m, NO_x emission is decreased from 180 to 120 ppm. It is explained that char in dense bed reacts with NO_x and low NO_x concentration can be obtained with long residence time in the dense bed. With increasing excess air ratio from 5 to 20%, NO_x concentration is increased from 5-50 ppm to 30-120 ppm. And with a high bed temperature, 950 °C, NO_x emission levels maintain slightly higher concentrations than those at 850 and 900 °C. One of the reasons for higher NO_x concentration with the increase of bed temperature is that the char in the dense bed is completely combusted. The rate of NO_x oxidation reaction is increased with high O_2 concentration and high temperatures [Johnsson, 1994].

The effect of coal feed rate on NO_x emissions is shown in Fig. 7. With increasing coal feed rate from 12 to 18 kg/hr the NO_x concentration is increased from 40-100 ppm to 130 ppm, and decreased to 40 ppm.

This is explained by the longer gas contact time with the particles, because of high char fraction in same dense bed volume. It is possible for char particle concentration to be high in the dense bed at high coal feed rate, if the coal consumption rate is constant. There is a reduction reaction between char and NO_x, with catalyzed components such as ash and sorbents [Allen and Hayhurst, 1991]. Given operating conditions, the increase of the coal feed rate means that the condition of high temperature, high excess air ratio and low superficial gas velocity is changed to the condition of low temperature, low excess air ratio and high superficial gas velocity. Superficial gas velocity and temperature are competing parameters to control the NO_x emission.

At 20% excess air ratio, NO_x emission concentration is 60 ppm higher than that at 5 and 10% excess air ratios. At high excess air ratio, a relatively low amount of coal is fed into a fluidized bed at the same superficial gas velocity. The rate of reaction to NO is increased with the increase of O_2 concentra-

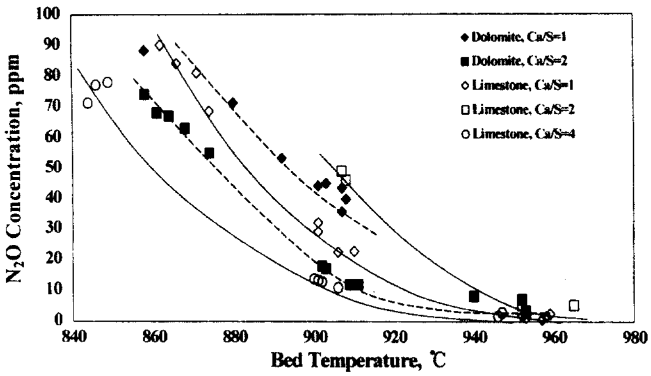

Fig. 8. Effect of bed temperature on N_2O emissions.

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5. N_2O Emission

In Fig. 8, N_2O concentration is shown as a function of bed temperature. Nitrous oxide is an air pollutant which acts both as a greenhouse gas and stratospheric ozone depletant. The emission of N_2O from fluidized bed combustion results from formation and reduction of N_2O. Combustion temperature, excess air ratio, and fuel type and bed material sorbent are also very important. It is seen that N_2O concentration decreases from 90 to 10 ppm as bed temperature increases from 850 to 950 °C.

Many researchers [Johnsson and Johanssen, 1995; Johnsson, 1994] have reported that N_2O concentration decreases since N_2O is oxidized and converted to nitrogen oxide as temperature increases. The type of sorbent and Ca/S mole ratio does not affect the N_2O concentration.

6. SO_x Emission

SO_x emission characteristic is shown as a function of the temperature of the bed in Fig. 9. SO_x emission characteristic is investigated with the type of adsorbents, i.e., limestone and dolomite, and Ca/S mole ratio. Dolomite is not used at atmospheric pressure since it is less effective than limestone in desulfurization ability [Sarofim, 1994; Shun et al., 1996]. At pressurized condition it is known to be better than limestone in desulfurization ability. It is reported that the reason is owing to $MgCO_3$ included in dolomite decarbonates around reaction temperature and pressure producing porous particles, into which the SO_2 can readily enter to react with $CaCO_3$. It is seen that SO_x concentration reaches a minimum around 850-900 °C with Ca/S ratio and increases as temperature increases above 900 °C. SO_x concentration is maintained under 120 ppm even with a Ca/S mole ratio of 2 in case of dolomite at bed temperature below 900 °C. SO_x concentration is less than 50 ppm with 4 of Ca/S mole ratio of limestone under 900 °C. The types of sorbents do not much affect sulfur retention in these experimental conditions, even though many researchers [Podolski et al., 1983; Sarofim, 1994] published that the dolomite is superior to limestone for sulfur retention ability at high pressure, and at temperature between 850-950 °C.

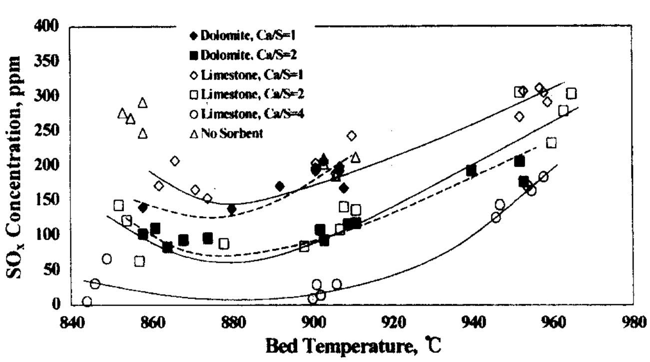

Fig. 9. Effect of bed temperature on SO_x emissions.

The effect of Ca/S mole ratio of sorbent on the concentration of SO_x is shown in this figure. The concentration of SO_x decreases from the range of 200-350 to the range of 10-180 ppm as Ca/S mole ratio increases up to 4. SO_x concentration decreases as Ca/S mole ratio increases.

CONCLUSION

In this study, it is found that PFBC as a next generation coal-fired combustor is outstanding equipment for coal combustion and considerably reduces the emission of CO, NO_x, and SO_x. The detailed conclusions obtained in the combustion experiment are as follows:

1. Combustion efficiency is higher than 99.8% in the experiments. The freeboard temperature little affects combustion efficiency.
2. CO concentration with increasing freeboard temperature is decreased from 100 ppm to 20 ppm.
3. Heat transfer coefficient is affected by gas velocity, bed temperature and coal feed rate. It is between 550-800 W/m^2 °C, which is higher than that of AFBC and CFBC.
4. There is a maximum value of heat transfer coefficient with the gas velocity in the experiments.
5. Heat transfer coefficient with increasing temperature is slightly increased.
6. NO_x concentration in flue gas is in the range of 5-130 ppm in this facility and increased with increasing excess air ratio.
7. N_2O concentration in flue gas is decreased from 90 to 10 ppm when the bed temperature increases from 850 to 950 °C.
8. The maximum sulfur retention temperature is between 850 to 900 °C in this experiments

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NOMENCLATURE

A : area [m^2]
D : diameter [m]
F : feed rate [Kg/sec]
H : heating value [MJ/kg]
h : individual heat transfer coefficient [W/m^2·°C]
k_m : heat conductivity of exchange tube [W/m·°C]
Q : heat flow rate [W/s]

x_w : thickness of heat exchange tube [m]
ΔT_l : log mean temperature [°C]
U_o : overall heat transfer coefficient [W/m^2·°C]

Subscripts

b : bed ash
c : coal feed
f : fly ash
g : flue gas
i : inside
o : outside

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