Combustion Characteristics of Thai Lignite in a Fluidized Bed

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Abstract

Combustion characteristics of Thai lignite, with 42% ash and 1.9% sulphur content, in a laboratory scale bubbling fluidized bed was investigated. The aims were to determine the influence of fuel particle size, excess air, and air staging on combustion efficiency, combustion regime, and concentrations of major combustion gas species. The results show that the combustion efficiency is strongly influenced by excess air with a drop of about 20% when the latter is varied from 20 – 100%; and that smaller-sized lignite particles yield higher combustion efficiency at the expense of much higher CO emission. With low excess air, in-bed combustion predominates, while both in-bed and freeboard combustion takes place at high excess air. It is shown that NO can be reduced by decreasing excess air, while N_2O tends to be minimized when the excess air is the range of 40 – 60 %. SO_2 decreases significantly with increasing excess air in the lower excess air region. Ash particles, and hence their size, are believed to play an important role as a sulphur absorbent. In-bed air staging results in a decrease in CO emission but also a drop in combustion efficiency. A slight increase in combustion efficiency is possible with freeboard air staging, which is also seen to be more effective in NO reduction than in-bed staging. N_2O decreases with increasing air staging--both in-bed and freeboard--with the latter being more effective. While in-bed staging has little effect on the emission of SO_2, the ability for in-bed sulphur retention can be adversely affected by freeboard staging.

1. Introduction

Fluidized bed combustion (FBC) technology offers significant advantages for generating steam from a broad range of fuels. FBCs can be designed to burn almost any solid, semi-solid, or liquid fuel without the use of a supplemental fuel, as long as the heating value is sufficient to heat up the fuel and to drive off the moisture, and preheat the combustion air. FBC technology has proved to be particularly effective in the utilization of low rank coal (lignite and subbituminous) [1,2,3]. Although the use of FBC technology for burning low rank coal has been widely studied, the characterization of the combustion behaviour of each particular coal to be used is necessary in order to obtain useful information for evaluation, design and proper operation of FBC facilities. The aim of this work is to investigate the fluidized bed combustion characteristics of Thai lignite, which generally has a high ash content of more than 30 %, a high sulphur content in the range of 2-7 %, and a low heating value. Special interest is focussed on determining the influence of major operating parameters including fuel particle size, percent excess air, and the degree and mode of air staging (in-bed staged and freeboard staged) on combustion efficiency, combustion regime, and various gas emissions, including CO, NO, N_2O and SO_2.

2. Experimental set-up

The experiments were performed in a fluidized bed reactor schematically shown in Figure 1. The reactor is made of 310S stainless steel cylindrical tube of 115 mm internal diameter and 2.4 m height, with outside ceramic fiber insulation. Silica sand, 2,600 kg/m^3 density and 500 μm mean diameter, was used as the bed material. Fluidizing air from a compressor was introduced into the bed through an orifice-type distributor with 96 holes of 3-mm -diameter each.

The reactor was heated by hot air which was preheated by an 18-kW electric-heater. The temperature at the entry of the bed was typically 600 °C and in the sand bed about 500 °C. This temperature is sufficient for the ignition of lignite used as start up fuel, and to raise the bed temperature to the operating point, whose typical value lies above in the range of 750-900 °C. For air staging operation, secondary air was introduced at either one of two separate ports located above the distributor plate at 225 mm (in-bed) and 525 mm (freeboard). Analysis of the fuel is given in Tables 1. and 2.

Table 1. Proximate analysis (%wt) of lignite (Mae Moh Mine)

Moisture	Volatile Matter	Fixed carbon	Ash	LHV (MJ/kg)
8.5	29.8	19.97	41.7	11.6

Table 2. Ultimate analysis (% d.a.f.) of lignite (Mae Moh Mine)

C	H	O	N	S
52	6.5	38.5	1.1	1.89

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Entrained particles were separated from the exhaust gas by a cyclone. Eighteen K-type thermocouples allowed the continuous measurement of temperature variation along the height of the bed, and pressure drop was also monitored continuously with a pressure transducer.

Gas samples could be taken through sampling probes at various positions along the bed. In this experiment, the gas samples were taken continuously at 2,325 mm above the air distributor, just before the freeboard exit. All the samples were taken in the axis of the reactor. The flue gas was pumped into an on-line analyzers: NDIR for carbon oxides, NO_x, N_2O and SO_2, and paramagnetic for oxygen. Data sampling was done when steady state was reached, with at least 300 data points collected for each condition. Temperatures, concentrations and carbon combustion efficiencies are defined by $x(t)$ and are random variables. Their distributions are unimodal, mode value is x_M. The intervals on which the probabilities prob$\{x(t)\}$ =0.9 are calculated and the corresponding limit confidence intervals appear in error bars in each figure. During experiments, temperature varied on intervals $L_{tbed}=\pm$ 4 K, for fluid bed and $L_{tfreeboard}=\pm$ 10 K, for freeboard. Concentration values are given in ppm and convert to 6% O_2 in all case. After completing one steady-state measurement air flow was changed to another value, a time delay corresponding to fifteen to thirty minutes was necessary for the random variable to reach a new steady-state and the sampling was renewed.

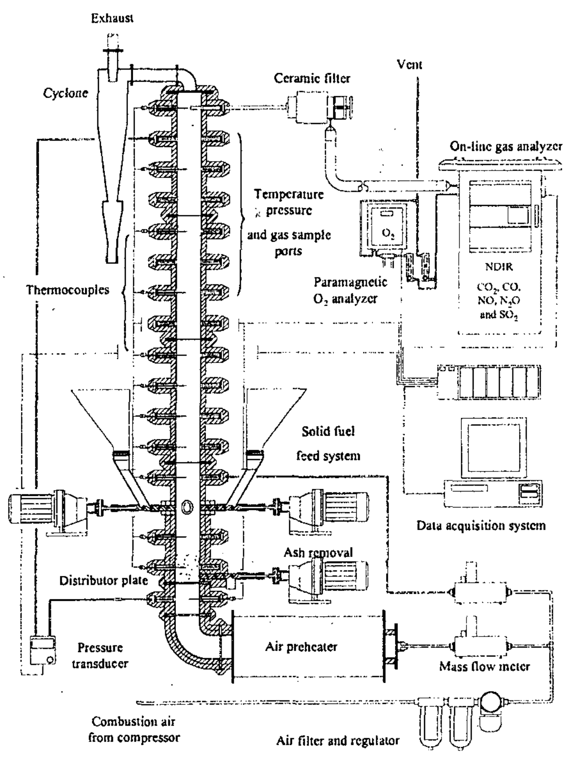

Fig. 1. Schematics of the fluidized bed reactor and measurement system.

Normally the combustion efficiency is calculated from the following equation:

$$\eta_{CE} = \frac{\%CO_2 \ in\ flue\ gas \times 100}{(\%CO_2 + \%CO)\ in\ flue\ gas} \qquad (1)$$

This efficiency calculation procedure is based on the knowledge of flue gas composition only and assumes that there are no carbon losses and that carbon present in the feed is completely converted to carbon monoxide and carbon dioxide.

In fluidized bed combustion, unburnt lignite could be elutriated. More accurate combustion efficiency is calculated, taking into account all of the pollutants present in the flue gas [4],

$$\eta'_{CE} = (B/C) \times 100 \qquad (2)$$

where B and C are, respectively, the mass fractions of burnt carbon and total carbon in the fuel.

3. Results and Discussion

3.1 Influence of excess air and fuel particle size

Combustion efficiency

To study the influences of excess air (EA) and fuel particle size on combustion characteristics, two sizes of lignite, 0.2-1 mm and 1-5 mm, were used. The lignite feed rate was kept constant at 975 g/h. Excess air was varied from 20 to 100 %, with no secondary air. Fig. 2 shows the effects of fuel particle size on carbon combustion efficiencies calculated from equations (1) and (2), being η_{CE} and η'_{CE} respectively, as a function of excess air. η_{CE} ranges from 98-99.9 %, whereas η'_{CE} shows lower values in the range of 76 and 97 %. The fact that η_{CE} is higher that η'_{CE} is not unexpected. As explained by Cliffe and Patumsawad [5], some carbon losses occur due to mechanical elutriation of fuel particles in the combustor, leading to lower η'_{CE}. This suggestion can be reinforced when it is noted that the gap between the two combustion efficiencies widens as

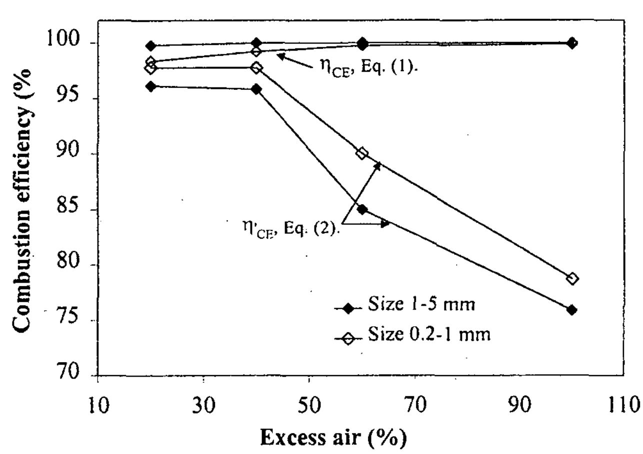

Fig. 2. Effect of fuel particle size and excess air on combustion efficiency

excess-air (and hence combustion air velocity) increases, giving rise to more elutriation.

The two types of efficiency also exhibit opposing tendencies under the influence of excess air. As excess air increase, η_{CE} tends to increase, which is consistent with the decrease in CO as seen in Fig. 3 and the expected increase in the CO to CO_2 conversion rate. η'_{CE}, on the other hand, remains high for low excess air (EA=20-40%) but drops sharply as excess air is increased further, the overall drop being about 20%. This suggests that at low excess air, the carbon-to-CO conversion rate is greater than the particle elutriation rate. Beyond a certain EA value, the air velocity, and hence fluid drag, is high enough to cause the elutriation rate to become more significant than the carbon-to-CO conversion rate.

The carbon combustion efficiency, η'_{CE}, for small particles is always higher than that for coarse particles. This is because fine particles expose a larger specific area (surface area/volume) than coarse particles, and therefore burn with higher reactivity [6]. However because fine particles a have shorter resident time in the fluid bed, the CO-to-CO_2 conversion efficiency (η_{CE}) is lower than for coarse particles (see Fig. 2).

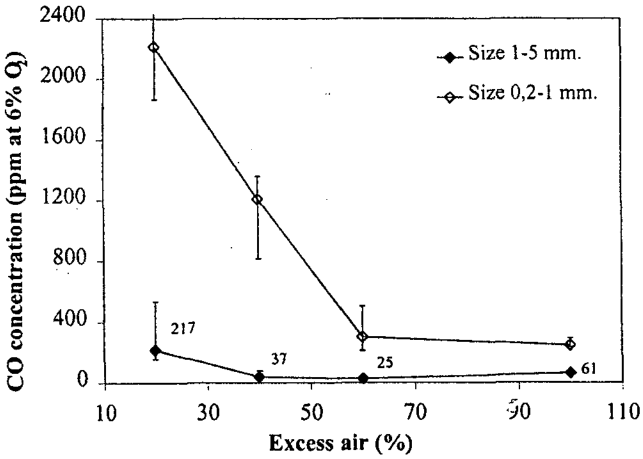

Fig. 3. Effect of fuel particle size and excess air on CO emission.

Combustion regime

Fig. 4 shows the axial distributions of mean (time-averaged) temperature for two different excess air levels, 20 and 100%, and the two mean particle sizes. In all cases the temperatures in the fluid bed are higher than those in the freeboard. However the in-bed temperatures with 20% EA are considerably higher than those with 100% EA for the same particle size; and in the transition zone between the fluid bed and the freeboard the temperature with 20% EA drops much more rapidly than the case of 100% EA, so that the freeboard temperatures for the latter case become higher. This implies that, at low EA, most of the fuel particles burn in the bed. At high EA, on the other hand, the combustion takes place both in the bed and in the freeboard. It should also be noted that the in-bed temperatures for fine particles are lower than those for coarse ones, while the reverse is true for the freeboard temperatures. The phenomena can be explained as follows:

(1) A large particle has a longer resident time in the bed and gas diffusion plays a dominant role in delaying gas evolution from the fuel during devolatilisation, so that most of the fuel burns in the bed.
(2) When burning fine particles both short resident time in the bed and chemical reaction kinetics play a role, leading to incompletely burnt gas in the bed, which continues to burn when passing into the freeboard.

Gas concentrations

Fig. 5 shows the evolution of mean concentration values for the various gaseous species as a function of excess air. NO is seen to increase with excess air for both particle sizes. This is because low oxygen concentrations generate more reducing gases such as H_2, CO, NH_3 and CH_4 that enhance the reduction of NO [7]. It is also seen that coarse lignite produces higher concentration of NO than fine lignite. Bibbs and Hedley [8] suggest that higher bed carbon (or char) concentration leads to enhanced NO reduction, but in case of high volatile content fuel such as lignite, the primary agent responsible for the reduction of NO is volatile matter [9]. Since fine particles have higher reactivity than coarse ones, due to larger specific surface area and faster particle heat-up, their rate of release of volatile matter is expected to be higher and hence greater NO reduction.

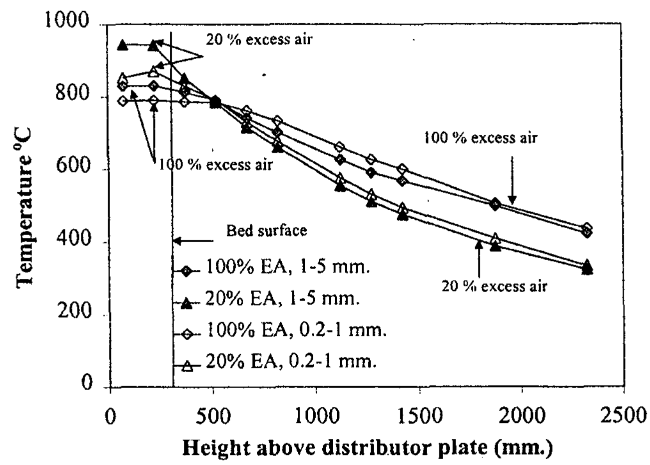

Fig. 4 Temperature profiles along the bed height.

For both particle sizes, N_2O concentration decreases with increasing excess air in the low excess-air region (20 <EA<40%) but increases in the high excess-air region. Amand et al. [10] suggest that the key formation reactions of N_2O in fluidized bed are oxidizing reactions that depend on oxygen concentration. They conclude that N_2O must increase with oxygen concentration. This is in agreement with the findings of Tullin et al. [11], which explains the increasing trend of N_2O in the high excess-air region. In the low excess-air region, N_2O has to compete with CO for OH and H radicals. Since CO concentration is more significant in the low excess-air region, particularly for fine particles, it is believed that N_2O reduction is hindered by the presence of CO.

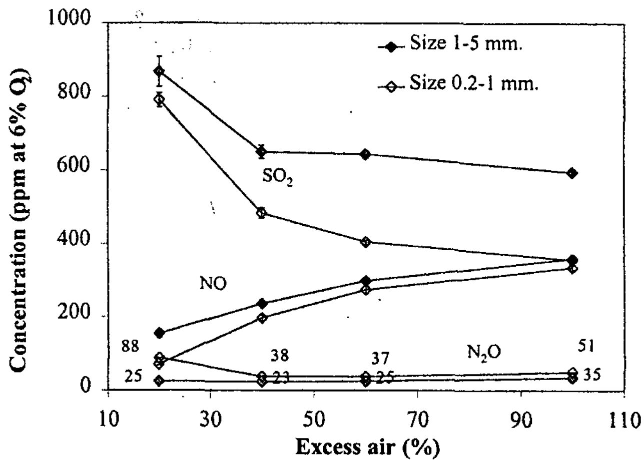

Fig. 5. Effect of fuel particle size and excess air on gas emissions

Emission of SO_2 is distinctively high at low excess air. This could be explained by the fact that SO_2 formation is a result of oxidation of H_2S, which is formed from partial combustion and pyrolysis of fuel in the reducing zone [13]. At low excess air, a reducing environment is more likely to be created in the fluid bed than at high excess-air, thus favoring the formation of H_2S. Although it may be argued that there is less O_2 available to oxidize H_2S in this case, it is also less likely that CaS, which is formed by the reaction of H_2S in the reducing zone with CaO present in the ash, would be oxidized to $CaSO_4$.

Considerably higher concentration of SO_2 is generated by the burning of coarse lignite than fine lignite. This has to do with the difference in the capacity for sulphur capture by ash particles of different size. As shown by the experiments of Aetos et. al. [3], depending on the amount of calcium (CaO) present in the coal ash, the ash particles are able to retain up to 50% of SO_2 formed during combustion. However the process is diffusion limited by a $CaSO_4$ product layer formation around the ash particles. Therefore the ash particles resulting from the burning of finer lignite, with more exposed surface area than larger particles, would be more effective in capturing SO_2.

It is generally accepted that the presence of SO_2 has a significant influence on the formation and destruction of other pollutants such as NO and CO, through its catalytic effect on radical recombination reactions, irrespective of limestone addition or coal type [15-17]. In particular, higher concentration of SO_2 leads to an increase in CO and a decrease in NO emissions. This result is in agreement with the present work, as can be seen from the comparison of the concentrations of the three species in Figs 3 and 5.

3.2 Effects of Air Staging

It has long been established that air staging in fluidized bed combustion offers the benefit of a large reduction zone (fuel-rich) for the control of NO formation, and SO_2 emission may also be decreased if in-bed staging is employed with the aid of limestone [13].

In our experiments, the effects of freeboard and in-bed air staging were examined by dividing the combustion air into primary air, introduced at the bottom of the bed, and secondary air, which was injected by means of a 6-mm-dia. stainless tube located below and above bed surface (i.e. 225 or 525 mm above the distributor plate). The primary and secondary air flow rates were varied to produce reducing or near reducing conditions in the fluid bed. The percentages of theoretical air provided to the lower portion of the fluid bed were 78, 91, 104, 117 and 130 % for in-bed staging, which was achieved by varying the secondary/total ratio (S/T) from 0 to 0.4 at 0.1 intervals. For freeboard staging, the corresponding values were 70, 84, 98, 112, 126 and 140 %. Excess air (primary plus secondary) was kept constant at 30 % for in-bed staging and 40 % for freeboard staging. The particle size of lignite used was 1-5 mm.

Combustion efficiency

The results show that the combustion efficiency η_{CE} is unaffected by air staging, while η'_{CE} is dependent on both the type and the degree of air staging (Figs 6 and 7). For in-bed staging, η'_{CE} decreases from 95% to 91% when S/T is varied from 0 to 0.4, as shown in Fig. 6. Gibbs et al. [18] suggest that at lower staged position, any combustible matter injected into the freeboard enters the secondary air mixing zone and is conveyed out of the combustor. This implies that there are high carry-over losses, which coincide with the difference already noticed between η_{CE} and η'_{CE} for non-staged air distributions. The evidence that this phenomenon is occurring to some extent is that lower temperatures are reached in the freeboard, when using S/T=0.4 as compared to the case where S/T=0.1 (see Fig. 8.). This means that the reaction in the freeboard for S/T=0.4 is less significant than that observed for S/T=0.1.

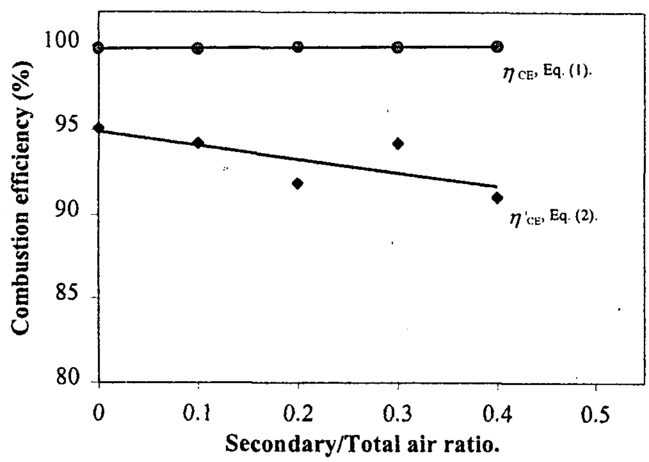

Fig. 6. Effect of secondary/total air ratio on combustion efficiency with in-bed staging

For freeboard staging, η'_{CE} increases slightly from 95% for S/T below 0.2, to a maximum of 98% at S/T around 0.35, before dropping off on further increase of secondary air. The increase of combustion efficiency in this case is probably the result of partial combustion in the sub-stoichiometric zone of the dense bed, following post oxidation in the freeboard with secondary air It can be seen in Fig. 9 that the temperature for S/T = 0.4 is higher than that for S/T = 0.1. This positive difference in the two conditions remains, up to the end of the reactor at H = 2400 mm, indicating that some of the reactions proceeded in the freeboard. However, excessively high S/T ratio severely affects combustion efficiency because there is insufficient air in the dense bed.

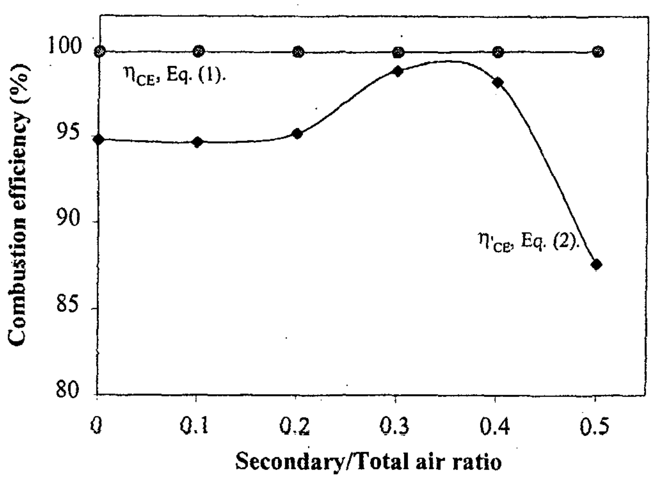

Fig. 7. Effect of secondary/total air ratio on combustion efficiency with freeboard staging

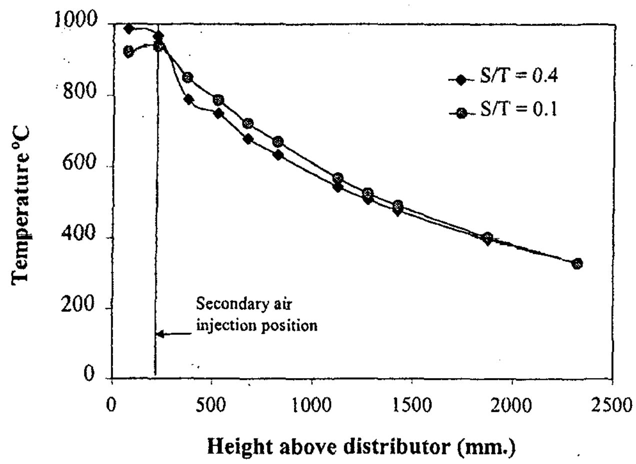

Fig. 8. Effect of secondary/total air ratio on temperature profiles with in-bed staging

CO concentration in the flue gas oscillates with air staging, but a general reduction of CO from the non-staged condition (S/T=0) is observed for both in-bed and freeboard staging, as shown in Figs. 10 and 11. This agrees with the results presented by Gibbs et al. [19]. The maximum CO reduction for in-bed staging is 90 % (249 to 14 ppm) and 60% (93 to 34 ppm) for freeboard staging both at S/T = 0.4. This is probably because for high secondary air ratio in in-bed staging, more elutriation losses of unburned carbon leads to a reduction in CO emission; and for freeboard staging, it is the result of post oxidation of CO to CO_2.

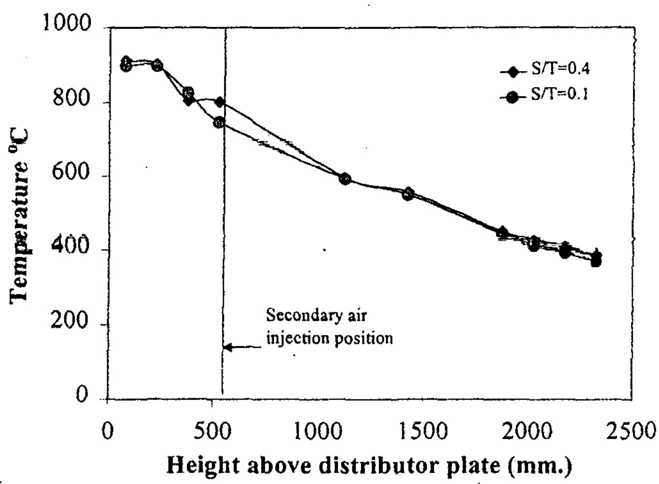

Fig. 9. Effect of secondary/total air ratio on temperature profiles with freeboard staging

Emission of NO and N_2O

The emissions of NO for both in-bed and freeboard air staging are lower than those obtained for unstaged combustion: a maximum reduction of 18% (177 to 145 ppm) for in-bed staging and 45 % (239 to 133 ppm) for freeboard staging, both at S/T=0.3 and 91 and 98% of theoretical air respectively. It is noted that the maximum decrease in NO occurs near the stoichiometric condition on the fuel-rich side. When using in-bed air staging, NO is mainly reduced by heterogeneous reactions with carbonaceous materials within a limited reducing zone in the lower portion of the fluid bed, where the resident time for lignite particles is limited. In the freeboard the reducing region extends, allowing longer resident time for particles in the reducing zone and more NO abatement [18]. The increase in NO beyond S/T=0.3, both in freeboard and in-bed staging, is probably due to the decrease of fluidization velocity, leading to a reduction of turbulence in the bed that hinders the mixing of NO with the reducing species.

N_2O emission decreases with secondary/total air ratio in all experiments. A 50% decrease in the registered values are observed for in-bed staging at S/T=0.4 and up to 87% for freeboard staging at the same S/T ratio. This is the result of the oxidization dependence of N_2O formation explained previously. It is also seen that freeboard staging has greater advantage over in-bed staging for N_2O control. Since in-bed staging leads to a reducing zone smaller than for the case of freeboard staging, more N_2O is thus produced in the former case. In any case the magnitudes involved are very small.

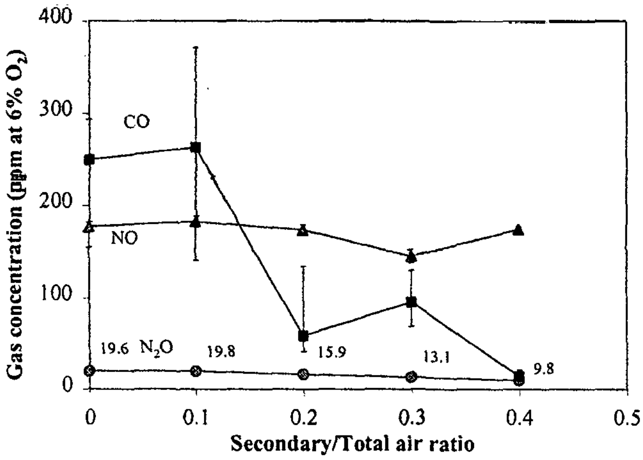

Fig. 10. Effect of secondary/total air ratio on gas emissions with in-bed staging

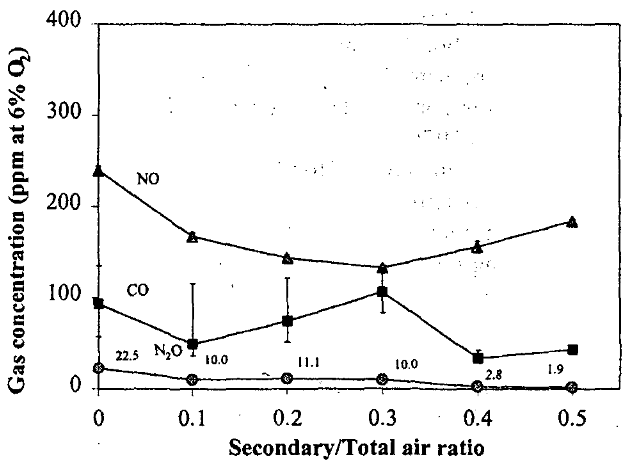

Fig. 11. Effect of secondary/total air ratio on gas emissions with freeboard staging

Emission of SO_2

Fig. 12 shows the different behavior in SO_2 emission for in-bed and freeboard staging. With in-bed staging, the secondary/total air ratio has little effect on SO_2 emission, which remains around 600 ppm. This can be explained by considering the formation of SO_2 and sulphur capture in the reducing zone. Although the decrease of oxygen enhances SO_2 formation as pointed out previously, the sulphur capture increases when in-bed staging is used due to effective sorbent (ash)-gas contact. On the contrary, a significant increase and decrease in the level of SO_2 is noted in case of freeboard staging. A steep increase in SO_2 formation is observed when increasing the secondary/total ratio near stoichiometric condition (corresponding to S/T=0.3). This is the result of a better H_2S formation, in the more fuel-rich zone, which enhances SO_2 formation that coincides with less effective sulphur capture but higher oxidation of H_2S by secondary air in the freeboard. This is in agreement with previous experiments in the case of non-staged combustion.

With a high degree of air staging in the freeboard, a large reducing zone would result in the fluid bed. Although the formation of H_2S would be enhanced, defluidizedtion would occur as evidenced by the drop in temperature in the upper part of the fluid bed (at 225 mm above the distributor) (see Fig. 13), so that oxidation of H_2S to SO_2 in the bed is constrained. In the freeboard, the injection of a large amount of secondary air at ambient temperature also decreases the oxidation rate. However the reduction in SO_2 is obtained at the expense of lower combustion efficiency, and may result in increased H_2S emission.

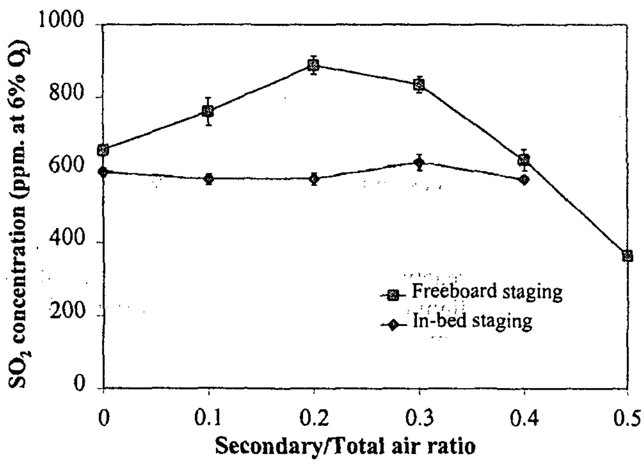

Fig. 12. Effect of secondary/total air ratio on SO_2 emissions

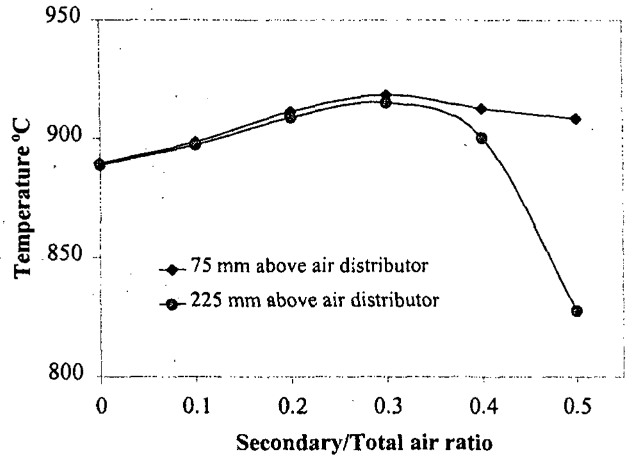

Fig. 13. Effect of secondary/total air ratio on in-bed and top of reactor temperature

4. Conclusion

The experimental investigations on the burning of Thai lignite in a laboratory fluidized bed reactor lead to the following conclusions:

Influence of excess air and particle size

1. The carbon combustion efficiency is strongly influenced by excess air with about 20% drop when the latter is varied from 20 to 100%, due

largely to elutriation loss of carbon particles. Smaller sized lignite particles yield higher combustion efficiency, but at the expense of much higher CO emission.

2. The effect of excess air on the combustion regime of lignite is such that, at low excess air most of the fuel burns in the fluid bed, while at high excess air, combustion takes place both in the fluid bed and in the freeboard.
3 The reduction of NO by decreasing excess air can be significant; and fine fuel particles generate less NO than coarse particles (40%<EA <60%).
4. There appears to be an optimum excess air range for the control of N_2O; and large lignite particles generate slightly lower concentration of N_2O than smaller particles.
5. Significant SO_2 reduction can be achieved by increasing excess air, but only up to about 50% excess air. Considerably lower SO_2 emission is seen for the burning of smaller lignite particles than their larger counterpart despite their higher reactivity, indicating the significant role of lignite ash particles as a sorbent for sulphur capture.

Influence of air staging

1. Carbon combustion efficiency decreases slightly (up to 4%) when in-bed air staging is applied, but significant CO reduction can be achieved as a trade off. On the other hand, a maximum of 98% efficiency--up from 95% under non-staged condition--is possible within a narrow range of secondary/total air ratio for freeboard staging, with CO reduction being relatively insignificant.
2. Both in-bed and freeboard air staging result in a reduction in NO, with the latter technique being much more effective by providing up to 45% reduction at about 30% air staging.
3. N_2O generally decreases with increasing secondary air for both in-bed and freeboard air staging, with the latter being more effective.
4. SO_2 emission is relatively unaffected by in-bed air staging. With freeboard staging, the ability for in-bed sulphur retention is adversely affected in the range of excess air where the carbon combustion efficiency tends to be optimized. Therefore limestone addition into the fluid bed cannot be avoided for further abatement of SO_2 emission.

5. Acknowledgements

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