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Time Series of Combustion Characteristics and Particulate Emission during Combustion of Thai Lignite in a Fixed Bed

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Abstract: The combustion characteristics and particulate emission during combustion of Thai lignite with 30% of secondary air to total air (SA:TA) in a fixed bed combustor have been investigated in real-time. The results have shown that particle formation is governed by competing reaction between the formation of the nucleated sized-particles (D_p < 0.1 μm) and the coagulated particle (D_p 0.1-1 μm). Temperature and burning rate are the highest priority factors to control the emission of particulate. Furthermore, the co-firing of coal/rice husks at 60:40% mass fraction with 10%SA:TA could be the alternative options to further reduction of particulate and to be recommended.

Key words: Combustion characteristics, particulate, Thai lignite, combustion, fixed bed.

1. Introduction

The most important solid fuel used in Thailand for energy production is Thai lignite. However, it is necessary to verify whether such a use for energy production is indeed environmentally friendly (low emission), especially in terms of PM, which may affect the atmosphere, health and internal-parts of the combustion system. It is known that such problems are mostly related to submicron particles (particle having diameter size below 1 μm; PM_1). In fact, characterization in real-time of particulate emission/formation from combustion of Thai lignite has not been investigated in Thailand yet. This study has, therefore, been employed in order to describe on-line the correlation between combustion characteristics (e.g. bed temperature, burning rate) versus particulate emission/formation, as a function of time. The highest expectation of this study is to use such data as a guideline to optimize the operating conditions to get the high temperature obtained and to reduce particulate concomitantly, based upon the combustion source, without requiring expensive PM removal devices.

2. Material and Methods

2.1 Fuel

Thai lignite was supplied by the Electricity Generating Authority of Thailand (EGAT), and originated from its Mae Moh Mine in Northern Thailand. Lignite was crushed and sieved to 3-5 mm in diameter, as shown in Fig. 1.

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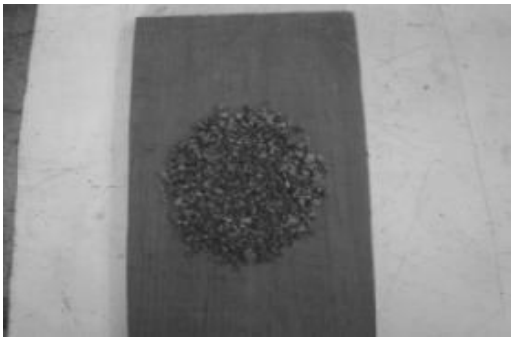

Fig. 1 Thai lignite.

2.2 Test Rig

The experimental was achieved by using a lab-scale fixed bed combustor and system, which schematically typified by Fig. 2. The combustor is a vertical cylindrical combustor of 12 cm internal diameter, 280 cm total height, isolating with 4.5 cm of refractory material and 2 cm of rock wool. The grate is located at the bottom of the combustor and consists of a perforated plate made from stainless steel, with approximately 30 orifice holes of 3 mm each. Eleven thermocouples (types K chromel-alumel) were used to detect the temperature along the reactor. All thermocouples were connected with a 40 channel Data Acquisition System; (Yokogawa, MV200) to display the temperature data. Six thermocouples (T1-T6, equidistant: 14 cm each of working distance) were used to monitor the temperature in bed, when they were plugged into the combustion zone (84 cm from the grate) and the left five thermocouples (T7-T11) were used in freeboard to monitor the temperature of

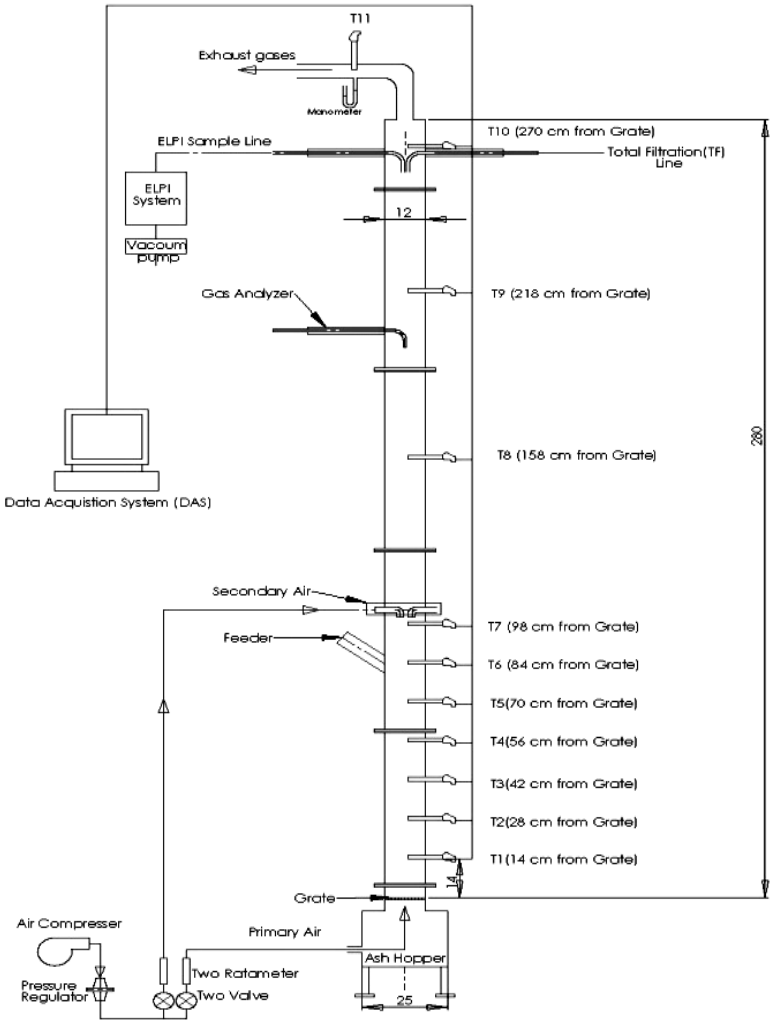

Fig. 2 Fixed bed system and sampling lines.

the flue gases. The clean air is divided into two paths, as primary and secondary air. Primary air enters the reactor underneath the fuel bed through the orifice hole of the grate, while secondary air is introduced at the top of the fuel bed (i.e. 14 cm above the top bed). The primary and secondary airflow were regulated by two rotameters. The sampling location of the ELPI line was located at about 270 cm from the grate and the sampling line is as short as possible to avoid the particle loss on the surface of sampling probe.

2.3 Specific Air Supply

In this study, Total Airflow (TA) rate is specifically designed at 0.005 m^3/sec (acms; actual cubic meter per second), together with air partitioning at 10 and 30% of secondary air to total air (SA:TA). By implication, 30% SA:TA defines as combustion under condition of primary airflow rate: 0.0035 m^3/sec and secondary airflow rate: 0.0015 m^3/sec.

2.4 Particulate Monitoring

Thirteen-stage ELPI (Electrical Low-Pressure Impactor) was used to monitoring particulate in real-time. The principle of ELPI step-by-step will be explained in the three steps that follow: (1) the sample are charged electrically by small ions, produced in a corona charger; (2) the charged particles are segregated by size, by thirteen successive stages of impactor, corresponding to their aerodynamic size (i.e. from the biggest size; 10 μm (13th stage-on top) to the smallest size; 0.04 μm (1st stage-on bottom)); and (3) the charged particles, collected in a specific impactor stage, produce an electrical current, which is recorded by the respective electrometer channel. The current value of each channel is proportional to the number of particle collected, and thus to the particle concentration. Apart from particulate, the emissions of the major gases were also monitoring and the burning rate could be calculated from the flue gas data. The detail will be described in Appendix.

3. Results and Discussion

For emissions of particle concentrations in each size, only the obvious trend of submicrometer sized-particles (PM_1) will be chosen to display. For example, D_p 0.07 μm ($PM_{0.1}$) is the representative of nucleate sized-particles (fresh particle; ultrafine particle), while D_p 0.2 and 0.48 μm (i.e. $PM_{0.1\text{-}1}$; particle having a diameter size between 0.1-1 μm) will represent the emission of the agglomerated particles. For emission of the gas, carbon mass loss as CO_2 (in mass basis) will mainly be represented as a burning rate as a function of time.

3.1 Characteristics of Temperature

Temperature histories during the combustion of Thai lignite with 30%SA:TA (Fig. 3A) can be stated as follows: the time lag for the flame front to reach the bottom of the bed is about 104 minute (at 6280 sec.). After about 155 minute (at 9330 sec.), the top thermocouple (T6) begins to drop off, suggesting that the fuel bed height has decreased, and/or the top of the fuel bed is starting to burn out. Two features are obvious for the inside-bed temperature profile, which are similar to Ref. [1]. One is the sharp drop in local temperature from the peak level, after the flame front passed by. An explanation for this is the lower-bed surface temperature, as the result of hindered char burning on the top bed, so that the thermocouples were exposed to the cooler environment, as the bed collapses below the measuring points. The second is the rise in bed temperature after about 146 minutes (8800 sec), at all measuring points, when combustion has proceeded for a significant time-period. This latter example can be regarded as the second combustion stage, where both moisture and volatile matter inside the bed solids have been completely driven out, and only char burning and coked-carbon are took place, enhanced by the full oxygen supply. This can be attributed to a change in stoichiometry, generated by

the small amount of fuel left, in which a secondary reaction originated due to oxidation of the char [2]. This caused an increase in the bed temperature, in which the maximum bed temperature obtained is about 1200 ^{o}C. Apart from the two features of the temperature profile, it also can be acknowledged that the flame front was very thin, as indicated by the very steep temperature increase at each inside-bed measuring point, i.e. T5, T4, T3, T2, and T1. This indicates that there is still a substantial layer of solid materials remaining, after ignition, and that is why there is a burnout process at the end of the experiment. It should be borne in mind that a plateau at a temperature that is representative of the combustion products follows (i.e. particulate).

3.2 Characteristics of Oxygen in Flue Gas

Oxygen detected in flue gas is also important as indicator of sufficient or insufficient status of oxygen in a fuel bed (See Fig. 3B). The details of the oxygen profile can be discussed as follows: the oxygen content does not drop below 5.5 ± 1.6% at which t = 100-2600 second (or at first 41 minute of entire time), according to the burning of the top bed to the lower bed, i.e. at 560-840 mm from grate; T6→T4. This feature indicates that there is insufficient oxygen in the bed for combustion. However, after the flame front reached the deeper bed (T4→T1), the oxygen concentration dropped to about 2.4 ± 0.89%. In the last stage (i.e. flame front moves from T1→grate), according to the time interval: 6620-8325 sec, the oxygen content fell to about 0.6-1.8%. These figures indicate that in the lower bed, there is more oxygen.

3.3 Correlation between Temperature Profile and Oxygen in the Flue Gas

Referring to Fig. 3 (A vs. B), the significant connection between bed temperature and oxygen concentration in flue gas can be discussed, as follows. At the warm-up phase, especially at t = 0-1100 sec, a visible decrease in slope can be observed from the temperature history (T6 reference). This slower temperature rise is characteristic of strong endothermic degradation of the fuel. The temperature at which the change in slope occurs (~600 ^{o}C) is the characteristic of the pyrolysis step. This particular temperature history corresponds to an equivalence ratio of less than one ($e_1 < 1$); e_1 is excess oxygen in bed, indicating that the oxygen does not drop below 4 percent. Therefore, the partial gasification of the fuelis expected to occur under a low oxygen concentration (in bed). This lack of oxygen concentration in the bed tends to favor endothermic pyrolysis pathways, and thus results in a decrease in the slope of temperature histories at the warm-up stage. When the flame front falls to at 560 mm from the grate (T4; thermocouple no. 4) or at t = 2660 sec, gasification still occurs but it is characterized by an oxygen richer environment in bed, as indicated by the fall in oxygen concentration to about 2.4%, and combustion occurring in the gas phase. This results in a temperature peak (1000 ^{o}C) that occurs at approximately 3000 sec. This peak corresponds to a narrow reaction zone, occurring above the fuel. All aforementioned features at the T4 location are also clearly seen at T3, T2, and T1 locations.

3.4 Relationship between Oxygen Concentration and Formation of CO_2

From Fig. 3 (B vs. C), it may be observed that the decrease in O_2 concentration is caused by a rise in CO_2 concentration, or defines as improvement of burning rate and combustion is marked by the dominance of exothermic reaction. It is important to further note that after oxygen depletion, there is still some solid material left, which might be partially consumed by gasification of H_2O and CO_2. Gasification causes a bed temperature decrease to mean values about 800 ^{o}C, at which these reactions proceed slowly. This feature is really seen (see behind dot line in Fig. 3) at time intervals: 3000-8700 sec. i.e. sees T4, T3, T2 and T1 for the relationship between

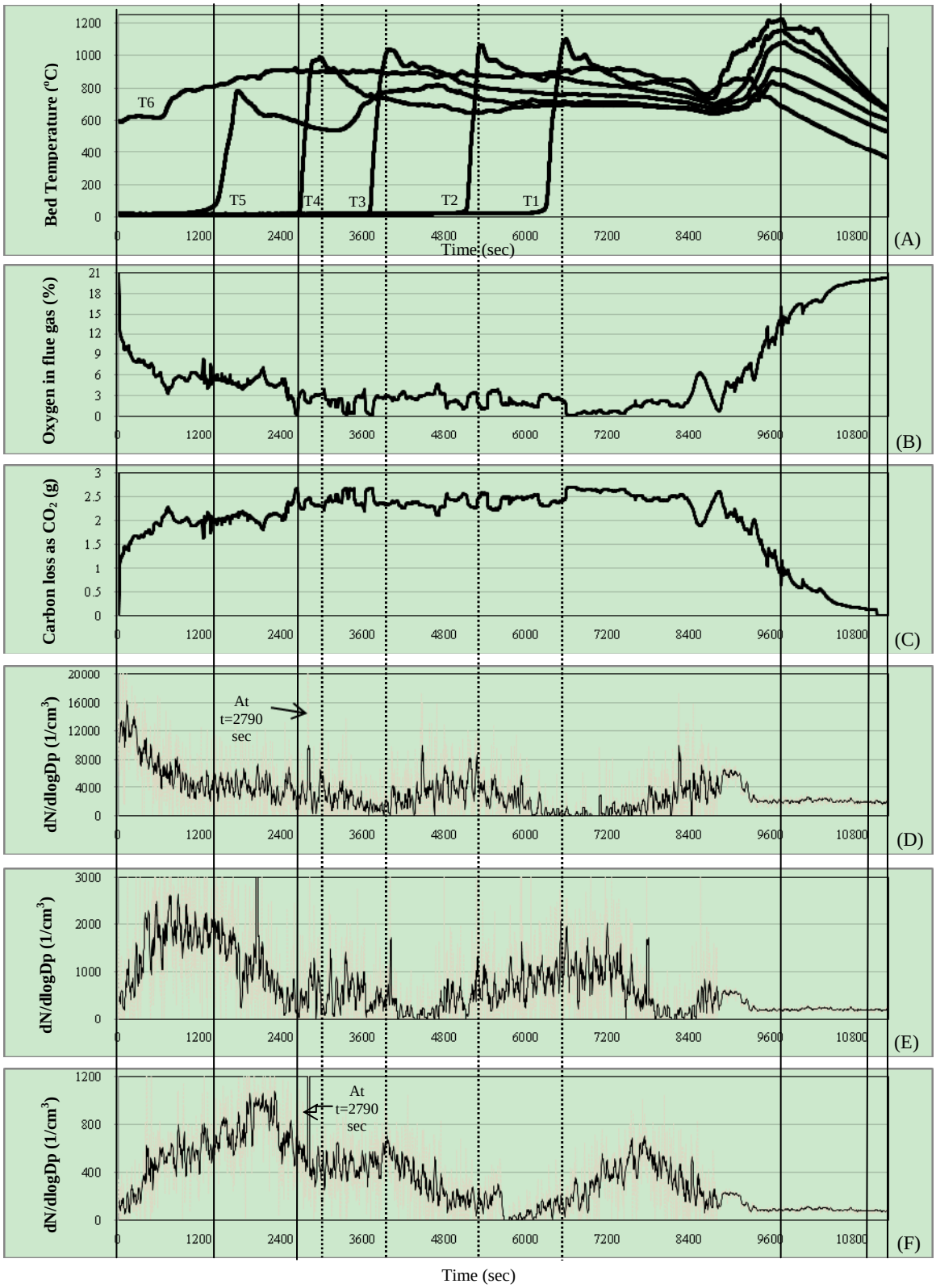

Fig. 3 Real-time correlation of operating conditions and number concentration of particles, during the combustion of Thai lignite, with 30%SA:TA; (A) Bed temperature; (B) Oxygen in flue gas; (C) Burning rate as CO_2 released in mass-basis; (D) D_p 0.07 µm; (E) D_p 0.20 µm; (F) D_p 0.48 µm.

CO_2, O_2 and bed temperatures.

3.5 Relationship between Operating Conditions and Particulate Emission

For particulate, the significant observation of their emission and connection with operating conditions can be made, as follows. At the warm-up stage at t = 0-1200 sec, the bed temperature (T6) is below 600 ^oC and bed TEMP tends to increase slowly (because of a low burning rate; CO_2 formation is low), the level of nucleate sized-particles or primary particles (0.07 μmin diameter size) are high for a moment. Afterwards, when temperatures rise above 600 ^oC along with a higher burning rate, this smallest size particle (0.07 μm) tends to decrease apparently, while amount of the accumulated particles increases; D_p 0.2 and 0.48 μm, are prevalent instead. This simply indicates that increasing of the bed temperature results in an increase of particle fusion or thermal coagulation, and formation of particle is a competing reaction, in which temperature is the main factor. Moreover, it could be said that the supply rate of secondary air (0.0015 m^3/sec) is a good compromise to mixing with the flue gas, and further oxidation in gas-phase. This means remaining gases above the bed are low, so that the chance of those vapors/gases converting to the smallest size of particle through condensation process is low. Once the flame front approach to T5 location and after peak temperature occurs (see a plateau), it is observed that there is a rise of agglomerated particles, especially those having the size: 0.48 and/or 0.2 μm. At the same time, the emission of the smaller size of emitted particles: 0.07 μm are kept low. The corresponding factors that play an important role in this increase of agglomerated particles are the decrease in burning rate (as seen from low CO_2 formation), due to low oxygen available in the bed, because of the low bed temperature in this time (< 600 ^oC). So now, it could be summarized that the limited interaction of oxidizer/fuel surface, due to insufficient oxygen in the bed at the time, will result in a limited conversion of fuel-C to CO_2 or CO to CO_2. This indicates a low burning rate, so the rise of temperature from exothermic reaction will be limited as well. This finally leads to incomplete combustion and thus an increase in PM emitted. Another point of view of the high formation of the ultrafine (D_p 0.07 μm) and the coagulated particles (D_p 0.48 μm) can be observed at t = 2790 sec. (i.e. during the rising of bed temperature at T4 location), this indicates that increasing of bed temperature results in more pronounced ash volatilization and the subsequent condensation of them [3-4]. At the intermediate phase (flame propagated from T4 to the grate), and the char-burnout phase, both phases represent the most steady state and the greater good in higher bed temperatures and/or in a burning rate, compared to both parameters in the warm-up phase.

Due to time spent in each location inside the bed is not equal, so that analysis will be based on flame propagation, from one place to another (i.e. from T6 to T5, T5 to T4, unitlsT1-grate). In this way, the number of particle emitted at each level of the fuel bed can be summarized in Table 1.

The key point in Table 1 is that the total number of particles tends to reduce, apparently as a function of fuel bed height, in which the average of total number are 7.4×10^9, 3.63×10^9 and 3.0×10^9 at warm-up phase, intermediate phase (steady state) and char burnout phase, respectively. Above all, the highest priority factors influencing this trend are the difference in temperature and burning rate for each phase; higher bed temperature and/or burning rates results in more complete combustion, and, thus, low emission of particulates, which can clearly be seen at the intermediate phase (vs. warm-up phase). Note that during char burnout phase, the burning rate (as CO_2 formation) is rather low (vs. warm-up, intermediate) due to there is so little fuel left at that time, and oxygen concentration is starting to increase steadily, so that the surface reaction of char and oxidizer might be obstructed as usual. However, the very high local

Table 1 Emission of particles in number versus operating conditions in each location of the fuel bed, during combustion of Thai lignite.

Each location in bed	Bed temp (^oC)	Gas temp (^oC)	Burning rate (g/sec)	$N(D_p)$ 0.07 μm	$N(D_p)$ 0.2 μm	$N(D_p)$ 0.48 μm	Total Number (N)
T6→T5	785	815	0.3302	6.4E + 09	1.2E + 09	3.7E + 08	8.1E + 09
T5→T4	770	910	0.3838	4.4E + 09	1.3E + 09	9.2E + 08	6.7E + 09
T4→T3	980	960	0.4449	2.1E + 09	4.2E + 08	3.9E + 08	2.9E + 09
T3→T2	1040	960	0.4332	3.0E + 09	3.2E + 08	4.0E + 08	3.7E + 09
T2→T1	1060	980	0.4416	2.3E + 09	5.9E + 08	9.8E + 07	3.0E + 09
T1→grate	1100	1000	0.4529	3.0E + 09	1.2E + 09	6.4E + 08	4.8E + 09
Char Burnout	1220	1000	0.3287	2.7E + 09	2.6E + 08	1.1E + 08	3.0E + 09

Important note: T6→T4: warm-up phase, T4→grate: intermediate phase (period of steady combustion), char burnout: final phase. Temperatures are labeled by peak temperature at each location of measuring points; T1-T6 (Bed TEMP) and T7 (Gas TEMP).

bed-gas temperature stills support the complete secondary combustion of reduced gases leaving char, so that the particles emitted still remain at a low level.

3.6 Some Implications to Further Reduction of Particulate through Co-Firing with Low SA

This example of case (coal: rice husk at 60:40% mass fraction with 10% of SA:TA) is documented as the recommended case, giving the lowest emission of particulate matter, in both terms of total number concentration and total mass concentration, in comparison with the other remaining fractions operated under 10-30% of SA:TA.

Moreover, this case could be compared to the lignite case, because they were burned at the same total airflow (i.e. TA = 0.005 m^3/sec). There are the most distinguishable phenomena in this case (10%SA:TA-basis) and lignite case operated under 30%SA:TA-basis.

Firstly, the whole combustion in case 10%SA is apparently marked by single-stage combustion; no sign of char burnout in the last stage, as found on case 30%SA (see in Fig. 3 vs. Fig. 4: bed temperature). This is caused by the adequacy of excess air in bed as a result of the reduction in the SA:TA ratio. This makes the char fraction that is supposed to burn in final char-burning-only stage, shift combustion in the bed to a more oxidizer rich environment. This also means there is thinner layer of solid materials left after the flame front passes away compared to case 30% SA:TA.

Secondly, oxygen in flue gas fell to zero level (see in Fig. 4B), during the significant time (i.e. at time = 60-2400 sec) in the whole combustion process. By implication, it means the mixing or surface reaction between carbonaceous material and oxygen (in the case of 10%SA) is optimum. In addition, that characteristic of oxygen also produces the highest stable of bed temperature and a higher burning rate; high formation and steadiness of CO_2 compared to 30%SA, that always leads to the stability of high temperatures obtained, especially in the time interval: 600-2400 sec (deeper bed = richer oxygen).

Thirdly, adding of rice husk (40%) may be considered as a part of help for more effective combustion. This is probably due to the lower ignition temperature and fuel properties as rice husk contains higher volatile matter and oxygenated compounds, which favor a progress of ignition and flame propagation.

Fourthly and it is the most important subject we are concerned with this study: the minimization of PM emissions. It can be recorded that the average of total number concentration in each location of the combustor are: 1.2×10^9 and 1.48×10^9 of particle numbers at the initial stage (T6→T4) and in a stable state until the end of combustion (T4→grate), respectively. Such ranges are much lower than those numbers retrieved by lignite case operated under 30%SA, which yielded finally that

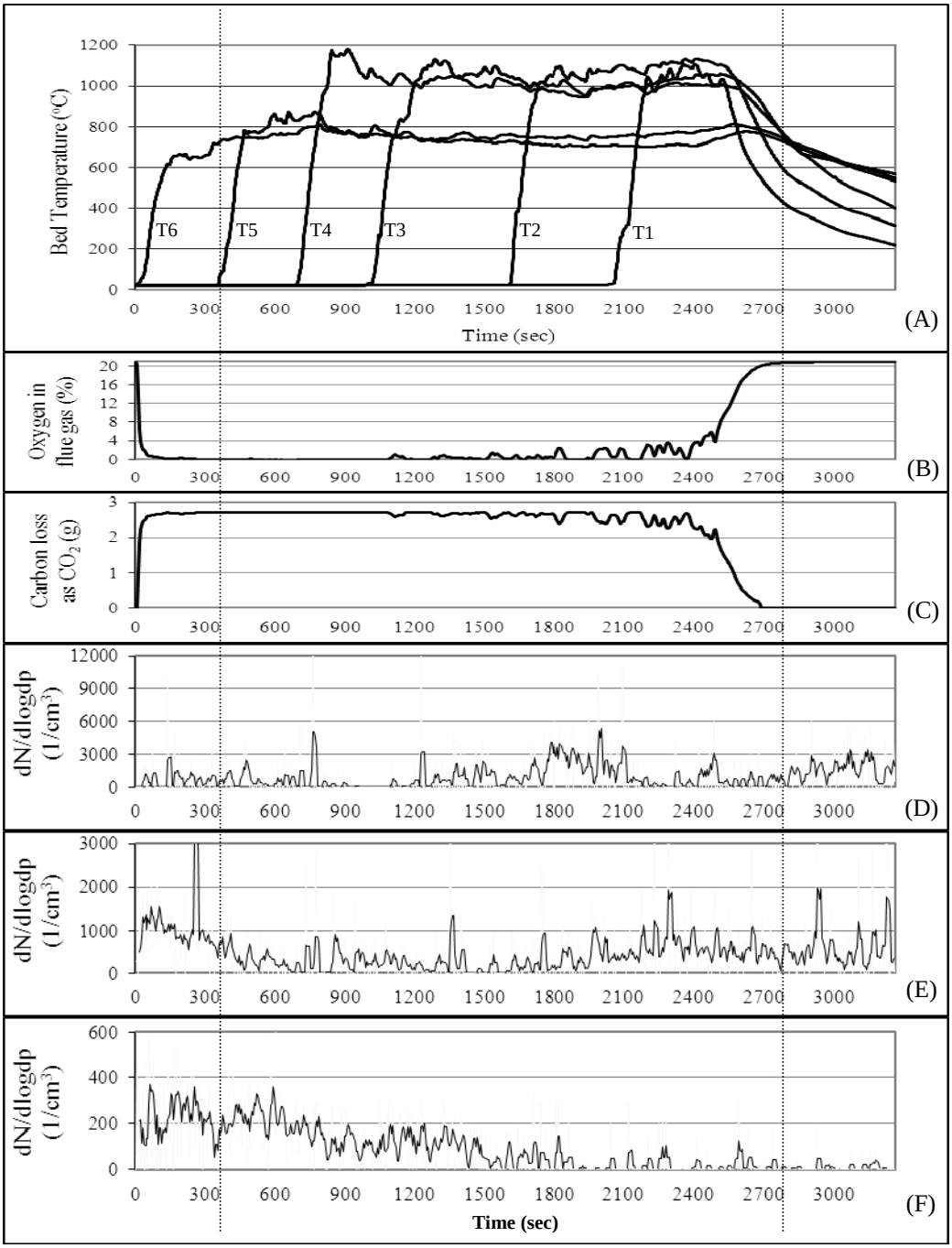

Fig. 4 Real-time correlation of operating conditions and number concentration of particles, during the co-firing of Thai lignite/rice husks (60:40), with 10%SA:TA; (A) Bed Temperature; (B) Oxygen in flue gas; (C) Burning rate as CO_2 released in mass-basis; (D) D_p 0.07 µm; (E) D_p 0.20 µm; (F) D_p 0.48 µm.

"the surplus of excess air in bed is more pronounced, than the surplus of secondary air quantity above the bed, to reduce the PM".

4. Conclusions

Real-time correlation between combustion characteristics and emission of particulate during combustion of Thai lignite has been elucidated. In addition, changing of fuel fraction and of air supply has been applied in order to observe the PM emission. The major conclusion can be documented as follow.

4.1 Combustion Alone of Thai Lignite with 30% SA:TA

Particle formation is governed by competing reaction between the formation of the nucleate sized-particles ($PM_{0.1}$) and the coagulated particle ($PM_{0.1\text{-}1}$). When the burning rate and temperature are low or rising slowly, nucleate sized-particles are dominated. In contrast, when burning rate and temperatures get higher the coagulated particles are prevalent instead and $PM_{0.1}$ will be partially suppressed. The maximum particle number formed is observed in warm-up phase. However, the greater stability in bed temperature and burning rate are always found in an intermediate and char burnout phase, result in more complete combustion and thus a low emission of total number of particulate, especially submicron particles.

4.2 The Alternative Options

Co-firing by adding rice husk in optimum mass fraction (40%) together with the reduction of secondary air to total air ratio (10%) can reduce further particulate emissions, compared to coal case.

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Appendix: Gases Monitoring and Calculation of Burning Rate

Gas emissions are monitored continuously by using Testo gas analyzer (model 350). The measuring range for continuous measurement of O_2 was 0-21%, while CO_2 concentration continuous measurement data were not available directly but were thus calculated from oxygen concentrations following equation:

$$CO_2 = \frac{CO_{2\max} \times (21\% - O_2\%)}{21\%} \quad (1)$$

When CO_2 max is fuel specific maximum CO_2 value (i.e. 18.4% for coal); 21% is oxygen content of the air in %; O_2% is the measured oxygen content in percent.

For burning rate**,** since carbon is the major composition of the fuel. The fuel degradation is therefore represented by carbon mass loss, which could be considered in terms of carbon dioxide released on a mass basis. Finding of burning rate from balancing of flue gas has been proposed as in Ref. **[**5**]**, as following equation:

$$m_{CT} - m_c(t) = MW_c x \cdot \left(\frac{1kg}{1000\,g}\right) \cdot \left(\frac{1}{100 \times 24.45}\right) \cdot Q \int_{t_i}^{t_f} [C_{co_2}(t)]dt \quad (2)$$

where m_{CT} is the total carbon mass loss in char particle (kg); $m_c(t)$ is the carbon mass in char particle at time t; MW_c is the molecular weight of carbon (12 g/gmole); Q is the volume of airflow rate (Liter/second); C_{CO_2} is the carbon dioxide content of flue gas at time t (vol.%).