Ignition behavior of single coal particle in a fluidized bed under O_2/CO_2 and O_2/N_2 atmosphere: a combination of visual image and particle temperature

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Abstract

Single coal particle ignition behavior was studied in a two-dimensional (200 mm × 20 mm × 400 mm) fluidized bed under O_2/N_2 and O_2/CO_2 atmosphere with O_2 volume concentration in the range of 10 ~ 40%, by a combination of visual observation and measurement of particle center temperature. A piece of transparent quartz glass was used as the front wall of the fluidized bed to allow visual observation. The investigated fuel particles were spherical bituminous coal particles with diameter in a range of 6~13 mm, which were artificially carved from selected original coal particles. The volatile combustion flame was recorded by a color video camera to analyze its ignition time delay and extinction behaviors. The temperature in the particle center was measured by a very thin thermocouple to detect particle heating process. Results indicate that under O_2/CO_2 atmosphere the ignition delay time is much longer than in O_2/N_2 atmosphere. The devolatilization process is almost unaffected by atmosphere under the conditions with same O_2 concentration, but controls by internal and external heat transfer. The effect of exothermic of volatiles reactions on heating and extinction time for larger coal particles can be neglected.

Keywords:
O_2/CO_2 atmosphere; fluidized bed; devolatilization; combustion behavior

Nomenclature and Units

A	m^2	surface area
c	J/(kg·K)	thermal capacity
E	J/mol	activation energy
F	kg/s	reaction rate
h	$W/(m^2\cdot K)$	Convection heat transfer coefficient
⊿H	J/kg	latent heat
R	kg/s	average evolution rate
k		pre-exponential factor
r	m	Radius
T	℃	Temperature
t	s	Time
U	m/s	fluidizing velocity

V	m^3	Volume
V^*	%	initialize volatile content
ε		emissivity of the particle
ζ		fraction of the enthalpy from combustion reaching particle
λ	W/(m·K)	thermal conductivity
ρ	kg/m^3	Density
σ	$J/(m^2·s·K^4)$	Stefan–Boltzmann constant
σ_E	J/mol	standard deviation activation energy
Subscripts		
b		fluidized bed
e		evaporation
mf		minimum
s		solid
v		volatile
vc		combustion of volatiles

1. Introduction

In the foreseeable future, coal would still be the predominant fuel in worldwide power generation. However, CO_2 emissions from coal combustion processes take up over 40% of global CO_2 emissions [1]. In view of high urgency to reduce CO_2 emissions, development of CCS (Carbon Capture and Storage) technology is promoted extensively in recent years [2],[3]. Among various carbon capture technologies, O_2/CO_2 combustion (also known as oxy-fuel combustion) is one of the most competitive technologies, where fuel is burnt with oxygen and recycled flue gas [4]. Then the generated high concentration of CO_2 (60~70 vol. %) can be compressed and transported for utilization or storage.

An extensive of studies [5]-[10]has been performed on comparisons of coal combustion characteristics under O_2/N_2 and O_2/CO_2 atmosphere. They indicate that coal combustion reaction rate is considerably reduced when N_2 is replaced by CO_2 with the same oxygen concentration. However, most of the existing studies are limited to pulverized coal (PC). As an alternative clean coal utilization technique, fluidized bed (FB) boiler has been utilized in many power plants, where the fuel combustion conditions are much different in FB and PC boiler, such as fuel particle size, furnace temperature, and flow dynamic behaviors. Currently, the O_2/CO_2 fluidized bed combustion technology is being actively developed in both academic and industry [11]-[16]. CanmetEnergy from Canada [17]-[20] reported pollutant formations and operating experiences in 100 kW_{th} and 0.8 MW_{th} pilot-plants with flue gas recycle under O_2/CO_2 atmosphere. The transition from O_2/N_2 combustion to O_2/CO_2 combustion was smooth. CO and NO_x emissions were lower than those under O_2/N_2 combustion. However, SO_2 concentration was significantly higher under O_2/CO_2 combustion. These results agree with the finding by Duan et al. [12] who conducted O_2/CO_2 combustion experiments in a 50 kW_{th} circulating fluidized bed. They found that NO emission could be drastically reduced by O_2 staging.

In view of laboratory-scale studies, Scala et al. [13],[21] presented a systematic study of char particles combustion in a lab-scale fluidized bed under O_2/CO_2 atmosphere, and found that both gasification of CO_2 with char and homogeneous CO oxidation were significant. Guedea et al. [22] conducted single coal particle devolatilization under O_2/N_2 and O_2/CO_2 atmosphere in a thermo-gravimetric oven and found that O_2/CO_2 mixtures resulted slightly longer devolatilization time.

However, regarding on single coal particle devolatilization in *fluidized bed*s under different atmospheres, there are few studies. Such information is critical for revealing combustion difference between O_2/N_2 and O_2/CO_2 combustion, and is important for oxy-fuel fluidized bed design and operation. Traditionally for studies on single coal particle combustion characteristics in fluidized beds, there are three main methods. (i) by measuring particle temperature, the devolatilization time was defined as the time when the temperature in the particle center reached to the bed temperature [23], (ii) by extracting coal particles suddenly from the reactor and quenching, the devolatilization time was defined as the time when the particle weight loss rate became less than 1% [22],[24], (iii) by optical access of the rector, the entire devolatilization and combustion process of fuel particle was observed visually. Prins et al. [25] designed a two-dimensional transparent fluidized bed for studying coal particle devolatilization and combustion and provided numerous of valuable information on coal combustion characteristics. Recently, based on visual measurements, Levendis' group [26] revealed coal devolatilizaiton and combustion mechanisms under O_2/CO_2 atmosphere, but again their studies are limited to pulverized coal.

In this work, two of the above three methods are combined, that both particle temperature measurement and visual access, to investigate the effect of CO_2 and enhanced O_2 levels on single coal devolatilization characteristics. The experiments are performed in an electrically-heated fluidized bed with a transparent front wall, meanwhile, temperature at the coal particle center is recorded continuously for analyzing devolatilization characteristics.

2. Experimental methodology

The current studies were carried out in a two-dimensional (200 mm × 20 mm × 400 mm) and electrically-heated (6.0 kW) fluidized bed (see Fig. 1). Two-dimensional fluidized beds, which can provide intuitive information of bubbles and solid mixing, have been widely used [27]-[28]. Besides, two-dimensional bed is considered to be appropriate device for the present study, as it allows the use of a single coal particle [25]. A piece of transparent quartz glass with size of 200 mm × 400 mm was fitted as the front wall of the fluidized bed. One stainless steel plate, fixed with four bubble caps as the gas distributor, ensured uniform distribution of the inlet gas. The pressure drop across the distributor was 0.26 times of the pressure drop across the bed when the static bed height was 150 mm. The furnace temperature profile was measured along the vertical axis of the furnace by four sensitive calibrated thermocouples, whose heights were 50 mm, 150 mm, 250 mm and 350 mm above the gas distributor. Thermal insulation material wrapped the fluidized bed to reduce heat loss (see Fig. 1). A self-adjusting PID controller was applied for bed temperature control. Bed materials and fuel particles were fed into the bed through a stainless-steel funnel which was located at the bed top.

Fig. 2 shows the schematic diagram of the experimental system. Three kinds of gases were supplied by different cylinders (nitrogen, carbon dioxide and oxygen) and calibrated rotameters were adopted to measure the gas flow rates. Before entering the bed, the gases were mixed in a mixing cylinder and heated to a desired temperature by an electric preheater with 1.5 kW.

A color video camera was used to record coal particle combustion processes through the quartz window at the speed of 32 frames per second. Digital videos were analyzed by slow playbacks (frame by frame) to account for fuel particle movement and combustion behaviors.

The investigated coal particle was produced by three steps, as shown in Fig. 3. (i) An original coal particle with anomalous edges was carved and resulted in a nearly spherical particle, and the particle diameter was measured repeatedly by a vernier caliper, ensuring the sphericity of the final investigated coal particles was larger than 0.9. (ii) A hole with a diameter of 0.4 mm was drilled into the particle's center. (iii) A thin and flexible, K-type thermocouple (TC Ltd.; 0.25-mm sheath diameter) was inserted into the hole and fixed with fireplace sealant which could endure temperature up to 1250℃.Scott et al.[29] and Scala [30] proved that this kind of thermocouple does not restrict free motion of coal particles in fluidized beds, and the response times is 3 ms for temperature change from 20℃ to 100℃ in boiling water. The thermocouple was wired to a data acquisition, making the transient thermocouple signal to be recorded continuously.

Silica sand (250~350 μm) with a weight of 1.3 kg was used as bed materials, giving an unfluidized depth of 150 mm. The minimum fluidizing velocity (U_{mf}) was experimentally measured to be 0.191 m/s at 850℃. All the experiments were performed at the condition of bed temperature with 850℃, and fluidizing gas velocity with 2 times of U_{mf}. In this study, bituminous coal was selected as the test coal because it has the lowest propensity of fragmentation and attrition compared with other coals [31]. The ultimate analysis of the investigated coal is listed in Table 1. The concentration of O_2 in the atmosphere with CO_2 or N_2 was set to 0, 10, 21, 25, 30, 35, 40% vol./vol., and coal particle diameter was varied from 6 to 13 mm. Each test condition was repeated at least 6 times to obtain averaged results. In each test, the single coal particle was dropped into the bed after the bed was regulated well and steady, and the test was discarded when fragmentation of the coal particle or thermocouple detached from the particle was observed.

3. Experimental results

3.1 Coal particle volatile combustion image

The zero time of video is defined as the moment when the coal particle was put into the funnel located in the bed top. It took some seconds for the coal particle landing onto the bed surface from the feed location, which was estimated to be around 0.35 s, given the vertical distance of 600 mm between the bed surface and feed location. The ignition delay time refers to the moment when volatile matter flame appeared for the first time, and the extinction time volatile matter flame disappeared [25], [32]. By playback of the video, the images of feeding, ignition, combustion and flame extinction can be determined. Fig. 4 presents these images for the coal particle (d_p = 10 mm) at the conditions with various O_2 concentrations in N_2 atmosphere. It is observed that the coal particle always floats on the bed surface during volatile combustion process. When O_2 concentration is 10%, a downstream yellow diffusion flame appears and extends over half of the bed height. When O_2 concentration is higher than 21%, a white flame appears and the flame size decreases. However, from Fig. 4, the coal particle ignition delay time and extinction time are not affected considerably as variance in O_2 concentration.

For comparison, Fig. 5 presents the determined images for the coal particle (d_p = 10 mm) at the conditions with various O_2 concentrations in CO_2 atmosphere. Compared with N_2 atmosphere, Fig. 5 indicates that the volatile combustion profile exhibits a blurred and yellow diffusion flame when the oxygen concentration is even 10%, and flame luminance increases with O_2 concentration. The ignition delay time of the coal particle under CO_2 atmosphere can be divided into two regions. When O_2 concentration is lower than 25%, the ignition delay time decreases as O_2 concentration increased. On the other hand, almost unchanged ignition delay time is observed by any further increase in oxygen concentration when O_2 concentration is higher than 25%, as shown in Fig. 5. By comparing Fig. 4 and Fig. 5, there is barely any difference of the extinction time between O_2/CO_2 and O_2/N_2 environment.

For a better comparison, Fig. 6 presents the ignition delay time and extinction time in the form of line graph. It strongly suggests the ignition delay time under O_2/CO_2 atmosphere is much longer than in O_2/N_2 atmosphere, especially when O_2 concentration in CO_2 atmosphere is lower than 25%.

Fig. 7 shows, for coal particles with different diameters (d_p = 6, 8.5, 10 and 13 mm), the typical coal particle devolatilization events under 30% O_2 concentration in O_2/CO_2 atmosphere. Both the extinction time and flame size increases with increase in particle diameter. By increasing particle diameter, the ignition delay time increases generally, however, when the particle diameter is larger than 10 mm, the ignition delay time is found to decrease slightly with further increase in particle diameter. A possible explanation is that the volatile content is much more for larger coal particles which could contribute to the larger quantities of released volatile compounds, resulting in a shorter ignition delay time.

3.2 Coal particle center temperature

Fig. 8 plots the temperature history with time for the coal particle (d_p = 10 mm) in various oxygen concentrations in N_2 atmosphere. After the particle center temperature reaching 100 ℃, it remains unchanged until the end of evaporation. As shown in Fig. 4, volatile flame extinction time is around 57.82 s. And the temperature profiles are analogous before 57.82 s with various oxygen levels, as seen in Fig. 8. It implies that the exothermic of volatiles reactions do not promote particle heating. This agrees well with Solomon [33] results, he indicated that the rate of devolatilization (pyrolysis) was controlled by both external and internal heat transfer when coal particle diameter is larger than 3 mm. After volatile extinction, particle center temperature continues to rise gradually under pyrolysis condition, and the eventual particle center temperature is still lower than bed temperature due to particle internal heat transfer resistance. For the devolatilization condition, the eventual value of center temperature increases by increasing O_2 concentration, even higher than bed temperature because char combustion attributes to higher temperature profile. For comparison, Fig. 9 plots the temperature history with time for the coal particle (d_p = 10 mm) in various oxygen concentrations in CO_2 atmosphere. Similar trends of particle center temperature are observed between O_2/CO_2 and O_2/N_2 atmosphere. Therefore, coal particle devolatilization process is almost unaffected by atmosphere, which coincides with Guedea et al. [22] results who measured mass loss during the devolatilization process under O_2/CO_2 and O_2/N_2 atmosphere.

Fig. 10 plots coal particle center temperature profiles under 30% O_2 concentration in CO_2 atmosphere with different particle diameters (d_p = 6, 8.5, 10 and 13 mm). Compared with larger size of coal particle, small one has lower moisture content and internal heat transfer resistance, which result in shorter time of drying and higher temperature rising ratio. As result of char combustion, the particle center temperature of the coal particle with d_p = 6 mm reaches nearly 1000 ℃. The ignition delay time of the coal particle with d_p = 13 mm is 2.70 s, and its particle center temperature at this moment is 59.70℃, as shown in Fig. 7 and Fig. 10, which indicates there exists a strong internal heat transfer resistance.

4. Discussion

Experimental results have shown different ignition and combustion behaviors of coal particle between O_2/N_2 and O_2/CO_2 atmosphere. In this section, a transient devolatilization model is developed to reveal these different behaviors. Generally, coal devolatilization (pyrolysis) includes particle heating, drying, devolatilization and volatile combustion, and these processes are interrelated and mutually. To make the problem analytically tractable, several assumptions have to be made in the following [34],

(i) Sphere coal particle is an isotropic material;

(ii) Change of shape of the coal particle does not happen;

(iii) Gas phase reaction is a one-step global reaction;

(iv) $C_xH_yO_z$ is considered to represent volatiles.

During coal devolatilization (pyrolysis) process, a heat balance on the single particle is,

$$\frac{\partial(\rho_s T_s)}{\partial t}=\frac{\lambda_s}{c_s}\frac{1}{r^2}\left(\frac{\partial}{\partial r}r^2\frac{\partial T_s}{\partial r}\right)+\frac{1}{c_s}\frac{R_e(-\Delta H_{latent})}{V_e}+\frac{1}{c_s}R_v\rho_s(-\Delta H_v) \tag{1}$$

Symbols are defined in the list of Nomenclature.

Conservation of coal particle mass is,

$$\frac{\partial \rho_s}{\partial t}=-R_e-R_v \tag{2}$$

Initialize condition of coal particle temperature is,

$$t = 0,\quad 0 \le r \le r_0,\quad T = T^0 \tag{3}$$

Boundary conditions at the particle surface and center are,

$$t > 0,\quad r = 0,\quad -\lambda_s \frac{\partial T_s}{\partial r} = 0 \tag{4}$$

$$t > 0,\quad r = r_0,\quad -\lambda_s \frac{\partial T_s}{\partial r} = \zeta \frac{F_v\left(\Delta H_{vc}\right)}{4\pi R^2} + h_{conv}\left(T_s - T_b\right) + \sigma\varepsilon_r\left(T_s^4 - T_b^4\right) \tag{5}$$

The evaporation temperature at the drying front is 100℃, evaporation rate at particle surface is,

$$r = r_e = r_0,\quad R_e = \frac{h_{conv} A_e\left(T_b - T_s\right) + \sigma\varepsilon_r A_e\left(T_b^4 - T_s^4\right)}{\Delta H_{latent}} \tag{6}$$

And evaporation rate of particle internal is,

$$r = r_e \ne r_0,\quad R_e = \frac{\lambda_e A_e\left(\frac{\partial T}{\partial r}\right)}{\Delta H_{latent}} \tag{7}$$

The kinetic scheme with distributed activation energy is chosen for devolatilization of coal [35] as it eloquently incorporates lager number of simulations reactions and species [36], and the rate of devolatilization reaction is expressed as,

$$V\left(r,t\right) = V^* - V^* \int_{(E_0-4\sigma_E)}^{(E_0+4\sigma_E)} \exp\left[-k_0\left\{\int_0^t \exp\left(-\frac{E}{R_g T\left(r,t\right)}\right)dt\right\} f\left(E\right)\right] dE \tag{8}$$

$$f\left(E\right) = \frac{1}{\sigma_{\mathrm{E}}\sqrt{2\pi}} \exp\left[-\frac{(E-E_0)^2}{2\sigma_E^2}\right] \tag{9}$$

$$R_v = \frac{\partial V\left(r,t\right)}{\partial t}, \tag{10}$$

$$R_v = V^* k_0 \int_{(E_0-4\sigma_E)}^{(E_0+4\sigma_E)} \exp\left[-k_0 \int_0^t \exp\left(\frac{-E}{R_g T\left(r,t\right)}\right)dt\right]\left[\exp\left(\frac{-E}{R_g T\left(r,t\right)}\right)\right] f\left(E\right) dE$$

Volatile combustion is a global reaction,

$$C_x H_y O_z + \left(x + \frac{y}{4} - \frac{z}{2}\right) O_2 \rightarrow xCO_2 + \frac{y}{2} H_2O \tag{11}$$

with its,

$$F_v = \frac{d\left[C_x H_y O_z\right]}{dt} = A_0 \exp\left(\frac{-E_{cv}}{RT}\right)\left[C_x H_y O_z\right]^m \left[O_2\right]^n \tag{12}$$

The above equations from Eq. (1) to Eq. (12) make up the devolatilization model of the single coal particle, and R_v and F_v are the major parameters that affect the combustion behavior. Table 2 lists the value of simulation parameters, and shows that devolatilization kinetic parameters are highly similar in N_2

and CO_2 environment, which agree with the results reported by Guedea et al. [22] and Wang et al. [39].

Before volatile ignition occurs, not only a sufficient concentration of O_2 and volatiles must exceed the lower flammability limit, also gas temperature should exceed auto-ignition temperature. Fig. 11 plots the experimental and model predicted particle center temperature profiles under CO_2 and N_2 atmosphere. The model predicts agree well with the experiments. Volatile releases once the particle temperature reaches a certain value, and temperature profiles indicate that volatile evolution rate R_v is similar in both N_2 and CO_2 environment. However, the volatile diffusion rate in N_2 environment is 1.25 times that of in CO_2 environment at temperature of 850℃, and this leads to higher volatile concentration and lower oxygen concentration in a local filed. Besides, in this work, gas temperature under N_2 atmosphere was around 830℃, which was 10℃ higher than CO_2 atmosphere. And the different gas temperature is due to higher gas density and molar heat capacity of CO_2 (The ratio of $\rho_{CO_2} C_{CO_2} / \rho_{N_2} C_{N_2} = 1.7$ at 850℃). Therefore, lower volatile diffusion rate, higher density and molar heat capacity of CO_2 result in slower volatile reaction rate (Eq. 12) in CO_2 atmosphere than that in N_2 atmosphere.

5. Conclusion

In this work, single bituminous coal particle ignition and combustion behaviors were studied in a two-dimensional, transparent fluidized bed under O_2/N_2 and O_2/CO_2 atmosphere at 850℃. A combination of visual observation and measurement of particle center temperature was established.O_2 concentration was ranged from 0% to 40% and coal particle size was 6, 8.5, 10 and 13 mm. In addition, a transient devolatilization model was employed for discussion. The main conclusions of this study are listed as follow:

1. Under O_2/CO_2 atmosphere, coal particle ignition delay time is much longer than O_2/N_2 atmosphere and can be divided into two regions: (i) with O_2 concentration lower than 25%, the ignition delay time decreases by increasing O_2 concentration; (ii) with O_2 concentration higher than 25%, ignition delay time is almost constant by increasing O_2 concentration. The volatile combustion flame exhibits a blurred and yellow diffusion flame with low oxygen concentration, and flame luminosity increases by increasing O_2 concentration.
2. Under O_2/CO_2 atmosphere with 30% O_2 concentration, by increasing the particle diameter, both the ignition delay time and flame size increase, but when particle diameter is larger than 10 mm, the ignition delay time decreases a little which could be contributed by more amount of volatile released from the larger coal particles.
3. Under O_2/N_2 atmosphere, volatile combustion luminosity increases with increase in O_2 concentration, however, there is no obvious change of coal particle ignition delay time for different O_2 concentration.
4. Temperature raising profiles during devolatilization of the coal particles for different atmosphere (CO_2 vs. N_2) are similar, since millimeter-scale coal particle heating process is controlled by internal and external heat transfer. The effect of exothermic of volatiles reactions on heating and extinction time for larger coal particles can be neglected.

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Fig. 1. Photograph of experimental apparatus.

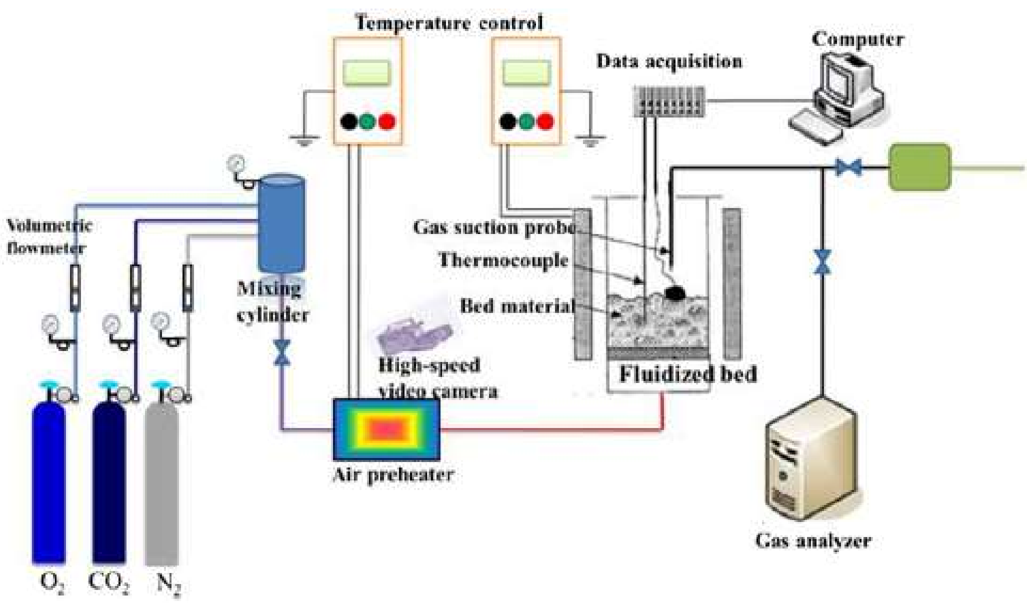

Fig. 2. Schematic diagram of the experimental system.

Fig. 3. Processes for the production of experimental coal particle, (i) spherical particles with a certain diameter, (ii) drilled a hole to the particle center, (iii) cemented a fine thermocouple into this hole.

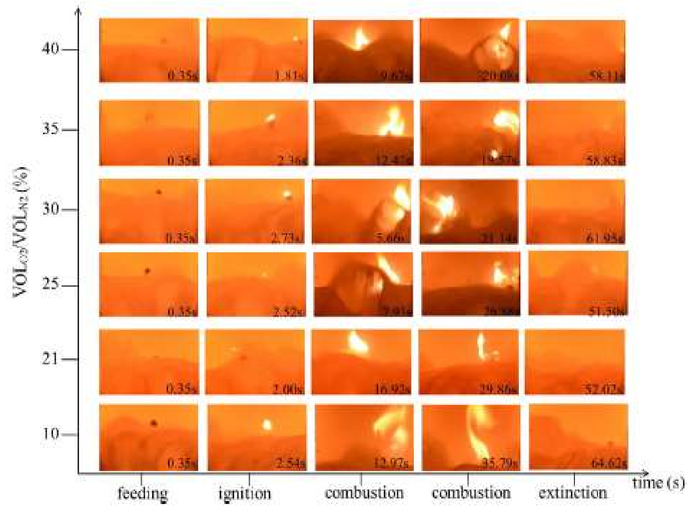

Fig. 4. Typical coal particle devolatilization events under various O_2 concentrations in N_2 atmosphere, particle diameter is 10 mm, bed temperature 850 °C.

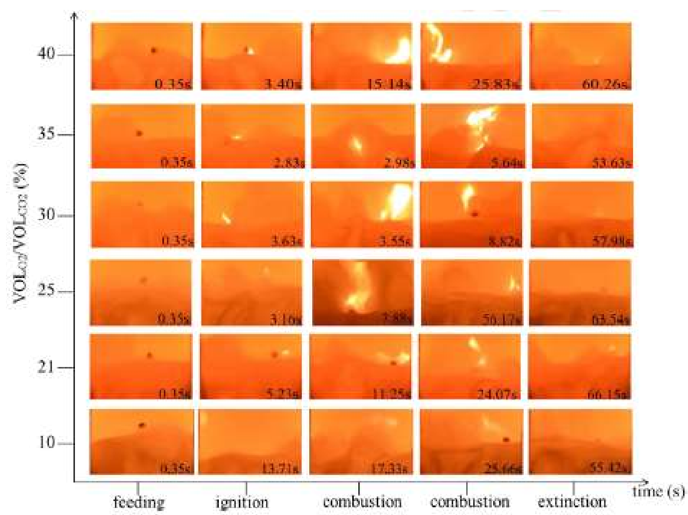

Fig. 5. Typical coal particle devolatilization events under various O_2 concentrations in CO_2 atmosphere, particle diameter is 10 mm, bed temperature 850 °C.

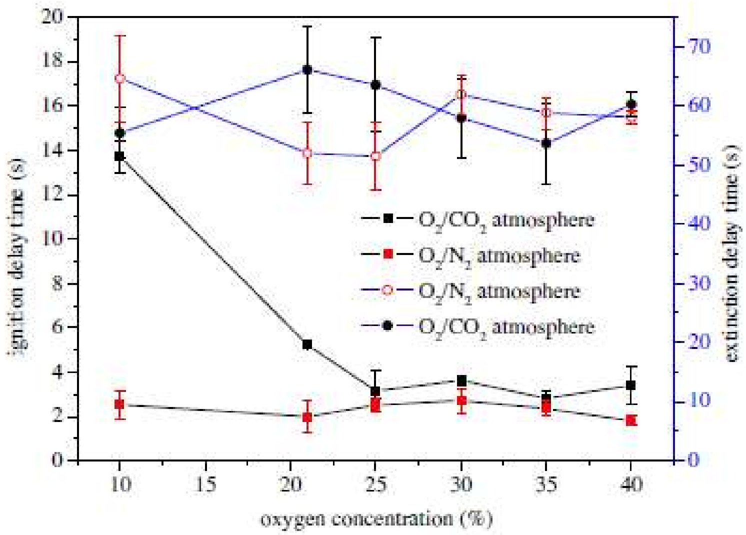

Fig. 6. Comparison of coal particle ignition delay time and extinction delay time under O_2/CO_2 and O_2/N_2 atmosphere, bed temperature 850 °C, particle diameter 10 mm.

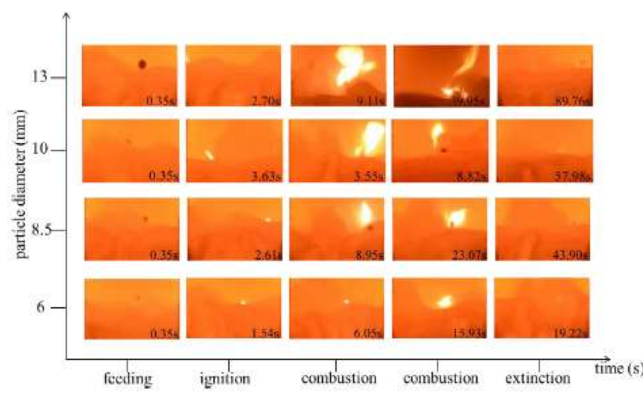

Fig. 7. Typical coal particle devolatilization events with different coal particle diameters under 30% O_2 concentrations in CO_2 atmosphere, bed temperature 850 °C.

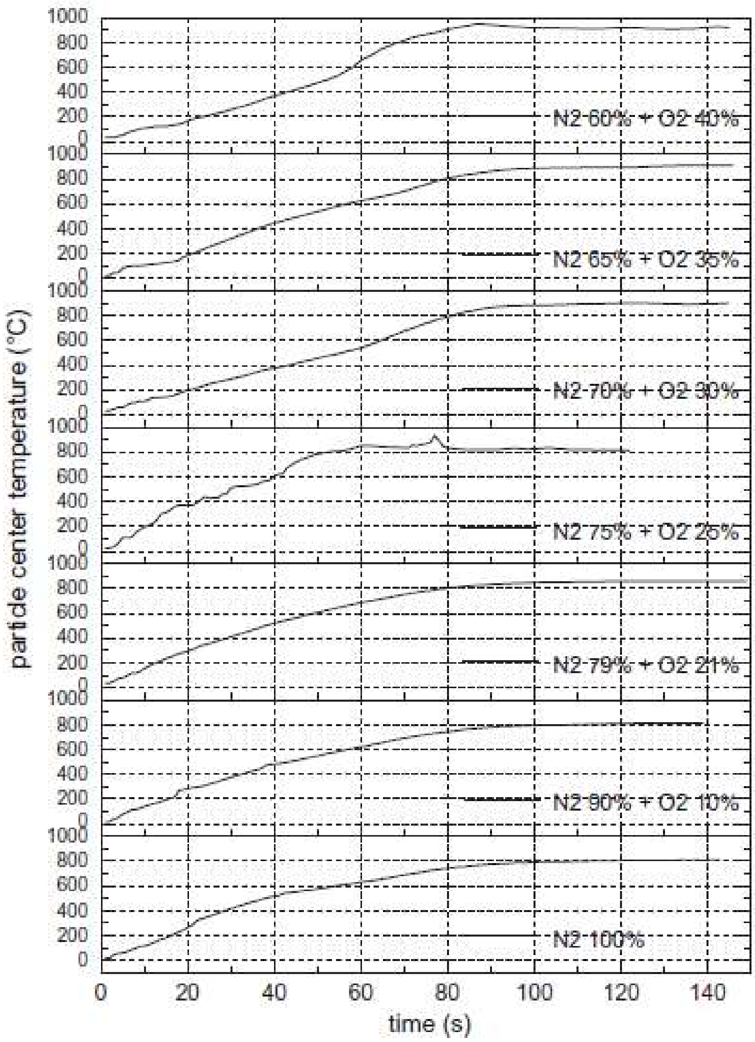

Fig. 8. Coal particles center temperature profile under various O_2 concentrations in N_2 atmosphere, particle diameter is 10 mm, bed temperature 850 °C.

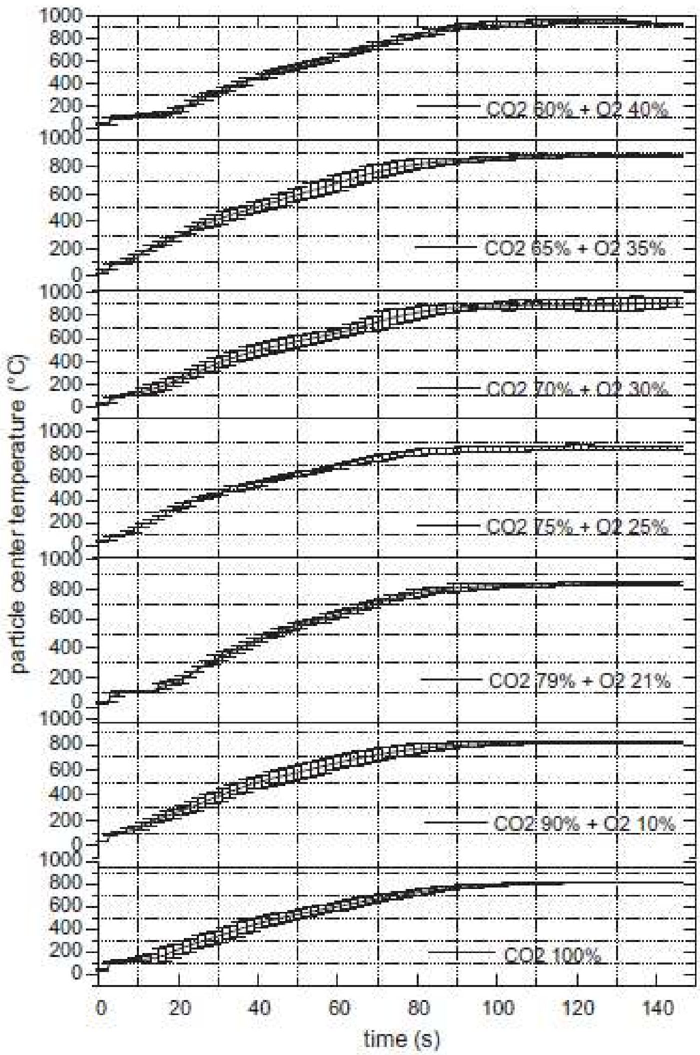

Fig. 9. Coal particles center temperature profile under various O_2 concentrations in CO_2 atmosphere, particle diameter is 10 mm, bed temperature 850 °C.

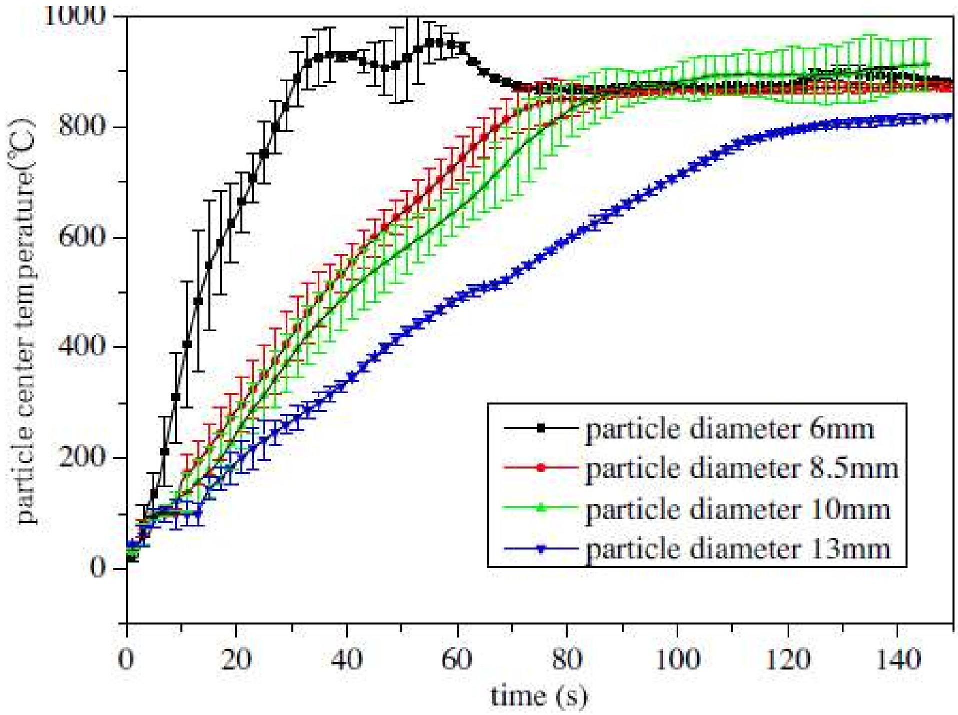

Fig. 10. Coal particle center temperature profile under 30% O_2 concentration in atmosphere with different diameters, bed temperature 850 °C.

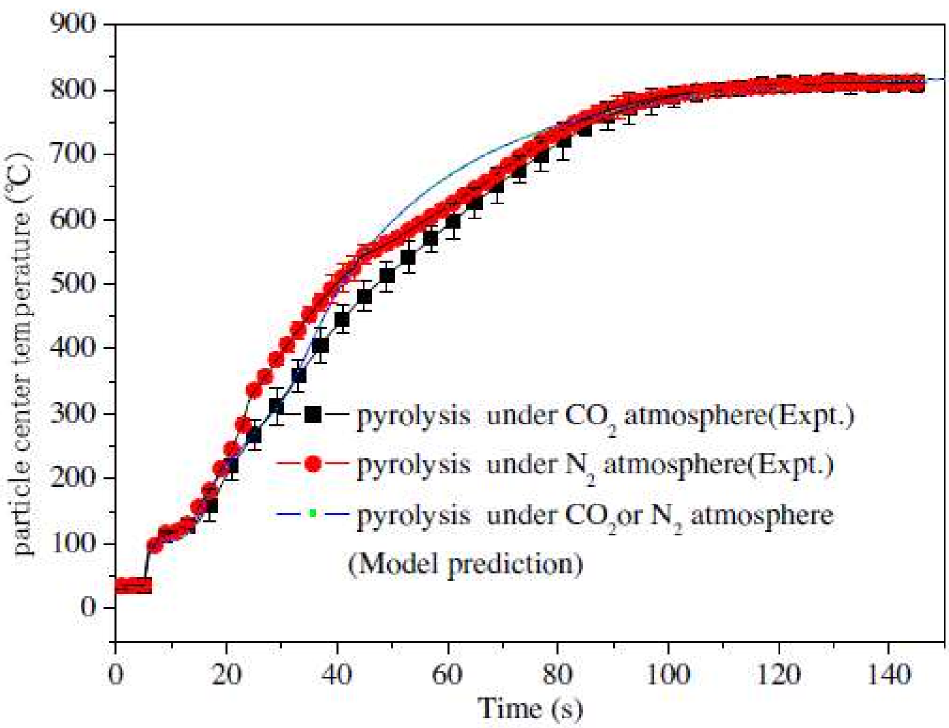

Fig. 11. Experimental and model predicted particle center temperature profiles under CO_2 and N_2 atmosphere, particle diameter 10 mm, bed temperature 850 °C.