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Thermal Analysis and Kinetics of Coal during Oxy-Fuel Combustion

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The pyrolysis and oxy-fuel combustion characteristics of Polish bituminous coal were studied using non-isothermal thermogravimetric analysis. Pyrolysis tests showed that the mass loss profiles were almost similar up to 870°C in both N_2 and CO_2 atmospheres, while further mass loss occurred in CO_2 atmosphere at higher temperatures due to char-CO_2 gasification. Replacement of N_2 in the combustion environment by CO_2 delayed the combustion of bituminous coal. At elevated oxygen levels, TG/DTG profiles shifted through lower temperature zone, ignition and burnout temperatures decreased and mass loss rate significantly increased and complete combustion was achieved at lower temperatures and shorter times. Kinetic analysis for the tested coal was performed using Kissinger-Akahira-Sunose (KAS) method. The activation energies of bituminous coal combustion at the similar oxygen content in oxy-fuel with that of air were higher than that in air atmosphere. The results indicated that, with O_2 concentration increasing, the activation energies decreased.

Keywords: oxy-fuel combustion, pyrolysis, bituminous coal, TGA, DTA, kinetics analysis, KAS method

Introduction

Oxy-fuel combustion is one of the leading technologies considered for capturing CO_2 from power plants with CCS (Carbon Capture and Storage) [1]. This technology can reduce significantly emissions of NO_x and improve the thermal efficiency of the combustion process by reducing the flue gas volume. In the oxy-fuel combustion, coal particles are burnt in a mixture of pure oxygen and recycled flue gas. Because nitrogen is eliminated from the oxidizing gas, the flue gas that leaves the combustion chamber is highly enriched in CO_2, which implies that the combustion process occurs in an O_2/CO_2 environment. Partial recycling of flue gas helps to control the flame temperature in the combustion chamber. Extensive studies in both pilot-plant and lab scales have pointed out the pronounced influence of gas composition (air versus O_2/CO_2) on coal combustion performance. The heat transfer and temperature distribution in a furnace are greatly affected by the large specific heat capacity of CO_2. Coal ignition is delayed in O_2/CO_2 in comparison to in O_2/N_2 with the same O_2 concentration. To match the flame/particle temperature in air, a large amount of O_2 in CO_2, typically around 30%, is required. Coal conversion rate, char properties, and reactivity are also affected by the replacement of air with an O_2/CO_2 mixture [2-4].

A Thermal Analysis (TA) determines a set of methods to study selected physical properties of the substance under the temperature effect [5]. Thermogravimetry (TG) is a technique that monitors the sample mass as a function of temperature or time when the sample is subjected to a controlled temperature program. Differential Thermogravimetry (DTG) is based on the rate of mass loss. For example, the DTG profiles enable one to know the mass loss at a temperature during the combustion process. The Differential Thermal Analysis (DTA) enables measuring the thermal effects during the studied process. The

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obtained peaks correspond to exothermic or endothermic effects, which explain the thermal behaviour of fuel. The most important applications of the thermal analysis are kinetics calculations. Kinetic parameters such as the activation energy and pre-exponential factor can be obtained to analyse the non-isothermal solid-state kinetics [5, 6]. Solid-state kinetic data are the major interest in combustion processes. The kinetic studies of pulverized coal, anthracite, coal gangue and different types of biomass in oxy-fuel combustion have been studied by many research groups [7-18]. Despite of significant ongoing research in this area, there are few studies on the oxy-fuel combustion properties of solid fuel particles larger than 1 mm. The kinetic parameters for larger coal particles in oxy-fuel combustion are required for computational fluid dynamics (CFD) calculations to support efficient oxy-CFB boiler modelling and design. The absence of studies on pyrolysis and combustion behaviours of larger coal particles in TGA-DTA under oxy-fuel condition necessitates its investigation. To fulfil this requirement, in this paper, the pyrolysis and combustion behaviours and kinetics of Polish bituminous coal with particle size of 1-2 mm were investigated in air and O_2/CO_2 mixtures, whose oxygen concentrations were 21-40% vol., using TG/DTG and DTA. The activation energies of bituminous coal during air and oxy-combustion were calculated using Kissinger-Akahira-Sunose (KAS) method.

Experimental

Coal sample

Polish bituminous coal (Ziemowit coal mine), which is used extensively for firing in CFB boilers in Polish power plants, was selected for this study. The proximate and ultimate analyses of the tested coal are shown in Table 1.

Table 1 Proximate and ultimate analyses of bituminous coal

Proximate analysis (dry basis)	
Moisture (wt.%)	8.7
Volatile matter (wt.%)	26.8
Ash (wt.%)	18.9
Fixed carbon (wt.%)	45.6
Higher heating value (MJ/kg)	22.75
Ultimate analysis (dry ash free basis)	
Carbon (wt.%)	73.3
Hydrogen (wt.%)	4.3
Nitrogen (wt.%)	1.1
Sulphur (wt.%)	2.3
Oxygen (by difference) (wt.%)	19.0

Apparatus and procedure

The pyrolysis and combustion characteristics of bituminous coal were studied using a simultaneous TGA/DTA analyser (TA Instruments STD 2960). Approximately 14±0.5 mg of coal sample was heated from ambient temperature to 1300 °C during each experiment. All experiments were performed under non-isothermal conditions at a constant heating rate of 50 °C/min. The total gas flow rate of 160 $ml \cdot min^{-1}$ was used in all experiments. The experiments of the heating rate effect on combustion and for kinetics analysis were performed at four constant heating rates (5, 10, 15 and 25 °C/min) in different oxidizing atmospheres. The following gas atmospheres were used in the experiments:

- air-combustion:
  21% O_2/79% N_2,
- oxy-combustion:
  21% O_2/79% CO_2,
  30%O_2/70% CO_2,
  40% O_2/60% CO_2,
- pyrolysis:
  100% N_2,
  100% CO_2.

TG and DTG curves, which were obtained during the combustion experiments, were used to determine the combustion behaviour and some characteristic temperatures such as the initial decomposition temperature (T_{in}), ignition temperature (T_{ig}), peak temperature (T_m), and burnout temperature (T_b) of the tested coal. T_{in} represents the initiation of mass loss and is defined as the temperature at which the rate of mass loss reaches 1 $\%\cdot min^{-1}$ after the initial moisture-loss peak in the DTG profile [14]. The ignition temperature (T_{ig}) is defined as the temperature at which the fuel begins to burn. It is taken as the temperature at which the mass loss curves in the oxidation and pyrolysis experiments diverge. The peak temperature (T_m) is the point at where the maximum reaction rate (DTG_{max}) occurs. The burnout temperature (T_b) is the temperature when the sample oxidation is completed. It is taken as the point immediately before the reaction ceases, when the rate of mass loss is 1 $\%\cdot min^{-1}$.

Results and Discussion

Pyrolysis characteristics

Pyrolysis tests of Polish bituminous coal with particle size of 1-2 mm were performed in 100% N_2 and 100% CO_2 atmospheres at a heating rate of 50 °C/min. The mass loss (TG) and mass loss rate (DTG) curves of the tested coal pyrolysis are shown in Fig. 1. The pyrolysis TG/DTG curves show similar trends in N_2 and CO_2 until they reach a critical temperature. After this critical temperature, which is approximately 870°C for the tested bituminous coal, there is a drastic mass loss, which is attributed to the char-CO_2 gasification reaction.

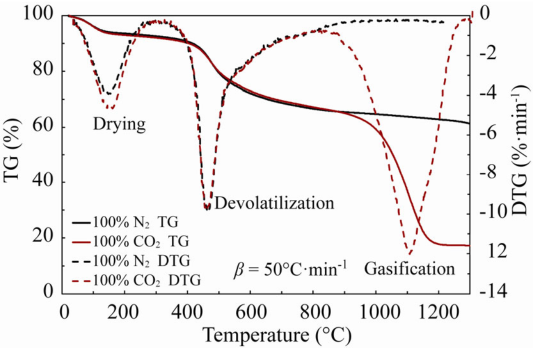


Fig. 1 TG/DTG curves of bituminous coal pyrolysis in N_2 and CO_2 atmospheres

Two main mass loss steps appear in the DTG profiles in nitrogen atmosphere, whereas an additional mass loss step is observed after 870°C in the CO_2 environment. The first mass loss step in the 25–200°C temperature range accounts for moisture release. The peak values of moisture release are approximately 150°C for 1-2 mm particles in N_2 and CO_2 atmospheres. The second mass loss step within 300–700°C corresponds to the volatile- matter release in both atmospheres. For Polish bituminous coal, the peak values of volatile-matter release are at approximately 465 °C both in N_2 and CO_2 atmospheres. This temperature is identical with the value that Wang et al. [8] observed for Lingxin bituminous coal in N_2 and CO_2 atmospheres. The maximum rate of mass loss of coal devolatilization is approximately 10 %·min^{-1} for 1-2 mm. The mass loss in the higher temperature range of 870-1250°C in the 100% CO_2 atmosphere is attributed to the char-CO_2 gasification reaction. The gasification peak occurs at approximately 1100 °C and the maximum DTG value is approximately 12 %·min^{-1}. There is a difference in critical temperature of char-CO_2 gasification as reported by different investigators. Rathnam et al. [19, 20] observed that the char-CO_2 gasification reaction began at approximately 700°C for pulverized lignite, 820°C for pulverized bituminous coal and above 930°C for pulverized semi-anthracite. Yuzbasi et al. [14] also observed an additional peak after 700°C in a CO_2 atmosphere for lignite, olive residue and their blend with 50/50 wt.%. Moreover, Wang et al. [8] indicated that at approximately 810°C the rate of mass loss for Lingxin bituminous pulverized coal in a CO_2 atmosphere began to significantly increase compared to that in N_2. They concluded that the char-CO_2 gasification reaction of Lingxin coal played an important role when the temperature was above 810°C in oxy-fuel combustion. They also calculated the apparent activation energy E_a and pre-exponential factor A of coal pyrolysis and gasification reactions. They found that both apparent activation energy and pre-exponential factor of coal pyrolysis in N_2 (E_a = 41.52 kJ·mol^{-1}; A = 60.52 s^{-1}) are consistent with those in CO_2 (E_a = 42.43 kJ·mol^{-1}; A = 72.38 s^{-1}). However, the activation energy of the char-CO_2 gasification reaction is 232.86 kJ·mol^{-1} and significantly exceeds that of the pyrolysis reaction because the char-CO_2 gasification is an intense endothermic reaction, which can only proceed at high temperature [8]. This phenomenon can be observed in the DTA curve during the pyrolysis of coal in CO_2 atmosphere in Fig. 2. The char-CO_2 gasification reaction is highly endothermic and may decrease the particle temperature.

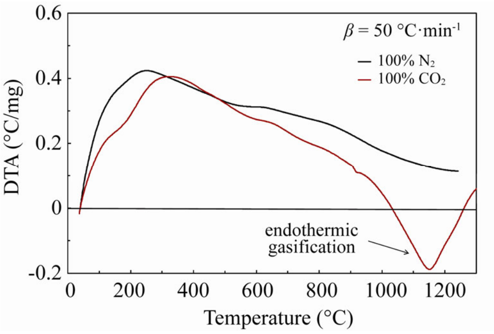


Fig. 2 DTA curves of bituminous coal pyrolysis in N_2 and CO_2 atmospheres

Combustion characteristics

The TG/DTG curves of the tested bituminous coal with particle sizes 1-2 mm in air (21%O_2/79%N_2) and oxy-fuel (21%O_2/79%CO_2) combustion are presented in Fig. 3. The first stage was heating and drying till approximately 300°C. In this stage, approximately 10% mass loss was observed. The second stage in the temperature range of 300-410°C accounts for the devolatilization of light components. In the third stage, the volatiles ignited at 415°C in air and 420°C in 21%O_2/79%CO_2.

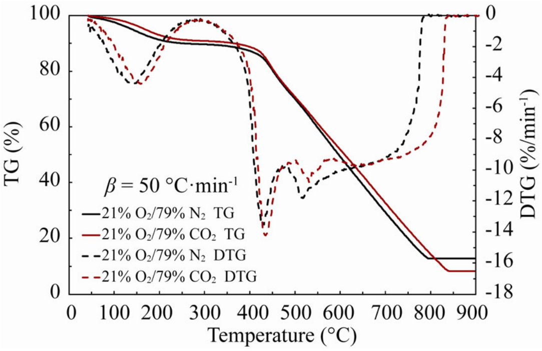


Fig. 3 TG/DTG curves of bituminous coal combustion in O_2/N_2 and O_2/CO_2 atmospheres of 21% O_2

The next stage was combustion of the volatiles, during which the peak temperature and maximum mass loss rate were observed. Above 530°C the combustion of char began. In this stage DTA_{max} (Fig. 4) was observed, which indicates ongoing exothermic reactions. At approximately 800°C the combustion process of bituminous coal ended, which indicates the constant mass of incombustible ash.

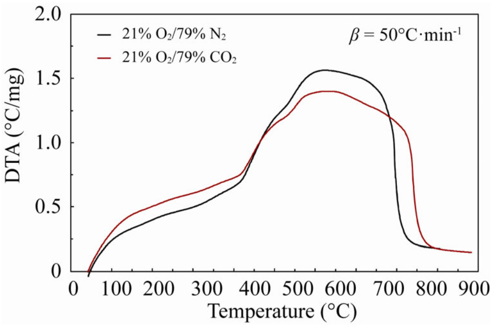


Fig. 4 DTA curves of bituminous coal combustion in O_2/N_2 and O_2/CO_2 atmospheres of 21% O_2

The total mass loss of the coal sample was approximately 87% in air and 91% in $21\%O_2/79\%CO_2$. The TG/DTG curves of bituminous coal in oxy-fuel combustion obviously shift to a higher temperature zone compared to those of air combustion under identical oxygen concentrations. The characteristic parameters of combustion in air and oxy-fuel combustion are also shown in Table 2. It can be observed that the process of bituminous coal combustion in the $21\%O_2/79\%CO_2$ atmosphere is delayed. Experimental results indicate that the initial temperature (T_{in}) and ignition temperature (T_{ig}) increase slightly whereas the peak temperature (T_{max}) and burnout temperature (T_b) increase significantly. Additionally, the maximum mass loss rate (DTG_{max}) and DTA_{max} in oxy-fuel combustion are less than those in air combustion. The $21\%O_2/79\%CO_2$ atmosphere may lower both gas and coal particles temperatures, mainly due to the higher specific heat of CO_2 than that of N_2. Consequently, the reduced temperature would lead to increased combustion time of coal particles in O_2/CO_2 atmospheres. In addition, the diffusivity of O_2 in CO_2 is lower than that in N_2, which would worsen the transport of O_2 to the coal particle surface [8]. These findings are in agreement with those in the literature [7-12].

Fig. 5 shows DTG curves of bituminous coal combustion in different oxygen concentration in O_2/CO_2 mixture. It can be observed that DTG profiles shift to a lower temperature zone, particularly in the $40\%O_2/60\%CO_2$ atmosphere.

It can be noted from Table 2 that the ignition temperature (T_{ig}), peak temperature (T_{max}) and burnout temperature (T_b) decrease with increased oxygen concentration. On the other hand, the maximum rate of mass loss (DTG_{max}) increased from 1.87 to 3.03 $mg \cdot min^{-1}$ in the $21\%O_2/79\%CO_2$ atmosphere and $40\%O_2/60\%CO_2$, respectively.

The DTA profiles of the tested coal samples during oxy-fuel combustion are shown in Fig. 6. It can be seen that the DTA curves become taller and slimmer with the increase of oxygen concentration. Consequently, the heat released more rapidly. This is in accordance with the conclusion obtained by Wang et al. [8].

Fig. 7 shows the effect of the oxygen concentration on the ignition and burnout temperatures of the tested bituminous coal with particle sizes of 1-2 mm in O_2/CO_2 atmospheres at a heating rate of 50 °C/min. The present study and the studies reported by Wang et al. [8] show a decrease in ignition and burnout temperatures with oxygen concentration increasing. However, the oxygen concentration significantly affects the burnout temperature and slightly the ignition temperature. These trends are consistent with those in the literature [8, 11, 14, 16, 18 and 20]. Lower temperature values obtained by Wang et al. (Fig. 7) were associated with smaller particle sizes (0.1-0.125 mm) and a lower heating rate (20 °C/min).

It can be concluded that the oxygen concentration conspicuous affects the coal particles combustion. At elevated oxygen levels the DTG profiles shifted through lower temperature zone, ignition and burnout temperatures decreased and mass loss rate significantly increased and complete combustion was achieved at lower temperatures and shorter times.

Table 2 Characteristic parameters obtained from the DTG and DTA profiles of bituminous coal during combustion in all atmospheres at a heating rate of 50 $°C \cdot min^{-1}$

Atmosphere	T_{in}	T_{ig}	T_{max}	DTG_{max}	DTA_{max}	T_b	*Total mass loss*
	°C	°C	°C	$mg \cdot min^{-1}$	$°C \cdot min^{-1}$	°C	%
21% O_2/79% N_2	340	415	430	1.98	1.56	780	87
21% O_2/79% CO_2	349	420	442	1.87	1.40	830	91
30% O_2/70% CO_2	330	405	415	2.21	2.02	730	93
40% O_2/60% CO_2	315	390	396	3.03	2.79	620	92

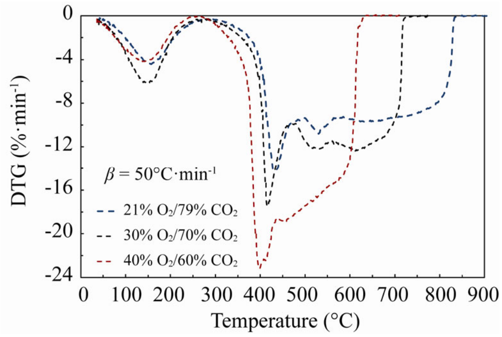


Fig. 5 DTG curves of bituminous coal combustion in O_2/CO_2 atmospheres

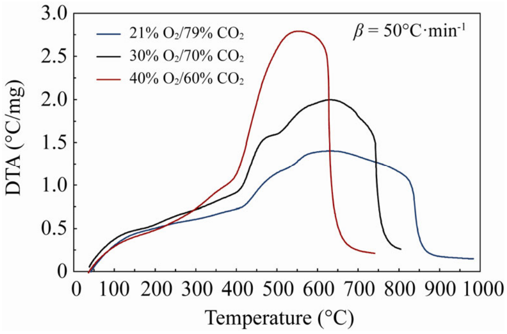


Fig. 6 DTA curves of bituminous coal combustion in O_2/CO_2 atmospheres

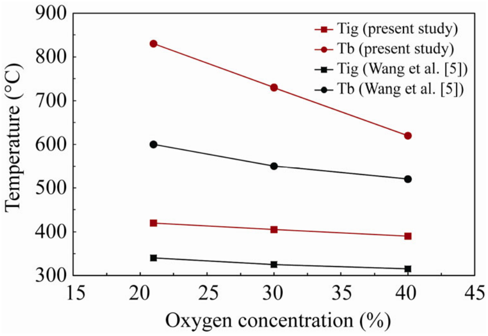


Fig. 7 Effect of the oxygen concentration on the ignition temperature (T_{ig}) and burnout temperature (T_b) of bituminous coal combustion in O_2/CO_2 atmospheres

Kinetics analysis

The general kinetic equation can be written as follows:

$$\frac{d\alpha}{dt} = k(T) f(\alpha) \tag{1}$$

where *á* is the conversion degree, *t* (s) is time, *T* (K) is the absolute temperature, *k(T)* is the temperature-dependent rate constant and *f(á)* is a function, the type of which depends on the reaction mechanism.

Conversion degree (α) is defined as follow:

$$\alpha = \frac{m_0 - m}{m_0 - m_{ash}} \tag{2}$$

where: m_0 (mg) is the initial mass of coal sample, m (mg) is instantaneous mass of sample and m_{ash} (mg) is final mass of ash.

The rate constant is usually described by Arrhenius equation:

$$k = A\exp\left(-\frac{E}{RT}\right) \tag{3}$$

where A (min^{-1}) is the pre-exponential factor, E (kJ·mol^{-1}) is the activation energy, and R (8.314 kJ·mol^{-1}) is the universal gas constant.

The function of *f(α)* is expressed as:

$$f(\alpha) = (1-\alpha)^n \tag{4}$$

where n is the reaction order.

Kissinger-Akahira-Sunose (KAS) method is an isoconversional linear integral method based on the equation [5, 11]:

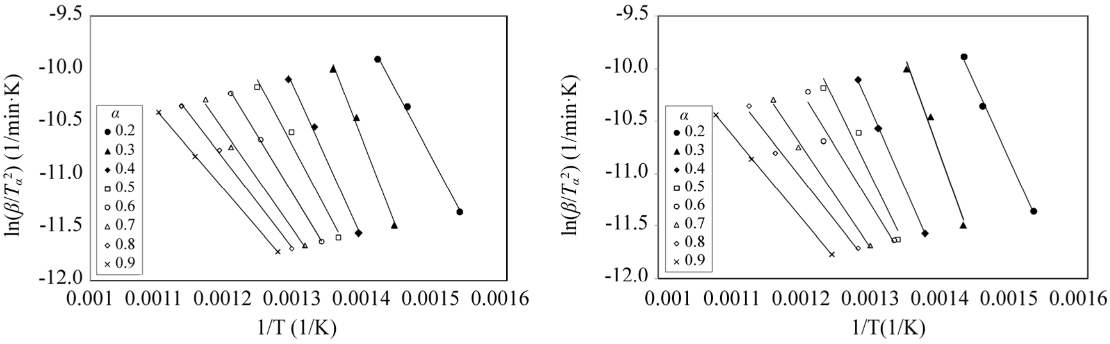
$$\ln\left(\frac{\beta}{T_\alpha^2}\right) = \ln\left(\frac{A_\alpha R}{E_\alpha g(\alpha)}\right) - \frac{E_\alpha}{RT_\alpha} \tag{5}$$

where β (K·min^{-1}) is the heating rate. The apparent activation energy can be obtained from a plot of $ln(\beta/T_\alpha^2)$ versus $1/T$ for a given value of conversion *á*, where the slope is equal to $-E_a/R$.

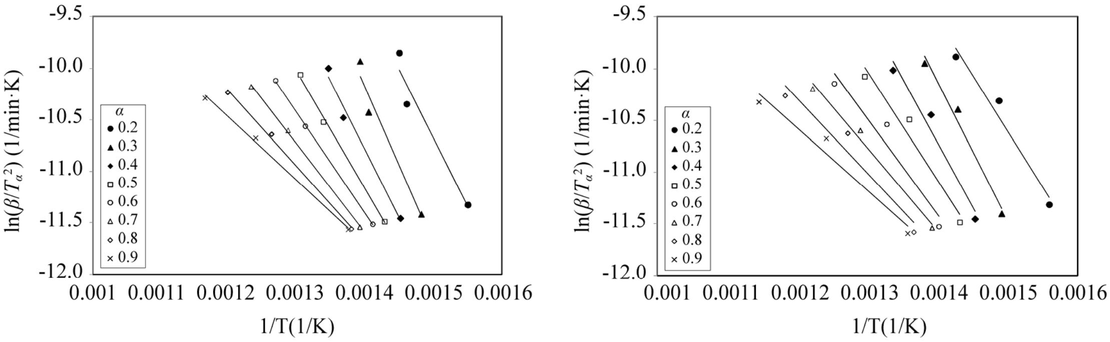
The apparent activation energy (E_a) and corresponding correlation coefficients (R^2), which were calculated using KAS method, are shown in Fig. 8 shows plots to determine of E_a in different atmospheres using KAS method. Table 3. E_a has high linear correlation coefficient (R_2, related coefficient) in the range of 0.94-0.99.

Table 3 Activation energies obtained from TG data at different α using Kissinger-Akahira-Sunose method

α	21% O_2/79% N_2		21% O_2/79% CO_2		30% O_2/70% CO_2		40% O_2/60% CO_2	
	E_a (kJ/mol)	R^2	E_a (kJ/mol)	R^2	E_a (kJ/mol)	R^2	E_a (kJ/mol)	R^2
0.2	102.69	0.99	124.90	0.99	109.9	0.94	89.92	0.97
0.3	141.16	0.99	158.13	0.98	126.32	0.96	111.29	0.97
0.4	121.23	0.98	130.28	0.99	109.62	0.98	103.03	0.96
0.5	102.93	0.98	116.38	0.96	94.57	0.99	87.31	0.96
0.6	89.41	0.99	92.20	0.97	81.22	0.99	77.04	0.94
0.7	78.34	0.99	83.56	0.99	71.49	0.99	66.56	0.98
0.8	70.89	0.99	71.88	0.98	61.31	0.99	59.94	0.94
0.9	63.67	0.99	67.97	0.99	51.07	0.99	50.26	0.96



(a) 21% O_2/79% N_2 (b) 21% O_2/79% CO_2



(c) 30% O_2/70% CO_2 (d) 40% O_2/60% CO_2

Fig. 8 Arrhenius plots to determine of E_a in different atmospheres using KAS method

The activation energy that corresponds to the bituminous coal combustion in a 21%O_2/79%CO_2 atmosphere is always higher than combustion in 21%O_2/79%N_2 (Fig. 9). According to the reaction kinetic theory, higher activation energy implies that it is more difficult to react. In an O_2/CO_2 atmosphere, the activation energy slightly decreases with an increasing oxygen concentration. The trends are consistent with those in the literature [7, 10, 11, 18].

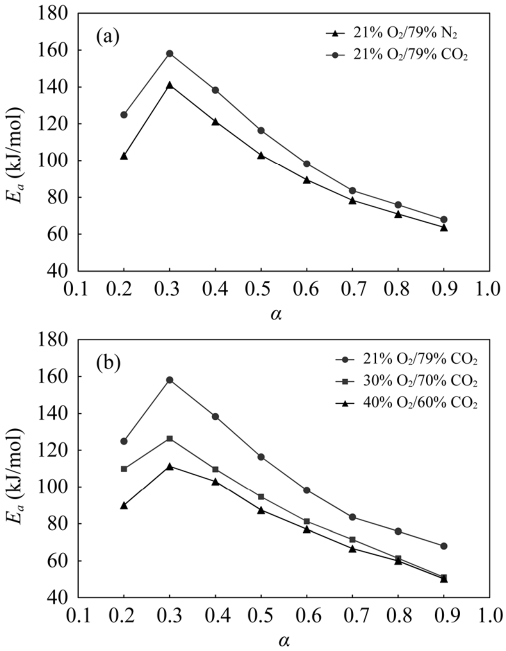


Fig. 9 Activation energies as a function of the conversion degree (α) during the bituminous coal combustion in (a) O_2/N_2 and O_2/CO_2 atmospheres of 21% O_2 and (b) O_2/CO_2 atmospheres, using KAS method

Conclusions

The pyrolysis and combustion behaviours and kinetics of Polish bituminous coal with particle sizes of 1-2 mm, which are dominant in the particle size distribution of feeding coal for the circulating fluidized bed boiler, were carried out using TGA/DTA. On the basis of the data obtain from experiments, the main conclusions are as follows:

(1) Pyrolysis tests in N_2 and CO_2 atmospheres showed that the mass loss profiles were almost similar up to 870°C in both atmospheres, while further mass loss occurred in CO_2 atmosphere at higher temperatures due to char-CO_2 gasification. The DTA curve indicated that the char-CO_2 gasification reaction was highly endothermic and may decrease the particle temperature.

(2) The TG/DTG curves of bituminous coal in oxy-fuel combustion obviously shift to a higher temperature zone compared to those of air combustion under identical oxygen concentrations. The 21%O_2/79%CO_2 atmosphere may lower both gas and coal particles temperatures,

mainly due to the higher specific heat of CO_2 than that of N_2. Consequently, the reduced temperature would lead to increased combustion time of coal particles in O_2/CO_2 atmospheres. In addition, the diffusivity of O_2 in CO_2 is lower than that in N_2, which would worsen the transport of O_2 to the coal particle surface.

(3) At elevated oxygen levels the DTG profiles shifted through lower temperature zone, ignition and burnout temperatures decreased and mass loss rate significantly increased and complete combustion was achieved at lower temperatures and shorter times.

(4) The kinetic parameters for larger coal particles (d > 1 mm) in oxy-fuel combustion are required for computational fluid dynamics (CFD) calculations to support efficient oxy-CFB boiler modelling and design. The activation energies of bituminous coal were in the range of 64-141 kJ/mol in the air combustion. With the increase of the oxygen concentration in the oxy-fuel combustion, the values of apparent activation energy in 21% and 40% O_2 decreased from 68-158 kJ/mol to 50-111 kJ/mol, respectively.

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