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Investigation of combustion characteristics and kinetics of coal gangue with different feedstock properties by thermogravimetric analysis

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ABSTRACT

The combustion behavior of eleven coal gangue was investigated by thermogravimetric analysis (TGA). The effects of feedstock properties (combustible matter and mineral matter) on combustion characteristics of coal gangue were analyzed. The kinetic parameters were obtained based on experimental results by distributed activation energy model (DAEM). The results show that the ignition temperature of coal gangue is affected by volatile content and oxygen adsorption behavior jointly. In the main weight loss stage, combustion reaction of combustible matter and dehydroxylation reaction of mineral matter proceeded simultaneously. Combustion conditions of volatile matter and fixed carbon, as well as, decomposition of kaolinite, illite and calcite exhibited different characteristic DTG curves. The activation energy for the combustion of coal gangue was in the range of 117–300 kJ/mol. Feedstock properties affected the change of activation energy with conversion values. The maximum relative errors between calculated and experimental results by DAEM model were 0.2–0.5%.

1. Introduction

Coal is the most widely consumed source of energy in China [1]. Coal gangue is a residue from the coal mining and washing process [2]. On a mass basis, coal gangue accounts for approximately 10–15% of the total coal produced [3]. Hence, coal gangue is one of the largest sources of industrial solid waste in China. Coal gangue disposal occupies a tremendous amount of land and results in serious environmental problems [4,5]. Therefore, Chinese government policies have been developed to encourage coal gangue utilization. It is estimated that the calorific value of coal gangue is 30% of the source coal. Owing to containing combustible matter in coal gangue and increasing demand for electricity, coal gangue is widely used in low-calorific value coal-fired power plant [4,6,7]. The chief advantage of coal gangue is wide-spread availability at little or no cost, which in-turn creates economic benefits.

When combusting coal gangue, the circulating fluidized bed (CFB) boiler plays an important role. CFB combustion has many advantages when utilizing low-rank fuel [4]. The high ash content of coal gangue does lead to some combustion problems, such as poor burnout and flame instability [4,6]. A systematic understanding of high-ash coal gangue combustion behavior is essential for maximizing efficiency while producing stable operating conditions. Coal gangue combustion behaviors have been investigated in previous studies. Some investigations focused on the effect of atmospheric conditions. Meng et al. found that coal gangue combustion under oxy-fuel differed from that under air [6]. The higher concentration of oxygen resulted in lower ignition and burnout temperatures. Ren et al. believed that the comprehensive combustion characteristic index increased slightly when methane was present in the combustion atmosphere [8]. Other researchers pointed out that combustible matter content also had significant effects since ignition and combustion of combustible matter are the rate-controlling steps [9]. Xiao et al. concluded that co-combustion of coal gangue with coal was beneficial for coal gangue combustion [2]. Ran et al. thought that coals with high volatile content could improve overall combustion performance of coal gangue [10]. High-ash coal gangue combustion behavior is not only influenced by combustible matter but also by mineral matter. This effect is most probably due to the heat absorption of mineral matter [9,11,12]. Moreover, the feedstock properties (combustible matter, mineral matter) of coal gangue are different depending on source, coal rank, and coal mining technique and washing technology. However, the effect of different feedstock properties on coal gangue combustion behavior is not very clear.

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Kinetic parameters are required for computational fluid dynamics (CFD) simulation to support efficient CFB boiler design and maintenance [13]. Actual CFB boiler conditions vary significantly depending on the operating set points and the different zones within the CFB. Influential factors are complicated [14]. The variances within the CFB make obtaining the kinetic characteristics of coal gangue combustion difficult. Thermogravimetric analysis (TGA) is the easiest and most cost effective technique used to determine kinetic characteristics [15]. Utilizing the thermogravimetric analyzer, the kinetic parameters of coal gangue combustion have been calculated using some models. The activation energy (E) of coal gangue combustion was determined via the Coats–Redfern method and determined to be 117.6 kJ/mol [8]. However, the E value obtained from a single TG curve is unreliable owing to the complexity of a solid state reaction. The Flynn–Wall–Ozawa method based on four TG curves should be more reliable, with reported E values of 120–170 kJ/mol [6]. However, there is no information concerning other kinetic parameters, such as frequency factor and reaction mechanism. The distributed activation energy model (DAEM) can obtain kinetic triplets. This model has been widely used in analyzing complex reaction systems, such as coal pyrolysis, biomass pyrolysis, co-combustion of oil shale and its semi-coke [16–20]. The coal gangue combustion system is similar to these systems, and includes many complex reactions. However, the application of DAEM to coal gangue combustion system has scarcely been studied.

In the present paper, combustion behaviors and kinetics of coal gangue with different feedstock properties were investigated under air using thermogravimetric analysis (TGA) and the distributed activation energy model (DAEM). The changes in mineral matter during combustion were analyzed by X-ray diffraction techniques (XRD). Experiments were conducted to study the influence of feedstock properties (combustible matter, mineral matter) on the combustion characteristics. Results provided insightful information for evaluation of coal gangue combustion. This information will contribute to the rational use of coal gangue.

2. Experimental

2.1. Materials

Eleven coal gangue samples were collected from different mines. These mines produce coal with obvious differences in the extent of coalification (ranging from lignite through bituminous to anthracite). Additionally, the mines utilize different coal preparation technologies, such as heavy media separation and jigging process. Table 1 lists the proximate analyses, ultimate analyses and mineral phases of the coal gangue samples. From Table 1, it is observed that eleven coal gangue samples have different calorific values. Their volatile content, fixed carbon content and ash content are in the range of 9.6–28.2%, 10.6–34.3%, 40.7–77.0%, respectively. The main mineral phases include quartz, kaolinite, illite, calcite and pyrite. In the laboratory, each sample was air-dried, milled and sieved to less than 75 μm to minimize heat and mass transfer restrictions.

2.2. Thermogravimetric analysis (TGA)

A PerkinElmer Pyris 1 TGA was used for thermogravimetric combustion experiments. The combustion experiments were carried out under air atmosphere with a flow rate of 60 ml min^{-1}. The sample (3 ± 0.1 mg) was heated from ambient up to a maximum temperature of 1000 °C at constant four heating rates: 10 °C min^{-1}, 20 °C min^{-1}, 30 °C min^{-1} and 40 °C min^{-1}. The measurements under same condition were necessary to produce the most repeatable and precise results. The experiments were replicated at least twice to determine reproducibility, which was found to be very good.

2.3. Determination of combustion characteristic parameters

Ignition temperature (T_i), burnout temperature (T_f), and peak temperature (T_p) are the combustion characteristic parameters reflecting thermal behavior during the combustion process. These parameters can be derived from TG and DTG curves [21,22]. T_i is defined as the temperature at which a sample starts burning, and determined by the TG–DTG tangent method [23–26]. T_f is taken as the point immediately before the combustion reaction is completed, when the rate of weight loss becomes less than 1 wt % min^{-1} [27,28]. T_p is the temperature corresponding to the peak of the DTG profile [29].

The ignition index (D_i) and the burnout index (D_f) are defined in Eq. (1) and Eq. (2) below, respectively [30,31].

$$D_i = \frac{\mathrm{DTG_{max}}}{t_p t_i} \quad (1)$$

where $\mathrm{DTG_{max}}$ is the maximum combustion rates, t_p is the corresponding time of $\mathrm{DTG_{max}}$,t_i is the ignition time.

$$D_f = \frac{\mathrm{DTG_{max}}}{\Delta t_{1/2} t_p t_f} \quad (2)$$

where $\Delta t_{1/2}$ is the time range of DTG/ $\mathrm{DTG_{max}}$ = 0.5, t_f is the burnout time.

Table 1
Proximate analyses, ultimate analyses and mineral phases of coal gangue samples.

Samples	Areas	Sources	Proximate analysis (wt%)				Ultimate analysis (wt%)					$Q_{net,v,ad}$ (MJ/kg)	Main mineral phases
			M_{ad}	V_{ad}	A_{ad}	FC_{ad}	C_{ad}	H_{ad}	O_{ad}	N_{ad}	S_{ad}		
S1	Datong	CHB	0.85	14.31	70.84	14.00	17.71	1.81	8.40	0.21	0.18	6.84	quartz, kaolinite
S2	Yangquan	CHB	1.56	10.96	68.75	18.73	17.64	1.70	7.88	0.40	2.07	5.20	quartz, kaolinite
S3	Shuozhou	CJB	1.65	17.59	58.59	22.17	24.27	2.41	12.49	0.43	0.16	9.28	kaolinite, illite
S4	Shuozhou	CHB	1.34	15.10	66.22	17.34	18.13	2.14	11.47	0.40	0.30	6.72	kaolinite, illite
S5	Jinzhong	CHB	1.32	10.38	75.58	12.72	12.62	1.44	4.57	0.18	4.29	4.38	quartz, kaolinite
S6	Jinzhong	CJB	0.78	17.17	59.11	22.94	26.63	2.07	9.51	0.31	1.59	7.59	calcite, kaolinite
S7	Luliang	CHB	1.13	11.29	77.00	10.58	11.36	1.62	7.84	0.19	0.86	4.00	quartz, kaolinite
S8	Luliang	CJB	0.94	12.37	67.74	18.95	19.78	1.95	7.25	0.31	2.03	9.98	quartz, kaolinite
S9	Changzhi	CJA	0.82	10.15	54.78	34.25	34.16	2.12	7.34	0.64	0.14	13.37	quartz, kaolinite
S10	Changzhi	CJA	0.84	9.61	55.59	33.96	34.01	2.04	6.76	0.64	0.12	13.11	quartz, kaolinite
S11	Neimongol	CJI	0.84	28.22	40.74	30.20	38.84	2.62	15.24	0.65	1.07	14.98	calcite, kaolinite

Note: CHB, coal gangue from heavy media separation for bituminous coal preparation; CJB, coal gangue from jigging process for bituminous coal preparation; CJA, coal gangue from jigging process for anthracite preparation; CJI, coal gangue from jigging process for lignite preparation.