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Pollutant emission characteristics and interaction during low-temperature oxidation of blended coal

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ABSTRACT

Pollutant emission characteristics and the synergistic interaction during low-temperature oxidation of blended coal were investigated. Two kinds of bituminous coals, high volatile bituminous coal (HC) and low volatile bituminous coal (LC), and their blends were heated from room temperature to 300 °C by 2 °C/min under air atmosphere, controlled and measured with the TGA–FTIR. The results showed that CO_2 emission was affected by both moisture content and pore structure of coal. The blended ratio of 60HC40LC inhibited CO_2 emission for the coupling effect of moisture content and pore structure. The minerals like sodium and potassium contributed to SO_2 generation, while calcium and magnesium inhibited SO_2 generation, thus the blended ratio of 40HC60LC inhibited SO_2 emission with the synergistic interaction. What's more, the blended coal could reduce HCN and HCl emission in different blended ratio. Thereby, blended coal technology was an effective way to reduce pollutant emission during low-temperature oxidation.

1. Introduction

With the rapid development of economy and improvement of living standard of China, the electric energy consumption is increasing. In China, the capacity of coal-fired power plant is account for about 70% of the national total generation capacity [1], thus coal is still the main fossil fuel of primary energy which is impossible to be replaced for a long time. However, because of the rising price of coal and the unbalance of supply and demand, many coal-fired power plants have import most of their coal besides the domestic coal [2]. Therefore, the coal types used in coal-fired power plants have been diversified and the quality has been further reduced.

To ensure adequate electric power supply, blending of different coal types is becoming common and promising in coal-fired power plant in recent years, especially in China. Coal-blend combustion technology is widely adopted for many advantages, such as improving combustion performance, meeting pollutant emission limits, controlling ash deposition, extending the range of acceptable coals, and reducing the fuel cost [1].

On the other hand, accumulation of heat of stockpiled coals leads to self-ignition [3,4] easily, which results in not only safety problem [5] and energy losses [6], but also uncontrolled pollutant gas emissions [7–9]. Low-temperature oxidation occurs whenever carbon-containing material is exposed to oxygen in the air. This process spans a wide range of temperature from room temperature to nearly 300 °C when ignition of coal occurring. As the temperature increasing, the organic matter and mineral substance of coal will become active and generate pollutants, including CO_2, SO_X, NO_X, CO and CH_4, etc. under the oxygen-enriched and oxygen-starved state. Many studies have found that, apart from mass transport considerations, the internal variables include composition and physical properties of coal, history of coal weathering/oxidation, as well as particle size, and the external variables involve temperature, partial pressure of O_2, and moisture content in the gas medium contribute to coal oxidation, and also have a great influence on the pollutant emission characteristics of coal [10].

Since the coal-blend combustion technology is still in development, and most of the current investigations are focusing on pollutant emission characteristics of individual coals during low temperature oxidation, the pollutant emission characteristics of blended coals has

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seldom been investigated. As the blending of coals is a popular way which is widely used in coal-fired power plants nowadays, further understanding about pollutant emission characteristics of blended coals and the interaction between the constituents during low temperature oxidation process, for instance, is of paramount importance. Therefore, the aim of this article is to study pollutant emission characteristics of blended coals and the interaction between different coals during low temperature oxidation under air atmosphere by TGA–FTIR.

The results could provide a detailed observation on pollutant emission characteristics of blended coals during low temperature oxidation, thus offering referential information for designing effective methods for suppressing the pollutant emission tendency of blended coals and perfecting co-combustion technology.

2. Materials and methods

A kind of high volatile bituminous coal from Indonesia (named HC) and a kind of low volatile bituminous coal from Inner Mongolia Autonomous Region of China (named as LC) were used as investigation objects in this paper. The proximate and ultimate analyses of two coals were shown in Table 1. The coal samples were pulverized and then passed through a sieve with a mesh size of 178 μm. The HC was added to LC at weight ratios of 20%, 40%, 60% and 80% (20HC80LC, 40HC60LC, 60HC40LC and 80HC20LC, respectively). The HC and LC were mixed a total amount of 40 g by a micro rotary mixer in all proportions for 3 h to ensure the uniformity of the blends, and then 10 g of the blends were performed for experiments. The blended samples were stored in the N_2 atmosphere for tests.

The experiments of low-temperature oxidation of coal were carried out via thermogravimetric analyzer (TGA) (METTLER TOLEDO TGA/DSC1). The temperature precision was ±0.5 °C and microbalance sensitivity was less than ±0.1 μg. A 15 ± 0.5 mg sample was placed on the sample pan quickly and loosely, then heating from room temperature to 300 °C in air atmosphere with a flow rate of 50 ml/min, at a heating rate of 2 °C/min.

The pollutant emission from low-temperature oxidation of blended coals was detected by a Fourier transform infrared measurements (Nicolet™ iS™ 10 FT-IR spectrometer) coupled with the TGA. Evolved gases from TGA passed through a heated transferred line into a beam conforming flow cell in the FTIR sample compartment and the FTIR spectra were collected for further processing. TGA and FTIR were connected by heated transfer line with a temperature of 215 °C in order to prevent the condensation of gases. FTIR spectra were collected with 4 cm^{-1} resolution, in the range of 4000–400 cm^{-1} IR absorption band. And the wavelength accuracy was less than 0.005 cm^{-1}. FTIR spectra of the gaseous production were collected continuously with the baseline corrected. According to the standard database of reference spectra (SDBS) and calibration curves, the absorbance spectra of CO_2 is at 2360 & 670 cm^{-1}; carboxyl at 1700 cm^{-1}; CO at 2119 cm^{-1}; SO_2 at 1342 cm^{-1}; S=O at 1440–1300 cm^{-1}; NO at 1762 cm^{-1}; HCN at 714 cm^{-1}; NH_3 at 966 cm^{-1} and HCl at 2798 cm^{-1}. All the experiments were carried out three times to decrease the test error, and the reproducibility was quite good.

In order to investigate if there was a synergistic interaction between blended coals, the calculated values of these blends were the weighted average of the individuals coal [11]:

$$W = \eta_1 W_1 + \eta_2 W_2 \tag{1}$$

where W was the calculated value of the pollutant amount of the blend, η_1, η_2 were the weight percentage of each individual sample in the blend and W_1, W_2 were the pollutant amount of each sample, respectively.

ΔW ($\Delta W = W_{\text{experimental}} - W_{\text{theoretical}}$) was defined as the pollutant amount difference between experimental value and theoretical value to further investigate the interaction.

3. Results and discussion

3.1. Pollutant emission characteristics

3.1.1. Emission characteristics of greenhouse gas and CO

The FTIR curves of CO_2 emission were shown in Fig. 1. Compared with all the blended coal, LC had the highest releasing temperature of CO_2. While HC had the lowest releasing temperature of CO_2. As shown in Table 1, LC had more moisture content than HC. The existence of moisture consumed large amount of heat for evaporation during the low-temperature oxidation process. Thus, the larger moisture content the blended coal had, the longer time it would take to dry the coal and start releasing CO_2. With the increasing ratio of LC in the blended coal, the releasing temperature of CO_2 delayed gradually, which were 63 °C, 75 °C, 120 °C, 125 °C, 181 °C and 200 °C for HC, 80HC20LC, 60HC40LC, 40HC60LC, 20HC80LC and LC, respectively. The releasing temperature of CO_2 of 80HC20LC, 60HC40LC, 40HC60LC, 20HC80LC and LC is 1.19, 1.90, 1.98, 2.87 and 3.17 times as that of HC, respectively. Moreover, the reactivity of chars enhanced at the same time. As shown in Fig. 1, the release rate of CO_2 increased dramatically with the increasing of moisture content of the blended coal, except for 20HC80LC and LC. The phenomenon above resulted from the moisture content in the blended coal. The increasing moisture content absorbed more oxygen, thus the following sequence of free-radical reactions would occur for auto-oxidation of coal.

Table 1
Proximate and ultimate analysis of coal (on air dried basis).

	Proximate analysis (wt%)				Ultimate analysis (wt%)					Lower heating value (MJ/kg)
	Moisture	Ash	Volatile matter	Fixed carbon	C	H	N	S	O	
HC	4.56	3.75	33.14	58.55	79.24	5.67	1.30	1.31	4.17	27.85
LC	8.45	8.44	38.7	44.41	68.07	4.64	0.82	0.62	8.95	25.75