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Research Paper

Combustion properties of coal gangue using thermogravimetry–Fourier transform infrared spectroscopy

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HIGHLIGHTS

- Five characteristic temperatures and mass losses of CG were investigated.
- Properties of the spontaneous combustion of CG were revealed through TG.
- Functional groups variation of CG was examining by FTIR under air atmosphere.
- S_n, D_i, and D_h values were calculated based on combustion performance method.
- S_n value increased with an increase in the $V_{ad} + FC_{ad}/A_{ad}$ value.

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ABSTRACT

This study investigated the combustion properties of coal gangue (CG) from the Gongwusu coal mine in northern China. Three CG samples collected from various parts of the spontaneous combustion gangue dump were evaluated using a proximate analyzer, thermogravimetry, and Fourier transform infrared spectroscopy. The results revealed that the total mass losses of the three samples were 15.5%, 30.3%, and 19.42%, which were consistent with the volatile and fixed carbon contents ($V_{ad} + FC_{ad}$). The calculated combustion performance indices (S_n) of CG for the three samples were 6.01×10^{-10}, 29.42×10^{-10}, and $15.65 \times 10^{-10}\%^2/(\min^2\ K^3)$. The S_n values increased with an augment of $V_{ad} + FC_{ad}/A_{ad}$. Sample 2 had a higher de-volatilization rate, burnout performance, and total mass loss, and it was easier to ignite during the spontaneous combustion process. The combustion performance of CG was more favorable when $V_{ad} + FC_{ad}$ was high and the ash content was low. In addition, the CG samples from the same mine area contained analogous hydroxyl, aliphatic, and oxygen functional groups, and aromatic compounds.

1. Introduction

Coal gangue (CG) is a mixture of organic and inorganic compounds and mineral materials [1], that are fused together with coal deposits during the coal formation process [2]. CG is a complex industrial solid waste generated from the coal mining and washing process [3]. Because of long-term stacking, spontaneous combustion frequently occurs in coal and carbonaceous materials [4]; this phenomenon has been reported worldwide [1,2,4–6].

Spontaneous ignition of gangue hills has generated numerous environmental and ecological hazards, such as toxic and harmful gas emissions that pollute the air [7] and heavy metal ions that contaminate the soil and water [8]. Moreover, toxic substances generated from CG hills, such as mercury, have threatened the health of people residing around and working in the mining industry zone [9]. In addition, the waste dump may lead to fire or explosion and cause casualties [6,10].

Coal production in China was 3.68 billion tons in 2015 [11], accounting for 47% of the global share. The discharge rate of CG reached approximately 10–15% of raw coal production [9], equivalent to 368–552 million tons. The total inventory of CG in China is estimated to be approximately 4.5–5.0 Gt [12], with an annual increase of 0.37–0.55 Gt [13]. Large amounts of waste are stacked to form a huge waste dump, which occupy large areas and are potentially hazardous [14–16]. More than 1600 CG hills are present in China, and over 300 of these hills have exhibited spontaneous combustion with smoke produced during the day and night. The western and northern regions of China are major coal-producing

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Nomenclature

DTG_{max}	maximum combustion rates (%/min)
t_p	corresponding time of DTG_{max} (min)
t_i	ignition time (min)
$\Delta t_{1/2}$	time range of DTG/DTG_{max} = 0.5 (min)
t_h	burnout time (min)
$(dw/d\tau)_{max}$	maximum rate of oxidation reaction at the peak (%/min)
$(dw/d\tau)_{ave}$	average rate of CG obtained by DTG area integration (%/min)
D_i	ignition index
D_h	burnout index
S_n	comprehensive combustion performance index
T_1	characteristic temperature of maximum evaporation of water and desorption of gases (°C)
T_2	characteristic temperature of maximum oxidization mass gain (°C)
T_3	characteristic temperature of ignition point (°C)
T_4	characteristic temperature of maximum mass loss rate (°C)
T_5	characteristic temperature of burnout point (°C)

areas, and the spontaneous ignition of CG hills in these areas has caused severe pollution [7,9].

Because of the growing public appeals for environmental protection, local governments and mining companies have made concerted efforts to govern spontaneous combustion of the CG hills. For example, in Wuhai, a city located in western Inner Mongolia, China and famous for its Wuda coalfield, which is a global hot spot for coal fire research, 80 of the total 196 ignition points in refuse discharge fields were extinguished by June 2016. Furthermore, of the entire fire area of 1.25 million $m^{2,}$ the fire management project processing reached 539,200 m^2. Although the fire control project was extremely expensive and time consuming, considerable efforts were made to extinguish 53 ignition points in the 199,300 m^2 area [17].

Several investigations have been conducted to elucidate the mechanisms underlying the spontaneous combustion of CG. Zhang et al. investigated the combustion behaviors and thermokinetic parameters of 11 CGs using thermogravimetry (TG) to evaluate the effects of feedstock properties [18]. Furthermore, Urbański and Walker have reported that the self-ignition spot is 1.5–2.0 m below the surface [19,20]. The phenomenon of spontaneous combustion during the initial stage occurs at 60–80 °C with organic and oxidized mineral materials [21]. Zhou et al. investigated thermal and trace-element partitioning behaviors during the cocombustion of biomass with CG [8,15]. In addition, Wang et al. reported five characteristic temperatures of six types of coal ranks at different oxidation stages [22]. Niu et al. examined the changes of the functional groups of Xundian coal using in-situ Fourier transform infrared (FTIR) spectroscopy [23]. Ran et al. examined the combustion and pyrolysis kinetic characteristics of CG under different conditions [24]. The coal-oxygen recombination theory has been accepted by numerous researchers [2]. According to this theory, a CG hill consists of a discontinuous porous medium. The oxygen penetrates into surface cracks, resulting in oxidation reaction with some active groups of gangue, and the heat generated in the oxidation process causes spontaneous combustion. However, ignition of a CG hill is a complex combustion system driven by internal and external factors. A uniform viewpoint on the mechanism underlying the spontaneous combustion of CG is not yet available.

In this study, we used simultaneous TG-FTIR spectroscopy to investigate TG, derivative thermogravimetry (DTG), FTIR, characteristic temperatures, and functional groups during the oxidation process of CG under air atmosphere. The study of CG and its oxidation characteristics is crucial for evaluating the relationship between the macroscopic properties of spontaneous combustion and the microstructure characterization of CG. Simultaneously, this study also provided an appropriate method for revealing the spontaneous chemical reaction and mechanism of CG in its spontaneous combustion process. Finally, this study offers evidence that can be applicable in the control engineering of self-heated colliery spoil heaps in China.

2. Experimental and methods

2.1. Sample preparation

CG samples were collected from the Gongwusu coal mine, located in Wuhai City, Inner Mongolia, China (Fig. 1). Fresh test samples were packed in bags and taken to the laboratory. Particles with a size between 80 and 120 mesh (0.125–0.180 mm) were sieved after pulverization. The selected gangue particles were then transferred into the reaction bed for testing under air atmosphere.

2.2. Proximate analysis

We determined the moisture, ash, volatile, and fixed carbon contents using the 5E-MAG6700 proximate analyzer (Qiulong Instruments, Changsha City, Hunan Province, China) according to the National Standards of China GB/T212-2008, which is equivalent to the American Standards of ASTM-D5142-2009.

2.3. TG-FTIR spectroscopy

The characteristic parameters of distinct oxidation stages of CG, such as the mass loss, mass loss rate, and characteristic temperature, were screened and tested through TG (Pyris 1 TG, PerkinElmer, Fremont, California, USA). Moreover, the related data of the functional groups of released gases in the heating process were scanned through FTIR spectroscopy (Spectrum 100, PerkinElmer, Fremont, California, USA), with a scanning range was from 4000 to 650 cm^{-1}. In addition, IR spectral peaks were analyzed using PeakFit® (Version 4.12). The method of second derivative minima was then used to detect hidden peaks in the spectra, and the peak type was reconstructed using the Lorentz–Gauss (amplitude) equation. The air flow rate was 20 mL/min and the heating rate was 20 °C/min from 30 to 900 °C.

2.4. Combustion performance method

The ignition index (D_i) and burnout index (D_h) are described in Eqs. (1) and (2), respectively [17]. In view of the applications, D_i and D_h can indicate the ignition and burnout performance of CG, respectively:

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