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Research paper

Influence of the hydrothermal dewatering on the combustion characteristics of Chinese low-rank coals

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HIGHLIGHTS

- Typical Chinese lignites with various ranks are upgraded by hydrothermal dewatering.
- Upgraded coals exhibit chemical compositions comparable with that of bituminous coal.
- FTIR show the change of microstructure and improvement in coal rank after upgrading.
- Upgraded coals exhibit difficulty in ignition but combust easily.
- More evident effects are obtained for raw brown coal with relative lower rank.

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ABSTRACT

This study investigates the influence of hydrothermal dewatering performed at different temperatures on the combustion characteristics of Chinese low-rank coals with different coalification maturities. It was found that the upgrading process significantly decreased the inherent moisture and oxygen content, increased the calorific value and fixed carbon content, and promoted the damage of the hydrophilic oxygen functional groups. The results of oxygen/carbon atomic ratio indicated that the upgrading process converted the low-rank coals near to high-rank coals which can also be gained using the Fourier transform infrared spectroscopy. The thermogravimetric analysis showed that the combustion processes of upgraded coals were delayed toward the high temperature region, and the upgraded coals had higher ignition and burnout temperature. On the other hand, based on the higher average combustion rate and comprehensive combustion parameter, the upgraded coals performed better compared with raw brown coals and the Da Tong bituminous coal. In ignition segment, the activation energy increased after treatment but decreased in the combustion stage. The changes in coal compositions, microstructure, rank, and combustion characteristics were more notable as the temperature in hydrothermal dewatering increased from 250 to 300 °C or coals of lower ranks were used.

1. Introduction

Low-rank coals (LRCs), which include lignite, brown coals and sub-bituminous coals [1], are an important feedstock for energy and chemical products. However, the current global coal market is dominated by high-rank coals (HRCs) which account for only half of the total coal deposits [2]. As the world's largest coal producer and consumer [3], China depends heavily on coal [4]. 70% of China's primary energy demand is supplied by coal, and coal-fired power plants account for over 97% in the total thermal power capacity [5]. Unfortunately, the country is rich in LRC reserves. The reserves of lignite are about 41.18% in the amount in China [1]. Therefore, the exploitation of LRCs is necessary for the growing energy demands and the depletion of HRCs.

The advantages of LRCs include high reactivity, abundance, and low market price [6], however, LRCs have not been utilized as much as HRCs [7] because they have many drawbacks, such as high moisture, low energy density, and high spontaneous combustion tendency [8]. In order to overcome these drawbacks and make LRCs' characteristics comparable with those of HRCs. LRCs should be treated by an upgrading process before use. The first step of the upgrading process is dewatering.

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Hydrothermal dewatering (HTD) process, also known as hot water drying, is a typical non-evaporative dewatering process. HTD has two major advantages: it can remove water in the liquid form, saving the latent heat of vaporization; it can damage oxygen functional groups, preventing the upgraded products from reabsorbing water. Due to its high temperature and pressure, HTD has been considered as the simulation of the coal forming process in nature.

The literature contains some investigations about the effects of HTD processing variables on the properties of the final products [9,10]. Morimoto et al. [11] studied the yield of the products during the HTD process, and found that the upgraded coal showed less oxygen content, higher calorific value, and higher gasification reactivity than the raw coal. Yu et al. [12] studied the effect of HTD on the slurryability of brown coals, and found that the maximum solid concentration of coal water slurry increased. Wu et al. [13] studied the gas-, solid-, and liquid-phase products obtained after HTD, and analyzed the occurrence of carbon and its migration mechanism during HTD. In recent years, there has been a growing interest of HTD in coal treatment and a number of potential applications of hydrothermal processing have been investigated, such as preparing coal water slurry with the solid and liquid products of HTD [14], improving the space time yield of brown coal liquefaction reactor [15], upgrading and dewatering of raw tropical peat [16], dewatering of sludge [17], generating alternative solid fuel from paper sludge [18], and producing liquid and solid biofuels [19,20].

However, only few studies investigated the changes in coal chemical properties and microstructures during the HTD upgrading process, which are closely related to some combustion problems. For example, the less volatile matter release may lead to an unstable flame, and the oxidation of carbon may result in a decrease in the burnout efficiency. In this context, the combustion characteristics of LRC before and after the upgrading process should be compared to improve burning efficiency, reduce emissions, and evaluate the design and construction of a boiler.

In this study, three types of brown coals (with different coal ranks) produced in China were processed by HTD at different temperatures. The changes in the coal composition and microstructure of LRC by HTD were investigated with emphases on the improvement of coal rank and on the influence of HTD on the combustion characteristics of LRC.

2. Experimental

2.1. Coal sample

Three brown coals, namely, Zhao Tong (ZT), Yi Min (YM), and Zhun Dong (ZD), were investigated in this study. They were formed in different geological ages (ZT in Tertiary, YM in late Jurassic, and ZD in early Jurassic) and produced in three different areas in China. The HRC used in this study was Da Tong (DT) bituminous coal. The raw coals were crushed and sieved through 2.5-mm meshes before the upgrading process. The raw and upgraded coal samples were milled for 30 s in a ring mill and sieved through 74-μm meshes for the analysis.

2.2. HTD upgrading process

HTD process was performed in a bench scale hydrothermal reaction system (WeiBa WHFS-2), which had a 2-L cylindrical autoclave with the maximum pressure of 25 MPa and the maximum temperature of 350 °C [12], as shown in Fig. 1. A mixture of brown coal and deionized water (under a proportion of 1:2.5) was added into the autoclave. The sealed autoclave was injected with N_2 to remove air and the pressure in the autoclave was set to be 4 MPa for 2 h to ensure the absence of leaks. After the N_2 was released, the temperature in the autoclave was increased by 3 °C/min up to the preset values (250 and 300 °C) and maintained for 1 h. The mixture was stirred at 100 rpm during the entire process. The autoclave was cooled to ambient temperature by an internal cooling coil with tap water after the reaction was completed. The upgraded coals were separated from the mixture by a qualitative filter paper and dried in air for approximately one week. The dried updated coals were then milled. All upgraded coals were abbreviated to “initials-treatment temperature”, such as “YM-250” and “YM-300”.

2.3. Coal composition analysis

The C, H, N and S contents of the coal samples were measured using an LECO-CHNS 932 Elemental Analyzer, whereas the O content was calculated by difference. The proximate analysis data was determined using a 5E-MACIII Infrared Rapid Analyzer, and the calorific value (CV) was obtained using the adiabatic bomb calorimetric method following ISO1928.

2.4. Fourier transform infrared spectroscopy (FTIR)

All the spectra of both raw and upgraded coals were generated via standard procedures with KBr pellets by using a Nicolet NEXUS-670 FTIR instrument. The measuring region ranged from 4000 to 400 cm^{-1}, and the spectra were generated by collecting 32 scans at a resolution of 4 cm^{-1}. Approximately 1 mg of ground coal was mixed with 100 mg of dried KBr powder, and the mixture was further ground with a mortar and a pestle under a baking lamp to avoid water absorption. The ground mixture was then pressed into a pellet with a hydraulic tabletting machine. OMNIC software 6.1a (Nicolet) was used for data preprocessing, and the spectral analysis processes were performed using the professional software PeakFit® (Version 4.12).

2.5. Thermo-gravimetric analysis (TGA)

TGA analysis was conducted using the non-isothermal method in a TGA-SDTA 851e apparatus produced by METTLER TOLEDO. 5 mg of coal sample were placed in an aluminum crucible and heated at 25 °C/min to the temperature until the weightlessness curve unchanged. The flow rate of standard air was kept at 60 mL/min.

The combustion characteristics investigated in this study include ignition (T_i), burnout (T_b) and peak (T_{max}) temperatures, and the maximum (k_{max}) and average (k_{mean}) combustion rates. They reflect the thermal behavior and burnout property during the combustion process and can be derived from the thermogravimetric and differential (TG-DTG) curves. Therefore, they were adopted to evaluate the combustion process. An index S, which is defined in Eq. (1) was used to compare the combustion characteristics of different coal samples. The coals with a higher S value have better combustion performance.

$$S = \frac{k_{\max} \times k_{mean}}{T_i^2 \times T_b} \tag{1}$$

3. Results and discussion

3.1. Influence of upgrading on coal composition

The moisture content (M_t) of the raw brown coals not processed by HTD—53.11 wt% for ZT, 30.92 wt% for YM, and 24.94 wt% for ZD—is dramatically larger than that of DT bituminous coal (4.59 wt