



Full Length Article

An experimental study on the effect of igneous intrusions on chemical structure and combustion characteristics of coal in Daxing Mine, China

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ABSTRACT

To investigate the effect of igneous intrusions on the structure and combustion characteristics of coal, the normal bituminous coal and thermally altered coal were collected from Daxing Mine in China. The chemical structural features of coal were characterized by X-ray diffraction (XRD) and Fourier transform infrared spectroscopy (FTIR). Structural parameters determined from XRD analyses, including the interlayer spacing of the crystallite (d_{002}), stacking height (L_c), aromaticity ($f_{a(XRD)}$), and coal rank (R_{XRD}), revealed that the altered coal showed more crystalline carbon and higher coal rank than the normal bituminous coal. FTIR analyses indicated that igneous intrusions increased the apparent aromaticity of the altered coal and elevated its maturation level, which was consistent with the XRD results. TG-DSC experiments were conducted to study the combustion characteristics of coal. The result showed that the combustion process of the altered coal was delayed to high-temperature region. Compared with the normal coal, the altered coal exhibited lower combustion reactivity and worse combustion performance, as evidenced by lower comprehensive combustion index, higher ignition temperature and burnout temperature, as well as larger apparent activation energy. Moreover, igneous intrusions decreased the heat release and calorific value of the altered coal during combustion, which was ascribed to the changes in chemical composition and structure of the altered coal. This study concluded that igneous intrusions increased the difficulty in igniting the altered coals and reduced its heat release during combustion.

1. Introduction

Coal is a major energy source for electricity supply, and the coal consumption will increase from 29.9% to 46% of global electricity generation by 2030 [1,2]. According to the latest forecast of International Energy Agency, coal will be the largest source of worldwide electricity supply until 2040. Since the large demand for coal, the mining depth continuously increases due to the exhaustion of shallow coal reserves, and more thermally altered coals affected by igneous intrusions are mined and exploited in many coalfields around the world [3–10]. Compared with coal undergone normal coalification, the thermally altered coal showed distinct geochemical signatures in structures and properties [3,11]. Most of the existing studies mainly focused on the pore structure and adsorption property of the thermally altered coal due to the frequent accidents of coal and gas outbursts, which indicated that igneous intrusions promoted the development of pore structure and improvement of adsorption capacity of coal, consequently increasing the risk of gas outbursts [6,7]. In contrast, little attention has been

devoted to investigate the effect of igneous intrusions on the chemical structure and combustion characteristics of coal.

The effects of igneous intrusions on coal have several similarities to the artificial pre-treated process, such as pyrolysis, microwave irradiation treatment, hydrothermal treatment. As shown in previous literatures [5,12–16], both of igneous intrusions and artificial pre-treated process decreased the moisture and volatile matter contents in the treated coal, and increased its fixed carbon and ash contents. Many papers have reported the effect of artificial pre-treated process on coal structure and properties. Ohki et al. [12] investigated the effect of hydrothermal treatment at 200–350 °C on the low-rank coal, and concluded that the raw coal and hydrothermally treated coal showed obvious differences in the properties and combustion characteristics. To remove the inherent moisture in coal, Ge [14] and Wang [16] used microwave irradiation to treat the low-rank coal, concluding that microwave irradiation reduced the moisture in the treated coal, and increased its calorific value. Nevertheless, unlike the aforementioned artificial process, igneous intrusions provided the special conditions for

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coal seams, including the high temperature and high pressure, as well as the epigenetic minerals from hydrothermal fluids, which may show the different influence on coal properties. Our previous study has revealed that igneous intrusions increased the oxygen adsorption and oxidation reactivity of the altered coals during low-temperature oxidation ($< 200\text{ }^{\circ}\text{C}$), mainly because of the development of pore structure [17]. However, the oxidation reactivity of coal may not represent its high-temperature thermal behavior and combustion characteristics. Typically, Avila [18] and Meng [19] have found that some coals showed high oxidation reactivity, while these coals demonstrated the low combustion reactivity during the combustion period. To the best of our knowledge, the changes in chemical structure and high-temperature combustion behavior of coal under the impact of igneous intrusions has not been investigated, which was very closely related to many combustion problems. For example, the less volatile matter release could result in the unstable combustion flame, and the oxidation of carbon in coal would lead to the decrease in the burnout efficiency. The comparative study on the structural features and combustion characteristics of the normal coal and thermally altered coal is highly necessary to increase the burning efficiency, control the flame stability, and reduce emissions during coal combustion.

In the present study, the normal coal and thermally altered coal samples were collected from Daxing Mine in China. We studied the effect of igneous intrusions on chemical structure and combustion characteristics of coal. The research findings could provide a theoretical basis for the efficient utilization of the altered coal by igneous intrusions.

2. Materials and methods

2.1. Study area and samples preparation

Daxing Mine is affected by large-scale igneous intrusions and located in the southwest of Tiefa coalfield, which is the most important production base of bituminous coal in Liaoning Province, China. Daxing Mine has the length of 6.4 km and width of 3.2 km, and the designed production capacity is 3.0 million tons per year, with four mining areas including North No. 1, North No. 2, South No. 2, and South No. 5. Now, Daxing Mine has two coalfaces, South No. 5 709 coalface ($S_5\ 709$) and South No. 2 905 coalface ($S_2\ 905$), which are severely effected by igneous intrusions.

Four coal samples were collected from Daxing Mine and the schematic diagram of sampling locations were shown in Fig. 1, including the normal coal (Sample #1) and the altered coal (Sample #2) from $S_5\ 709$ coalface, as well as the normal coal (Sample #3) and the altered coal (Sample #4) from $S_2\ 905$ coalface. These coal samples were milled to pass a 200 mesh sieve for analysis. The proximate and ultimate analyses were showed in Table 1.

2.2. Analysis methods

XRD patterns of coal samples were observed by a Bruker D8 ADVANCE X-ray diffraction machine with Cu $K\alpha$ radiation (40 kV, 350 mA) and a scanning rate of 0.2 s/step from 10° to 80° .

The in-situ series diffuse reflection FTIR was applied to investigate functional groups and chemical structure of coals using a Nicolet 6700 FTIR spectrometer. Before analyzing the coal, the pure KBr powder background was collected as a baseline reference. The infrared spectra were recorded by collecting 64 scans with the resolution of 4 cm^{-1} over the range of $4000\text{--}500\text{ cm}^{-1}$.

A thermogravimetric analyzer (NETZSCH STA 449 F5 Jupiter) was employed to study the combustion characteristics of coals. Firstly, the coal samples approximately 10 mg were placed in ceramic pan for each run and heated from room temperature to $800\text{ }^{\circ}\text{C}$ at a constant heating rate of $10\text{ }^{\circ}\text{C}/\text{min}$ with the dry air flow rate of 100 ml/min. The TG and DTG signal as a function of temperature was recorded continuously

with increasing the coal temperature. Moreover, DSC curve was used to analyze the thermal behavior during coal combustion. Based on TG and DTG curves, ignition temperature (T_{ig}), peak temperature (T_{max}) and burnout temperature (T_b) was obtained to describe the combustion process of coal samples. T_{ig} is the temperature at which coals start to burn and it can be determined by TG/DTG tangent method [20,21], as shown in Fig. 2. T_{max} is the point of the maximum reaction rate corresponding to the top peak of DTG curve. T_b is the temperature at which weight loss fraction of coals reach 98%.

3. Results and discussion

3.1. X-ray diffraction analysis of coal samples

As shown in Fig. 3, it could be clearly found that some minerals existed in these coals, mainly including quartz, kaolinite, and calcite. The characteristic peaks of kaolinite and quartz significantly existed in the normal coals along with carbon based peaks, whereas the peaks of calcite were especially observed in the altered coals. This was because the reaction between materials from hydrothermal fluids and generated CO_2 during igneous intrusions caused the precipitation of calcite to fill pores and veins in the altered coals, increasing the content of epigenetic minerals in these coals [17,22,23]. All profiles exhibited the high background intensity, which indicated that coal samples contained disordered structures and amorphous carbons. Moreover, these coals also contain layered graphite-like structures as evidenced by the existence of (0 0 2) band at $\sim 25^{\circ}$. Due to the asymmetry of the (0 0 2) band, there was another γ -band at $\sim 20^{\circ}$ on the left side of (0 0 2) band. The characteristics of (0 0 2) band reflected the spacing of aromatic ring layers, while γ -band showed the packing distance of saturated structures in coals [24]. However, Fig. 3 did not show the clearly (1 0 0) band at $\sim 42^{\circ}$, which may be ascribed to the high levels of background, indicating low levels of growth in the basal planes of graphite structures in coals.

The structural parameters of coals including the interlayer spacing (d_{002}) and stacking height (L_c) of the crystallite were calculated using Eqs. (1) and (2) [25]:

$$d_{002} = \frac{\lambda}{2\sin\theta_{002}} \quad (1)$$

$$L_c = \frac{0.89\lambda}{B_{002}\cos\theta_{002}} \quad (2)$$

where λ is the wavelength of X-ray (0.154 nm for Cu $K\alpha$ radiation), B and θ refer to the full width at half-maximum and position of (0 0 2) peak, respectively. The aromaticity ($f_{a(XRD)}$) and coal rank could be calculated by the areas (A_{002} and A_{γ}) and peak intensity (I_{002} and I_{γ}) of (0 0 2) band and γ -band, as follows [26,27]:

$$f_{a(XRD)} = \frac{C_{ar}}{C_{ar} + C_{al}} = \frac{A_{002}}{A_{002} + A_{\gamma}} \quad (3)$$

$$R_{(XRD)} = \frac{I_{002}}{I_{\gamma}} \quad (4)$$

XRD pattern and the broad hump in the 2θ region of $16\text{--}33^{\circ}$ was fitted to the Gaussian peaks mainly including γ -band at $\sim 20^{\circ}$ and (0 0 2) band at $\sim 25^{\circ}$, while other small peaks affected by mineral compositions were treated as residuals. The fitted XRD patterns of coal samples are shown in Fig. 4. Based on the peak information, the structural parameters were calculated and showed in Table 2. The d_{002} of four coal samples varied from 3.520 to 3.585 Å, much higher than the d_{002} of pure graphite ($\sim 3.37\text{ Å}$), which exhibited the low degree of crystalline order in coal samples. However, compared with d_{002} values of the normal coals, lower d_{002} values could be found in the altered coals, decreasing from 3.585 to 3.540 Å and from 3.566 to 3.520 Å, which indicated higher crystallinity for the altered coals under the

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