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Full Length Article

Experimental study on characteristics of pressurized grade conversion of coal

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ABSTRACT

In order to investigate the characteristics of pressurized grade conversion of coal, an experimental system was established, and the general properties of coal and semi-coke were studied by using the BET and SEM methods. Then, the effects of the following variables on production yields, pollutant gas emission, and the quality of high-grade raw materials such as tar and coal gas were investigated in the ranges as specified: pyrolysis temperature (500–700 °C), pressure (0.1–0.5 MPa), pyrolysis atmosphere (N_2 and coal gas) and particle size (0–3 mm and 0–6 mm). It was found that when the coal pyrolysis temperature is 600 °C, the tar yield reaches its maximum, and the coal gas atmosphere and pressure are both helpful to increase the tar and semi-coke yield as well as to improve the quality of tar and the heat value of coal gas. With the pressure increasing, the NO and SO_2 emissions decrease. As a result of the pressure increase, the combustion temperature of the semi-coke decreases, resulting in the reduction of high temperature minerals and the increase of low temperature minerals in ash. The results of this study will be useful to address some of the presently existing issues in design, optimization and scale up of pressurized grade conversion of coal processes.

1. Introduction

Compared to other fossil fuels, coal is expected to be the main energy sources of the future in China due to its largest accessible reserves and low price [1]. In the year 2016, coal accounted for more than 60% of China's primary energy consumption and is projected to remain the economical choice for power plants and investors. It can be expected that the energy structure of China will still be coal-based in the next few decades [2]. Therefore, the technical development for clean and efficient utilization of coal aiming at reducing the air pollution and increasing the coal utilization efficiency is critical to the future energy security and development in China [3].

In the past decades, most of the coals (especially the low-rank coal) combusted directly in a thermal power plant, resulting in problems such as high pollution and low efficiency. These problems have become major barriers to the utilization of clean coal technology [4]. In order to solve these problems, pressurized grade conversion of coal has been proposed in recent years [5]. This technology was based on the grade utilization of coal in which the thermochemical reaction of coal is the coupling of low temperature pyrolysis of coal and semi-coke combustion, the pyrolysis gas and tar are extracted as high-grade raw materials, while the semi-coke obtained can be used for power generation and supplying heat for coal pyrolysis [6,7]. When the system pressure is increased, the interaction between gas and particles is enhanced, and the reaction rate and efficiency of each process will be accordingly changed. Moreover, the production yields are altered with the change of pressure, so as to obtain raw materials of different high-grade. In the past, various studies have been reported on the mechanisms and pilot scale test of semi-coke combustion and coal pyrolysis, especially under the condition of atmospheric pressure [8–12]. However, to date, the dependence of the characteristics of pyrolysis or combustion, such as product yields and pollutant emissions on pressure has not been clearly established. As a result, there are difficulties in design, optimization and scale up of these processes.

To overcome these gaps, this paper presents a systematic investigation of the characteristics and mechanisms of pressurized grade conversion of coal, including coal pyrolysis process and its semi-coke combustion process. In particular, BET and SEM methods were first used to study the general properties of coal and semi-coke. Then, a pressurized conversion experimental system was established to

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Table 1
Proximate and ultimate analysis of HeiLonggou coal.

Item	Content (wt%) Value (MJ/kg)
Moisture[†]	10.26
Volatile[†]	34.42
Fixed carbon[†]	50.24
Ash[†]	5.08
Carbon[§]	70.39
Hydrogen[§]	5.175
Oxygen[§]	22.4
Nitrogen[§]	1.032
Sulphur[§]	1.003
Low heating value[a] (MJ/kg)	27.508

[†] Proximate analysis.
[§] Ultimate analysis.
[a] As received.

investigate the mechanism and characteristics of coal pressurized grade conversion. Focuses were the effects of pyrolysis temperature, pressure and particle size on production yields, pollutant gas emission, and the quality of raw materials such as tar and coal gas characteristics.

2. Experimental

2.1. Materials

As a low-rank coal, HeiLonggou coal was chosen as the experimental material in this paper. Proximate and ultimate analysis of the coal is listed in Table 1. In order to investigate the effect of different particle size on the behaviour of pressurized grade conversion process, two particle size distributions which are commonly used in industrial production were used, and the details are shown in Fig. 1.

2.2. Structural characterization

The structural characterizations of the coal and semi-coke were carried out by SEM analysis and gas adsorption. The semi-coke samples were prepared under different pyrolysis pressures (0.1, 0.3 and 0.5 MPa) and temperatures (500, 600 and 700 °C) under the atmosphere of both N_2 and coal gas.

2.3. Grade conversion experiments

An electrically heated bench scale pressurized grade conversion system shown in Fig. 2 was used to study the mechanism of pressurized grade conversion of coal. The coal sample was pulverized and sieved to 0–3 mm and 0–6 mm sizes prior to the next procedure. Pressurized pyrolysis experiments were first carried out in N_2 and coal gas atmospheres, respectively. The reactor was fed with 5 g coal sample, and then sealed. The pressure was set to constant values of 0.1, 0.2, 0.3, 0.4 and 0.5 MPa by adjusting the valves 4 and 9 in Fig. 2, and the inlet carrier gas was set to 200 ml/min. At the beginning of the pyrolysis, the furnace was heated to a fixed target pyrolysis temperature, which was varied in steps such as 500, 550, 600, 650, and 700 °C at a given rate of 10 °C/min, each target temperature being maintained for 30 min. When pyrolysis process finished, the furnace was cooled down to the room temperature in the same atmosphere. While keeping the same pressure, the carrier gas was switched to air, and the furnace was heated to the combustion temperature of 1000 °C at the rate of 10 °C/min. The furnace temperature was maintained at 900 °C for 30 min during the semi-coke combustion process. The emissions of gas species during the pyrolysis and combustion processes were measured by two gas analyzers, respectively. A tar filter was used to remove tar from pyrolysis gas. A list of experimental conditions is shown in Table 2.

3. Results and discussion

3.1. General properties of coal and semi-coke

First of all, semi-coke samples produced under different conditions were prepared and their proximate analyses carried out as presented in Fig. 3. It can be seen from Fig. 3 that after pyrolysis, the heat value of semi-coke has increased. This is mainly because the moisture content has been evaporated, the O content decreased and most of volatile matter was released. This phenomenon is named as upgrade in some previous studies. Due to the volatile matter released, the fixed carbon content has increased with the increase of pyrolysis temperature while it decreases as the pressure increases. There is no significant difference in the proximate analysis results of the semi-coke samples of different particle sizes under different atmospheres. These data will provide a basis for further mechanism experiments.

Scanning electron microscope (S-3400N II) manufactured by Hitachi Co., Ltd. Japan was used to observe the surface topography characteristics of coal char, and SEM micrographs of some representative samples are shown in Fig. 4. The specific surface area (S_{BET}), total pore volume (TPV) and average pore diameter (APD) of the samples are shown in Table 3. It was found that the APD of semi-coke is smaller than that of raw coal because the pore structure of semi-coke is developed, and S_{BET} increases more than the total pore volume (TPV), resulting in a decrease in the average pore size. According to Fig. 4, the surface of Heilonggou coal is relatively smooth and compact, with just a

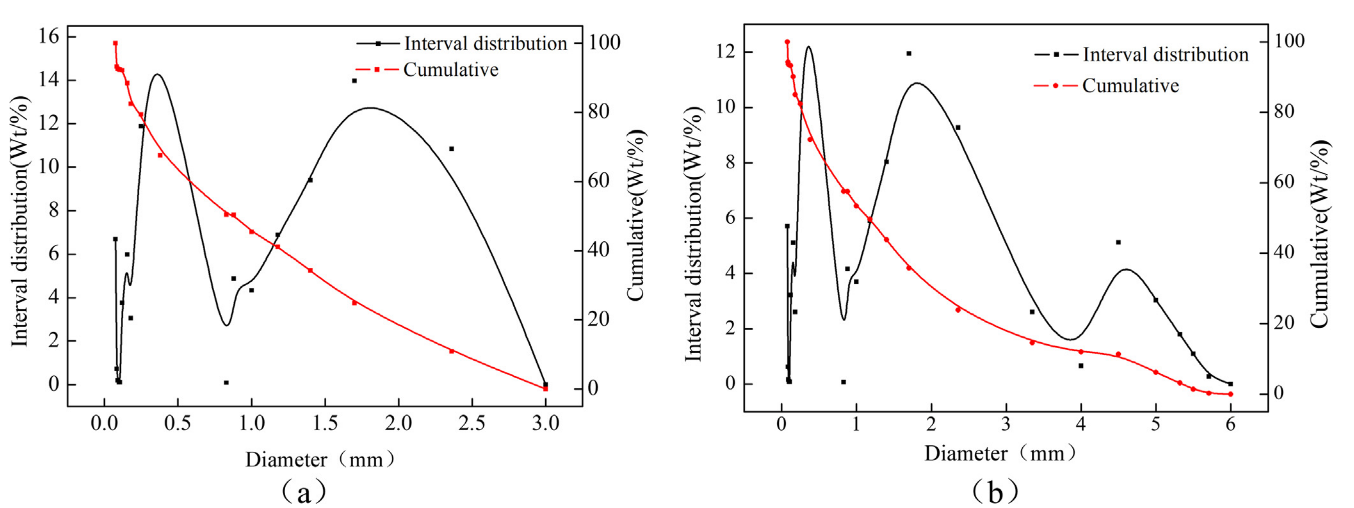

Fig. 1. Details of particle size distributions (a) 0–3 mm, and (b) 0–6 mm.