Fuel 223 (2018) 414–421

Contents lists available at ScienceDirect

Fuel

journal homepage: www.elsevier.com/locate/fuel

Full Length Article

Petroleum coke facilitate the upgrade of lignite under microwave irradiation for slurryability improvement

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ARTICLE INFO

Keywords:
Lignite
Petroleum coke
Microwave
Lignite water slurry

ABSTRACT

To remarkably reduce moisture and volatile content of lignite for the preparation of lignite water slurry (LWS) with high solid concentration, different additions of petroleum coke (PC) were used to increase the heating rate of microwave irradiation. After that the physical and chemical property changes of upgraded lignite were investigated through X-ray photoelectron spectra (XPS) and low temperature N_2 adsorption method. The results show that: the coal rank increased, the oxygen-containing functional groups decreased remarkably, the specific surface area and pore volume increased, the average pore diameter decreased, and the macropores gradually transformed to mesopores and micropores. Furthermore, these changes of upgraded lignite reached their maximum when 10 wt% PC was added to facilitate microwave pyrolysis. The maximum solid concentration increased from 51.60 wt% to 66.87 wt% and further increased to 67.51 wt% when the 10 wt% PC was mixed with upgraded lignite to prepare LWS. The LWSs prepared from upgraded lignite showed a weaker shear-thinning characteristic and lower static stability.

1. Introduction

Due to the abundant lignite (L) reserves (accounting for about 13% of the total coal reserves) in China, lignite-derived fuels may play an indispensable role in China's future energy demand [1,2]. However, direct shipping of lignite is not economic and safe because of its high moisture and volatile content, and low calorific value [3]. Preparing lignite into lignite water slurry (LWS) and then converting it to other fuel products is one of the most economic and safe ways for lignite further utilization. Its high inherent moisture and volatile content, developed pore structures, and abundant oxygen-containing functional groups makes the solid concentration of LWS quite low [4]. Therefore, exploring economic and effective ways to improve the solid concentration of LWS is the premise for further utilization of lignite reserves.

Some researchers have conducted into the preparation of LWS with upgraded lignite to improve its solid concentration. Microwave technology, a promising lignite upgrade method, has been used to dehydrate and make pyrolysis of lignite [5–12]. Compared with the conventional dehydration and pyrolysis techniques it is real-time, rapid, selective and efficient, which is because it forms the same direction of temperature gradient, pressure gradient and moisture migration, promotes the moisture migration and shortens the dehydration time [13]. From coal drying to coal pyrolysis, investigators have found promising advantages of microwave technology in future coal industry. Seehra [14] found that the efficiency of microwave dehydration is an order of magnitude higher than that of thermal heating. The internal high temperature and pressure from microwave irradiation could remove large amount of moisture in lignite, destroy the pore structures and remove some oxygen-containing functional groups of lignite at the same time. These variant changes on the physical and chemical properties of lignite after microwave irradiation improved its slurryability [15]. Zhu et al. [16] found that the maximum solid loading of lignite increased by 4.55% after being irradiated at 900 W for 6 min. Under a tunnel-type microwave oven Cheng et al. [17] increased the LWS solid concentration from 46.6 wt% to 54.0 wt% (viscosity is 1100 mPa.s at a shear rate of 100 s^{-1}). Wang [18] discovered that microwave irradiation could be employed to various lignite to improve the slurryability. Besides, Binner et al. [19] mentioned various other potential applications of microwave energy in coal processing and utilization, such as demineralization [20], grindability improvement [21], coke making [22], and coal liquefaction [23,24].

Some other researchers mixed L and petroleum coke (PC) to prepare mixed slurry with high solid concentration and good static stability [25–29]. As a carbonaceous byproduct of petroleum refineries, PC has led to an increasing interest in its utilization in recent years [30–33].

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https://doi.org/10.1016/j.fuel.2018.03.001
Received 11 December 2017; Received in revised form 26 February 2018; Accepted 1 March 2018
Available online 20 March 2018

Because its high calorific value, high carbon content and low ash content, PC is a desirable fuel. However, it is not economic and environmentally friendly to burn directly as a fuel source because of the difficulty in transporting the solid material and the high SO_x emissions (it can contain 5 wt% sulfur) [34]. Thus, preparing L and PC into mixed slurry for combustion or gasification can effectively meet the environmental regulations.

Recently the lignite microwave irradiation process can be further promoted through adding susceptors, including carbon materials (e.g., activated carbon [1], graphite [1,35], coke [36]), and some inorganic metal oxides (e.g., Fe_3O_4 [37], CuO, V_2O_5 and WO_3 [36]). However, the former is expensive and must be separated from coal for the preparation of coal water slurry; the latter needed to be separated from upgraded coal and could add extra inorganic component to the solid residues obtained after microwave irradiation. Thus, it is principal to find a relatively proper and economic susceptor to upgrade lignite under microwave irradiation, and directly prepare upgraded lignite and susceptor into mixed slurry with high solid concentration. As PC was employed to improve the solid concentration of LWS, it is also a strong microwave adsorbing material with a high dielectric constant of 67.38 ~ 44 [38], and its dielectric loss factor increases with increasing temperature, resulting in an increased ability to absorb microwave energy. Therefore, using PC as lignite microwave irradiation susceptor and preparing PC and upgraded lignite into mixed slurry for combustion or gasification can effectively use them and meet the environmental regulations. On the one hand PC is inexpensive, easily available in different textures, sizes, forms, etc.; on the other hand, without separating the petroleum coke susceptor from the upgraded lignite after microwave irradiation and directly preparing them into mixed slurry could save the trouble of separating and remarkably increase the solid concentration of LWS. Overall, it is feasible to facilitate lignite microwave irradiation process and improve the solid concentration of LWS through using petroleum coke as microwave susceptor. So, the aim of this study is to find a relatively proper addition of PC to upgrade lignite under microwave irradiation, and directly prepare upgraded lignite and PC susceptor into mixed slurry.

2. Experimental

2.1. Materials

Lignite from Inner Mongolia and PC from a petrochemical plant (Foshan, Guangdong Province) were used in this investigation. Lignite sample was crushed and sifted to a particle size of 0.3–3 mm, petroleum coke was crushed and then ground to a particle size less than 0.15 mm.

2.2. Experimental methods

2.2.1. Microwave pyrolysis process

Microwave pyrolysis experiments were carried out in a microwave oven with a frequency of 2450 ± 50 MHz, a maximum power output of 1800 W, and a multimode cavity dimensions of 200 mm * 180 mm * 260 mm. L and PC were mixed uniformly and loaded into a perfectly sealed quartz reactor with a nitrogen gas inlet, a gas outlet and a temperature measurement hole. Nitrogen gas inlet viscosity was controlled at 0.4 L/min to prevent lignite combustion and promote generated gases release during microwave irradiation. A K-type microwave-shielded thermocouple was inserted in the lignite sample to measure the real-time coal sample temperature. 120 g L with different PC additions (0g, 6 g, 12 g, 18 g, 24 g and 30 g respectively) were mixed evenly in a quartz bottle and irradiated under a microwave power of 1800 W for 30 min to investigate the effects of different PC additions on the microwave upgrading characteristics of L (the lignite temperature was controlled around 700 °C). Collect the residual solid samples (defined as S_0, S_5, S_{10}, S_{15}, S_{20}, S_{25}) separated from PC (with the sieving method) to measure the physical-chemical properties after microwave irradiation. And collect the gas products that emitted from lignite with aluminum foil packaging bags. Gas constituents were analyzed using a gas chromatograph (Agilent 7890A GC, USA). The L with upgraded physicochemical properties was finally used to prepare LWS with high solid concentration.

2.2.2. Preparation and determination of LWS

To avoid the inflation of LWS, all the upgraded lignite samples were vacuumed and heated in a vacuum oven for 6 h before the LWS preparation. KY33, composed of sodium salt of naphthalene sulfonate formaldehyde condensate (NSF), sodium carbonate and other additives, was used as a dispersant to prepare LWS. Then a certain mass of upgraded lignite (pulverized to 90% less than 0.074 μm) was mixed together with 1 wt% KY33 (based on the weight of dried lignite) and deionized water, then stirred together the mixture by using low-shear-velocity blend (300 r/min) for 4 min. After that the mixture was agitated using high-shear-velocity stir (1000 r/min) for 15 min to prepare LWS samples. The solid loading and the viscosity of LWS were analyzed with an automatic moisture analyzer (Sartorius MA35) and a viscometer (NXS11-B) respectively [39]. To meet the requirements of pumping, the viscosity of LWS should be no larger than 1200 mPa·s at a shear rate of 100 s^{-1} [40,41]. After LWS preparation, the slurry is filled into a 55 cm Turbiscan tube and placed into Turbiscan LAB™ to objectively characterize and analyze the changes in its static stability in 7 days. And the Turbiscan stability index (TSI) are used to quantify the stability of LWS samples [42].

2.2.3. Chemical properties measurements

Proximate and ultimate analysis of the PC and L (before and after microwave irradiation) were carried out to analyze the chemical characteristics of solid particles. A PHI Quantera (ULVACPHI, Japan) equipped with a hemispheric detector was employed for the XPS measurements.

3. Results and discussion

3.1. Heating characteristics of lignite

Fig. 1 shows the influence of PC addition contents on lignite temperature variation under microwave irradiation. The lignite temperature variation can be divided into four periods: under 100 °C, 100 °C to 180 °C, 180 °C to 700 °C and around 700 °C. During the first heating period (under 100 °C), the moisture, having high dielectric constant, mainly adsorbed the microwave energy, thus the heating rates of six samples were similar. After large amount removal of moisture, the

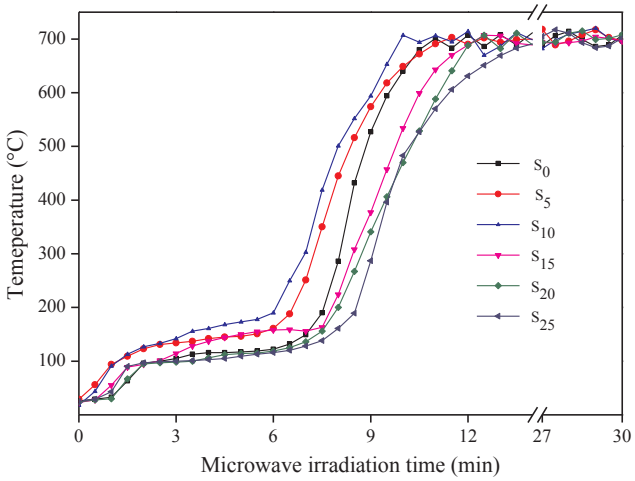

Fig. 1. Effect of PC contents on the heating characteristics of lignite under microwave irradiation.