Fuel 218 (2018) 275–281

Contents lists available at ScienceDirect

Fuel

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

Full Length Article

Improving physicochemical properties of upgraded Indonesian lignite through microwave irradiation with char adsorbent

Fan Zhou[a,b], Jun Cheng[a,*], Jianzhong Liu[a], Junhu Zhou[a], Kefa Cen[a]

[a] State Key Lab of Clean Energy Utilization, Zhejiang University, Hangzhou 310027, China
[b] Research Institute of Wood Industry, Chinese Academy of Forestry, Beijing 100091, China

ARTICLE INFO

Keywords:
Lignite
Microwave irradiation
Pyrolysis
Char
Upgrading

ABSTRACT

To upgrade Indonesian lignite by significantly reducing volatile matter content, char adsorbent was employed to accelerate lignite pyrolysis through microwave irradiation. Physicochemical properties of pyrolyzed lignite were determined with the assistance of char adsorbent on Fourier-transform infrared spectroscopy, X-ray photoelectron spectroscopy, X-ray spectroscopy, N_2 adsorption porosimetry, and contact angle analysis. Under the same microwave power of 800 W and irradiation time of 12.5 min, the lignite temperature remarkably increased from 150 °C to 656 °C with the aid of 10 g char adsorbent. Its apparent aromaticity increased from 0.50 to 0.97. The relative content of superficial carbon functional groups attributed to π–π^* transitions increased from 1.18% to 6.71%. The volatile matter content (dried basis) markedly decreased from 48.94% to 6.47% due to the release of volatile compounds in the lignite.

1. Introduction

Coal is an important energy source worldwide. The development and utilization of lignite will gradually attract considerable attention, as its abundance accounts for nearly 45% of the global coal reserves [1]. Lignite is classified as a low-grade fuel and has low calorific value and high moisture content, which critically restricted its direct and wide use. Consequently, upgrading lignite is important in coal research. Pyrolysis, which provides mild conversion of coal into low-carbon fuels and chemicals, appears to be a sufficient thermochemical technique for clean and efficient utilization of coal [2]. Upgrading of lignite through a pyrolysis process not only prevents its self-heating and spontaneous combustion during transportation and storage as the moisture and volatile matter contents were remarkably reduced [3] but also achieves efficient utilization of lignite as a resource generated char, tar, and gas products.

Microwave irradiation, which occurs through the direct conversion of electromagnetic energy to thermal energy within the material, offers volumetric and selective heating. Microwave energy has been applied as an alternative to conventional convective and conductive heating, and it is used in pyrolysis processes. Microwave pyrolysis is a prospective technique for upgrading and efficiently using low-grade fuel [4]. However, coal matrix except some minerals matters was generally unresponsive to microwave after large amount removal of moisture. Therefore the coal matrix, except for its moisture and some mineral matters, was generally unresponsive to microwave [5]. Enhancing the microwave-assisted pyrolysis of coal to reduce energy consumption was the objective of many studies. Lester et al. [6,7] showed that a bituminous coal was heated to coking temperature using a designed microwave cavity with high electric field operating at high microwave level. The co-pyrolysis of waste polystyrene with coal under microwave copper interaction was investigated by Hussain et al. [8]. Pyrolysis was believed to be the combined action of the high-temperature, microwave–metal interaction, and the active species produced during the process. Another feasible technique is the use of a microwave adsorbent. Monsefmirzai et al. [9,10] used copper oxide, vanadium oxide, tungsten trioxide, and ferroferric oxide for the microwave-assisted pyrolysis of coal. Wang et al. [4] did research about the effects of different Fe_3O_4 additions on temperature rising characteristics during coal pyrolysis in microwave field. Mushtaq et al. [11] used coconut activated carbon solid as absorber for tar and gas fuel production from microwave-assisted pyrolysis of coal. The effects of uniformly distributed coconut activated carbon absorber over coal solids to observe uniform process heating at various levels of CAC loading, microwave power, and N_2 flow rate conditions were studied. Activated carbon and graphite were employed by Zhou et al. [12] to accelerate the reduction of moisture and volatile matter content of Indonesian lignite under microwave irradiation for the preparation of quality lignite water slurry fuel.

However, the existing literatures mainly focused on the

* Corresponding author.
E-mail address: juncheng@zju.edu.cn (J. Cheng).

https://doi.org/10.1016/j.fuel.2018.01.044
Received 28 August 2017; Received in revised form 6 January 2018; Accepted 11 January 2018
Available online 30 January 2018
0016-2361/

characteristics of microwave pyrolysis, such as yields and compositions of tar and gas products. Little information was found on the various physicochemical properties of the pyrolyzed lignites following the microwave pyrolysis process. Generally, nearly no studies have been conducted on the effects of char (i.e., the pyrolyzed lignite) on the properties of its parent lignite upgraded by microwaves. Therefore, char was employed as adsorbent to accelerate microwave-pyrolysis of its parent lignite in this work. Physicochemical properties of pyrolyzed lignite with the assistance of char adsorbent were also determined on Fourier transform-infrared spectroscopy (FTIR), X-ray photoelectron spectroscopy (XPS), X-ray diffraction (XRD), N_2 adsorption porosimetry, and contact angle analysis. This work will help develop cheap, efficient, specific functional and directly reusable adsorbent for microwave pyrolysis of coals.

2. Material and methods

2.1. Material

An Indonesian lignite sample with a high moisture and volatile matter content was upgraded through microwave irradiation in the experiments. The char which was used as microwave adsorbent was produced as follows: A 40 g graphite cylinder Φ6 × 6 mm (with carbon and oxygen contents of 96.7% and 2.84%) with a microwave power of 800 W, microwave time of 25 min, and a lignite weight of 160 g was used for lignite pyrolysis. Using this graphite, the rapid pyrolysis of Indonesian lignite under microwave irradiation was confirmed in Ref. [12]. After pyrolysis, the char (i.e., the pyrolyzed lignite) was separated completely from the graphite cylinder with the sieving method, and further collected. The solid yield of lignite was 23%, indicating that the Indonesian lignite was pyrolyzed because its weight loss due to moisture content was 57.33%.

2.2. Microwave upgrading experiments

Microwave upgrading experiments were performed in a microwave oven with a maximum power output of 2000 W, a frequency of 2450 ± 50 MHz, and multimode resonant cavity dimensions of 400 × 400 × 420 mm. The sample was loaded into a quartz reactor with a nitrogen gas inlet and a gas outlet at the center of the microwave cavity. Nitrogen gas flow rate was controlled to 200 mL/min to facilitate the passage of gas through the sample bed and the release of the gases generated during the reaction. The investigation of the effects of char addition on the microwave upgrading characteristics of Indonesian lignite was conducted under the following conditions: 10 g char with a microwave power of 800 W, irradiation time of 0 min to 12.5 min, and a lignite weight of 60 g.

2.3. Methods

The physicochemical influences of microwave treatment with/without char on the Indonesian lignite were investigated. The modified lignite was pulverised to less than 150 μm. And part of the pulverized lignite was dried at 105 °C for 4 h.

FTIR of lignite was analyzed on a spectrophotometer (Nicolet 5700, Thermo, USA). The dried lignite powder was mixed with KBr and then pressed into pellets for the test. Data collection was performed with 64 scans in an infrared spectral range of 4000–400 cm^{-1}. Baseline correction processing of the spectra was done by means of the Omnic software.

XPS measurement of lignite (dried) was obtained by using an electron spectrometer (Escalab 250Xi, Thermo Fisher Scientific, USA). Data collection was performed in a binding energy range of 279–298 eV with a step scan of 0.10 eV. The spectrum was curve-fitted by a software XPSPEAK.

XRD of lignite was taken on a spectrophotometer (X'Pert PRO, PANalytical B.V., Netherlands). Undried sample powder was scanned from 5° to 80° in 2θ range with 0.0167 step interval.

Pore structure of pulverized lignite (dried) was determined on a nitrogen adsorption analyzer (ASAP 2020, Micromeritics, USA) under a relative pressure which ranged from 0.01 to 0.99 at 77 K.

The contact angle of pulverized lignite was measured on a contact angle meter (JC2000C, Shanghai Zhongchen, Ltd., China) equipped with a laser illuminator and a digital video image system. Contact angle test of lignite was measured on a contact angle analyzer (JC2000C, Shanghai Zhongchen Ltd., China) equipped with a laser illuminator and a digital video imaging system. 1 g pulverized lignite (undried) was pressed into a cylinder with 20 mm diameter and 2 mm thickness at 10 MPa for 30 min. A pendant droplet of distilled water was deposited on the cylinder lignite sample. The images were captured at the exact moment when the water droplet contacted the cylinder lignite. Then the contact angle was calculated using the built-in imaging software.

3. Results and discussion

3.1. Effect of char adsorbent on temperature rise and weight loss of lignite

Effects of char adsorbent on temperature rise and weight loss of Indonesian lignite upgraded through microwave irradiation are shown in Fig. 1. The rise in temperature and weight loss of lignite increased with the addition of char. When microwave time increased to 12.5 min, the temperature and relative weight content of lignite were 150 °C and 48%, respectively. Upon the incorporation of char, the temperature of lignite increased rapidly to 650 °C at a microwave time of 7.5 min. Simultaneously, the relative weight of lignite was 24.6%, which further decreased to 16.46% at a microwave time of 12.5 min. As the moisture content of raw lignite was 57.33%, this result indicated that the lignite without char was mainly dewatered during the microwave irradiation process. The addition of char evidently increased weight loss rate, implying the occurrence of pyrolysis of lignite with remarkably reduced volatile matter content. Compared with raw coal, the pyrolyzed coal possessed a relatively large amount of delocalized π electrons that move on its incipient graphenic structures [5]. Char adsorbent allowed the repeated motion of π electrons under microwave irradiation to generate electron flow and the bound charge for interface polarization and conduction loss effects promoted good microwave heating properties [13,14]. Thus, char was employed as adsorbent to facilitate lignite heating via microwave irradiation.

3.2. Effect of char adsorbent on chemical compositions of upgraded lignite

Effects of char adsorbent on proximate and ultimate analyses of

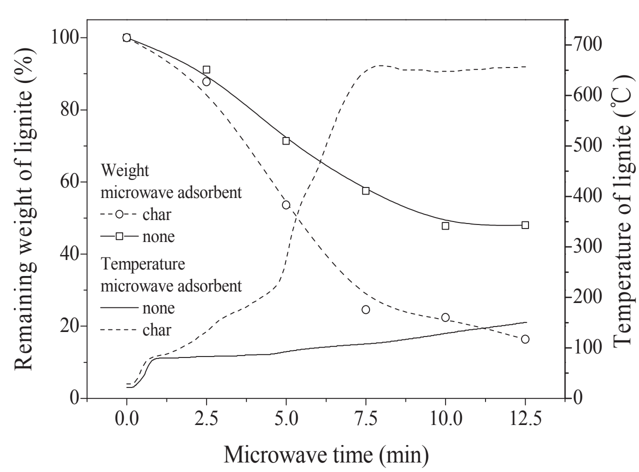

Fig. 1. Effects of char adsorbent on temperature rise and weight loss of lignite upgraded through microwave irradiation.