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The steam gasification of coal with molten blast furnace slag as heat carrier and catalyst: Kinetic study

Wenjun Duan^{*}, Qingbo Yu, Tianwei Wu, Fan Yang, Qin Qin

School of Metallurgy, Northeastern University, No 11, Lane 3, Wen Hua Road, He Ping District, Shenyang 110819, Liaoning, People's Republic of China

ARTICLE INFO

Article history:

Received 11 January 2016

Received in revised form

17 July 2016

Accepted 24 July 2016

Available online xxx

Keywords:

Coal

Steam gasification

Molten blast furnace slag

Kinetic model

Carbon conversion

ABSTRACT

In this study, we carried out a kinetic investigation to analyze the steam gasification of FS (Fu Shun) coal. The effect of reaction temperature (1573 K–1673 K) was studied, and the coal/slag ratios were in the range of 1:0–1:2. The reaction temperature and coal/slag ratio affected the carbon conversion and reactivity index of FS coal gasification. With reaction temperature increasing, the time for carbon conversion completing decreased. But the effect of temperature was non-significant, when it was above 1623 K. The reactivity index of coal at 1673 K was about 1.6 times faster than that at 1573 K, when the coal/slag ratio was 1:0. The BFS (blast furnace slag) acted as not only a heat carrier but also an effective catalyst in steam gasification. Compared with the steam gasification of “pure” FS coal, both carbon conversion and reactivity index of gasification were enhanced by BFS. Meanwhile, the Diffusion model (D_1 model) and Shrinking core model (R_2 model) were proved as the most appropriate model to describe the steam gasification without and with BFS as heat carrier, respectively. The kinetic parameters applicable to the established model with different coal/slag ratios were obtained. Under these conditions, the valid range of activation energy for gasification reaction was $20 \text{ kJ}\cdot\text{mol}^{-1}$ – $64 \text{ kJ}\cdot\text{mol}^{-1}$.

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Introduction

“Hydrogen energy economy” had a significant merit to reduce global CO_2 emissions [1]. At present, 96% of the total H_2 production was contributed by fossil fuels [2]. However, the fossil fuels caused numerous environmental problems, such as greenhouse effect, huge energy consumption, ozone layer depletion and acid rain [3]. Therefore, producing the sufficient H_2 economically and environmentally friendly was a major challenge.

In face of this challenge, Liu [4] proposed the theory of coal gasification reaction with molten BFS as heat carrier to produce hydrogen-rich syngas. Based on this theory, an attempt was made by Duan [5–10] to explore the potential of hydrogen-rich syngas production and waste heat recovery of slag. Li and Yu et al. [11–15] also carried on the thorough research in the aspect of gasification and waste heat recovery. They found that the BFS provided the energy needed for gasification reaction, while the higher content of hydrogen in the syngas was obtained. Luo [16,17] used the BFS waste heat to generate hydrogen-rich gas in a continuous moving-bed

^{*} Corresponding author. P.O. Box 345, Northeastern University, No 11, Lane 3, Wenhua Road, Heping District, Shenyang, Liaoning, People's Republic of China. Fax: +86 024 83672216.

E-mail addresses: duanwenjn@163.com (W. Duan), yuqb@smm.neu.edu.cn (Q. Yu).

<http://dx.doi.org/10.1016/j.ijhydene.2016.07.187>

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biomass gasification reactor. When temperature of BFS was 1473 K and its size was below 2 mm, the gas yield and H₂ content achieved the maximum of 1.28 Nm³·kg⁻¹ and 46.54%, respectively. The integrated CO₂/sludge gasification using the slag waste heat was explored by Sun et al. [18,19]. The gas yield reached to 0.422 kg·kg_{sludge}⁻¹ at 773 K, and the SO₂ reduced to 114 ppm at 1173 K.

The steam gasification of coal was a process for utilizing the energy in coal resources, without the traditional combustion process [20]. Sedghkerdar et al. [21] investigated the CO₂ capture and H₂ production during the steam gasification using a horizontal fixed reactor. The maximum fraction of H₂ (81.7%) and minimum CO₂ concentration (almost 0%) were obtained at 948 K. The experiments of H₂ production by coal gasification in supercritical water was conducted by Jin [22]. The carbon gasification efficiency was 95.7%, and the hydrogen yield was 42.22 mol·kg⁻¹. Phuhiran C [23] carried out the experiment of H₂ production by catalytic steam gasification. The highest H₂ fraction and the maximum H₂/CO ratio reached to 46.68% and 1.24 at 923 K, respectively.

As the research progressed, the kinetic analysis of steam gasification had been ever the subject of previous studies [24–29]. Jin [25] developed a novel kinetic model concentrating on gas species by lignite gasification in supercritical water. The model could be a useful tool for predicting the gas yield and the clarifying the mechanism. Kim [28] proposed a kinetic model of steam gasification of Ca-loaded lignite char. The model quantitatively described the change of the char conversion within the whole time range, all of gasification reactions obeyed Langmuir–Hinshelwood mechanisms. Jang [29] investigated the kinetic experiments in order to achieve the SNG production. In his study, the basic model was selected and limited kinetic values, and equations were applied. The model for catalytic coal gasification was executed about another different coal and it could obtain the result in the syngas composition which could be compared with the basic model.

From the above researches, the kinetic investigation of the steam gasification was benefited to control the reaction process. However, there was no kinetic study for steam gasification of coal with molten BFS as heat carrier. Although Li et al. [14,15] established the kinetic model of coal-CO₂ gasification with molten BFS as heat carrier, there still was significant difference of the coal reactivity between steam gasification and CO₂ gasification, which would result in various differences in design and operation of the coal gasifier. Therefore, it was necessary to conduct the detailed and systematic research on steam gasification of coal with molten BFS as heat carrier.

In present paper, the isothermal steam gasification of coal with molten BFS as heat carrier was systematically investigated using thermo-gravimetric method. The effect of temperature and coal/slag ratio on the carbon conversion and reactivity index were obtained. Furthermore, the steam gasification of coal was simulated using typical kinetic models. The inherent kinetic parameters of steam gasification were obtained and the most appropriate model was established.

Methods and mechanism

Experimental apparatus and procedure

A SETSYS Evolution TGA coupled with a Wetsys was used to perform atmospheric steam gasification reaction in molten BFS. The schematic diagram of the experimental apparatus was presented in Fig. 1. In experiment, the sample was hung below an electronic balance by suspension wire, and the weight loss of sample was recorded by the balance. The gas flowed in the reactor to form the circulation reaction area, and then flowed out after the reaction completing. Ar stream was adopted as a protective gas and controlled with a flowmeter. The steam stream was controlled and heated to 308 K by Wetsys. At last, the reaction gas of steam and Ar were adopted in the thermo-gravimetric experiment.

In experiment, the coal sample mixed with/without BFS (~10 mg) was placed in alundum crucible (Φ 5 × 4 mm) and Ar stream flowed through the balance at 50 mL·min⁻¹ as the protective gas. As shown in Fig. 2, the coal char preparation and gasification proceeded consecutively in the thermo-gravimetric analyzer. The sample was pyrolyzed by heating it to 1273 K at 20 K·min⁻¹ in Ar atmosphere. The sample was maintained at 1273 K for 30 min to complete the pyrolysis. After it, the sample was heated to the desired temperature (1573 K, 1623 K and 1673 K) at 20 K·min⁻¹ in Ar atmosphere. Then, the steam mixed with Ar was flowed into the reactor to complete the coal gasification in molten BFS. The total gas flow was 50 mL·min⁻¹, and the relative humidity was controlled at 75%. The gasification reaction continued for 60 min to ensure the reaction running completely. The mass of the sample could be measured by the thermo-gravimetric analyzer and recorded by a computer.

Sample characterization

Fu Shun (FS) coal was utilized in this kinetic study. FS coal sample with particle size about 75 μm was prepared to ensure that the intrapore diffusion did not control the coal gasification rate [30–32]. The ultimate analysis of the sample was conducted using a CHNS/O Analyzer, Perkin Elmer PE 2400 series II. The proximate analysis of the sample was performed at atmospheric pressure using an auto-measuring industrial analyzer, SUNDY SDTGA 5000 according to the ASTM Standard Methods (ASTM D 5142-90, ASTM D 5373-93 and ASTM D 6349-98) for coal. The proximate analysis and ultimate analysis of FS coal sample were shown in Table 1.

In addition, the water quenched BFS supplied by a factory was used as heat carrier and catalyst. To confirm the glassy phase of the quenched slags, XRD (X-ray diffraction) analysis was carried out, as shown in Fig. 3. The XRD test was performed in a 2θ degree range of 10°–70° with a speed of 4°·min⁻¹, where θ was the angle between the incident ray and the scattered plane and the intensity of the reflected ray was shown in Y-axis. The chemical compositions of BFS were analyzed by X-ray fluorescence (XRF, SE-Explore, Bruker). The

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