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Experimental study on steam gasification of coal using molten blast furnace slag as heat carrier for producing hydrogen-enriched syngas

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ABSTRACT

The new method for producing hydrogen-enriched syngas (HRG) by steam gasification of coal using molten blast furnace slag (BFS) as heat carrier was established. In order to achieve the HRG production, a gasification system using this method was proposed and constructed. The carbon gasification efficiency (CE), hydrogen yield (YH_2) and cold gasification efficiency (CGE) in the molten slag reactor were measured, and the effects of temperature, S/C (steam to coal) ratio and coal type on the reaction performance were accessed. The results indicated that the preferred temperature was 1350 °C, which ensured the miscibility of coal–steam–slag, the diffusion of reactant in molten BFS as well as recovering waste heat. The optimal S/C ratio was 1.5–2.0 for producing HRG. Under these conditions, the hydrogen fraction was higher than 63% and the gas yield reached to 1.89 Nm^3/kg. The CE and CGE were higher than 96% and 102%, respectively. The YH_2 also reached to 1.20 Nm^3/kg. Meanwhile, different types of coal were successfully gasified in molten BFS reactor for producing HRG. The proposed method enhanced the gasification efficiency of different types of coal, recovered the BFS waste heat effectively, and had important guidance for industrial manufacture.

1. Introduction

Hydrogen was a very versatile environmental energy carrier and the development of cheap and clean hydrogen generation technology had been the most hot research topic [1,2]. The method of steam gasification with fossil fuels or renewable to produce hydrogen was widely accepted [3,4]. However, for steam gasification process, some key reactions were intensely endothermic reactions and demanded a large amount of energy [5–7]. In general, the energy was provided by fossil fuel or syngas combustion. Meanwhile, a certain amount of CO_2 was produced in steam gasification process and not friendly for the environment [8,9]. Therefore, it was essential to find a reasonable energy supply to achieve the environmental and economic benefits in steam gasification process for producing hydrogen.

As we all know, iron and steel industry was an energy-intensive industry, which accounted for approximating 5% of the total energy consumption with a waste heat recovery rate of only 17% [10–12]. BFS was exhausted at extremely high temperature (about 1500–1600 °C), and the energy carried was about 1700 MJ/t_{slag}. For example, in 2013, the crude steel production reached up to 822 million tons in China, which meant that the waste heat of hot slag was equivalent to 14.8 million tons standard coal [13]. Therefore, establishing an integrated system of recovering BFS waste heat to provide the energy needed for the steam gasification process was necessary. In the theoretical analysis, Ishida [14] established the enthalpy-exergy diagram to analyze the BFS waste heat recovery. Akiyama et al. [15] described a feasibility study of recovering BFS heat using nine chemical reactions by enthalpy-exergy diagram method. By this method, thermodynamic analysis of BFS waste heat recovery system integrated with coal gasification was conducted by Duan et al. [16]. The results showed that the exergy efficiency and recycling efficiency of the established system were 52.6% and 75.4%, respectively. The exergy loss of the integrated system was only 620.0 MJ/t_{slag}. Meanwhile, the thermodynamic analysis of steam gasification using BFS as heat carrier and recycling its waste heat to produce HRG were investigated by Duan et al. [12,17]. The results suggested that the preferred condition for HRG from Datong coal was achieved at 775 °C, atmospheric pressure and S/C ratio of 2.0–3.0. In experimental studies, Luo et al. [18] established a continuous moving-bed biomass gasification reactor to evaluate the feasibility of recovering BFS particles waste heat and producing HRG at 1000 °C. Zhao [19] investigated the possibility of producing the combustible gas from municipal solid

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Nomenclature

n_{H_2}	the mole of the product hydrogen, mol
m_{coal}	the mass of the coal, kg
m_H	the mass fraction of hydrogen in the coal, %
h_{fg}	the enthalpy of vaporization of water, MJ/kg

Abbreviations

BFS	blast furnace slag
HRG	hydrogen-enriched syngas
DT coal	Datong coal
FX coal	Fuxin coal
FS coal	Fushun coal
SN coal	Shennan coal
DZ coal	Dezhou coal
CE	carbon gasification efficiency
YH_2	hydrogen yield
LHV	lower heating value
HHV	higher heating value
CGE	cold gasification efficiency

waste using BFS waste heat. In his study, the effects of temperature, gasifying agent and BFS particles on the gas production were studied at 600–900 °C. Sun et al. [20] also investigated the characteristics of biomass gasification and syngas release behavior using hot slag at the low temperature (200–450 °C). The syngas (0.149 L CO, 0.036 L H_2 and 0.069 L CH_4) could be produced by per gram of wheat straw when the biomass was mixed with slag. Obviously, the current methods of producing hydrogen by steam gasification using BFS waste heat were entirely feasible. But these methods were only limited to using slag particles at relatively low temperature. However, for BFS waste heat, the high-grade waste heat mainly concentrated in molten slag at 1250–1600 °C. The process of hydrogen production by steam gasification using molten BFS as heat carrier was never referred to and lack of comprehensive study. Therefore, it was necessary to investigate the steam gasification process using molten BFS as heat carrier, which would be more benefit for hydrogen production and recovering high-grade waste heat.

In the present study, the approach for steam gasification of coal in molten BFS to produce HRG was proposed and implemented. The effects of temperature, S/C ratio and coal type on steam gasification reaction were investigated, so as to obtain the optimal operation parameters and guide the industrial production.

2. Materials and methods

2.1. Feedstock

Five types of coal (Datong (DT) coal, Fuxin (FX) coal, Fushun (FS) coal, Shennan (SN) coal and Dezhou (DZ) coal) were obtained from various big coal mines in Northern China for gasification experiments to produce HRG. Their compositions and properties were shown in Table 1. 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 conducted using an automatic coal proximate analyzer. During preparing coal samples, the first step crushing coal pieces with a hammer and crusher for the appropriate size. Then these crushed particles were segregated into about 75 μm using a series of sieve shakers.

Table 1
The proximate and elemental analysis of the coal.

Coal	DT	FX	FS	SN	DZ
Proximate analysis (wt%)					
Moisture (wt%)	9.05	3.13	1.83	2.30	4.96
Volatile matter (wt%)	38.38	32.78	28.72	26.38	4.82
Fixed carbon (wt%)	38.42	34.79	20.81	22.85	19.11
Ash (wt%)	14.15	29.30	48.64	48.47	71.11
Ultimate analysis (wt%)					
Carbon	64.53	52.27	39.05	42.59	21.57
Hydrogen	3.746	4.204	3.489	3.379	0.710
Nitrogen	0.956	1.09	1.13	0.74	0.19
Sulfur	0.561	1.210	0.619	0.967	0.457
Oxygen	7.007	8.796	5.242	1.554	1.003

BFS from an iron and steel company was used in experiments as heat carrier with chemical compositions of 41.21 mass% CaO, 34.38 mass% SiO_2, 11.05 mass% Al_2O_3, 8.22 mass% MgO, 2.78 mass% Fe_2O_3, 0.35 mass% TiO_2 and some minor constituents of sulfur, manganese and phosphor oxides. Its chemical compositions were analyzed by X-ray fluoroscopy (XRF, SE-Explore, Bruker). In the experiment, the pulverized coal was conveyed by carrier gas and the reactor should be replenished with shielding gas before the experiment. The carrier gas and shielding gas used in experiments were N_2 with 99.99% purity.

2.2. Experimental system

The experiment system of steam gasification of coal using molten BFS as heat carrier for producing HRG was established, as shown in Fig. 1. The system was composed of seven parts: feeding system, primary reactor system, temperature measurement system, gas purification system, flow measurement system, gas composition analysis system and off-gas treatment system.

The primary reactor system was the key part of the integrated system, as shown in Fig. 2. The high purity corundum with high corrosion resistance was adopted as the reactor so as to perform the reaction for a long time. The reactor, which was externally heated in an electric furnace (SG1-15-16), had a height of 100 cm and a volume of 196 cm^3. Meanwhile, the feeding system was also a key part to ensure the successful experiment. The powder feeder (TWLP 15) was adopted in the feeding system, and the rate was controlled in the rage of 2.0–25 g/min. The bulk density of the pulverized coal in the powder feeder was about 0.74 g/cm^3. At last, the portable gas analyzer (Gasboard-3100P) was used to ensure the accuracy of the experimental results.

2.3. Experimental procedure

In a typical test, the suitable height of the molten BFS was about 300 mm for the gasification reaction [21]. Therefore, about 1.8 kg BFS particles sample was crushed and ground to about 2 mm and then put into the reactor. The loaded reactor was placed in the electric furnace and heated to the target temperature, and then kept it about 2 h so as to the temperature remain stable and evenly distribute. The rated power, rated voltage and rated temperature of the electric furnace were 15 kW, 380 V and 1600 °C, respectively. The reactor was insulated with glass wool material to maintain the temperature of gasification reaction in molten BFS. Furthermore, there was a temperature controller with a thermocouple (type K) placed into the reactor to control and record the temperature. When steam stream and the target temperature were stable