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Direct production of high hydrogen syngas by steam gasification of Shengli lignite/chars: Remarkable promotion effect of inherent minerals and pyrolysis temperature

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ABSTRACT

The steam gasification of Shengli (SL) lignite and the demineralized lignite/chars were tested on a laboratory fixed-bed micro reactor system. The evolution rates of syngas, carbon conversions to gas, yields of gaseous products, and H₂/CO molar ratios of SL lignite/chars were systematically investigated. Meanwhile, the gasification performance of SL lignite/chars was compared with the corresponding demineralized samples. The results showed that the inherent minerals in SL lignite/chars not only effectively promoted the steam gasification reaction but also considerably improved the gasification selectivity towards the production of H₂ and CO₂. Further characterizations by the scanning electron microscope (SEM-EDS) and X-ray fluorescence (XRF) demonstrated that it became more difficult for the minerals in SL lignite being eluted by hydrochloric acid solution with the temperature increasing of pyrolysis process. The results confirmed that the inherent minerals and lignite formed metals-lignite intermediate complex during pyrolysis process. The formed metals-lignite intermediate complex works as the active sites which effectively promote the steam gasification performance of SL lignite/chars, especially in favor of the direct generation of high hydrogen syngas.

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Introduction

The development of all-around technology for integrated gasification combined cycle (IGCC) with the core of coal gasification has already become an important direction of the efficient and clean utilization of coal resources [1–6]. Indeed, using the

technology of circulating fluidized bed to gasify the low-rank coals such as lignite at low temperatures (<900 °C) is an important and essential way to utilize lignite and other low-rank coals efficiently. The high hydrogen syngas could be produced by steam gasification of coal [7]. Steam gasification made coal to be a major source of clean H₂ energy and led to high

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energy conversion efficiencies of coal [6]. The high hydrogen syngas was applied in many fields which included fine chemical industry [8,9], microwave plasma treatment [10], turbine applications [11], etc. Low-rank coals were more likely to produce the high hydrogen syngas by steam gasification because of the natural structure properties [6–12]. Various factors could influence the steam gasification of coal, such as the structures or chemical constituents (surface area, pore structures, mineral substances, chemical constituent, etc.) [13–15], and other parameters relating to the reaction conditions (particle sizes, temperature, and gas pressure) [16,17]. Some researchers have reported in the early 1980s that metal components in coal remarkably promoted steam gasification performance and affected product selectivity of gasification [18–20].

Li et al. [21] found that the inherent mineral presented a distinct catalytic effect on the steam gasification reaction of SL lignite. Luo and Smoliński et al. [22–24] reports that blast furnace (BF) slag as a catalyst could improve the gasification reaction to hydrogen-rich gas. One possible reason was BF slag contained some metal oxides, such as Al_2O_3 , CaO and MgO . Jin et al. [25] investigated the Na transformation rule of Zhundong coal gasification in supercritical water by fluidized bed reactor. Zhang et al. [26] selected Na_2CO_3 and $\text{Ca}(\text{OH})_2$ as catalysts, and Na^+ , Ca^{2+} and $\text{Ca}^{2+}/\text{Na}^+$ loaded in coals were prepared by an ion-exchange procedure using a sub-bituminous coal. The research founded Na^+ and Ca^{2+} catalysts showed remarkable activity for gasification, and the $\text{Ca}^{2+}/\text{Na}^+$ -loaded coal exhibited the highest reactivity among the coal samples prepared. Domazetis et al. [27,28] indicated that iron species in brown coal formed small iron clusters by heating, and the active site for steam gasification was shown to be Fe–C. Besides, many studies revealed that the metal elements in coal, especially alkali metals [26,29–32], alkaline earth metals [26,33,34] and transition metals [19,27,35–37] had a significant catalytic effect on the steam gasification reaction of coal. Although there have been a lot of reports on the catalytic effects of metal constituents on the steam gasification of coal, the opinions on some key issues achieved in these reports are inconsistent or even contradictory, which is mainly owned to the complexity of coal microstructures and the complicated mechanisms of mineral catalysis during gasification reaction [16,28,38–40].

In this work, we investigated the steam gasification reactivity of SL lignite/chars and their corresponding demineralized lignite/chars to evaluate the effects of inherent minerals and pyrolysis temperature on the characteristics of steam gasification. It was found that inherent minerals in SL lignite had effectively promoted the hydrogen yield by steam gasification of SL lignite and formed the active complex with lignite at pyrolysis stage. This work has the potential to provide theoretical basis and technical support for the high hydrogen syngas by steam gasification of SL lignite at lower temperatures.

Experimental

Coal sample and demineralized chars preparation

In this study, lignite samples obtained from Shengli coal field in Inner Mongolia of China were used as the raw material

(labeled as SL). The lignite coal was crushed and sieved to 0.15–0.18 mm in diameter and stored under N_2 atmosphere.

- (1) The pyrolysis of coal samples was carried out in a stainless steel reactor (40×1000 mm) which was heated by an electric furnace. Dried coal sample (20 g) placed into central zone of the reactor were pyrolyzed for 120 min under flowing argon atmosphere at 300, 500 and 700 °C. The chars labeled as SL-300, SL-500, and SL-700, respectively, were ground with agate mortar and pestle. The powdery chars (particle size, <0.15 mm) were used in subsequent work.
- (2) The demineralized SL lignite/chars were obtained by acid leaching. Approximately 10 g of lignite/chars was mixed with 100 mL 17% hydrochloric acid (analytically pure, 38% wt), aqueous solution and stirred for 2 h at room temperature. The resulting slurry was filtered using a vacuum filter and then rinsed continuously with distilled water to remove the residue chloride ions, dried at 105 °C for 4 h. The solid products were labeled as SL-HCl, SL-300-HCl, SL-500-HCl and SL-700-HCl, respectively. The results from the proximate analysis of tested samples are presented in Table 1.

Steam gasification

The reactor system (Scheme 1) was mainly composed of a steam generator and a fixed-bed reactor that was made of stainless steel (8×350 mm) heated by an electric furnace [21]. For each experiment, 0.5 g coal sample was placed in the central zone of the tubular reactor between quartz wool and quartz sand where the temperature was controlled homogeneously. Prior to the gasification, the steam generator was preheated to 200 °C, and pressure in the reactor was increased to 0.6 MPa by introducing argon (purity: $\geq 99.99\%$) at a flow rate of 200 mL min^{-1} . After the desired steam generator temperature and system pressure were reached, the reactor was heated to 500 °C at the heating rate of 15 °C min^{-1} , then changed the heating rate to about 1.2 °C min^{-1} until 850 °C, and maintained the temperature of 850 °C for 2 h. In the meantime, the steam (0.12 mL min^{-1}) was introduced into the steam generator when the reaction temperature reached 500 °C. The outgoing gases passed through an ice-water trapper and then a cooler controlled at -20 °C to prevent steam and tarry matters into the GC analyzer. The concentrations of hydrogen (H_2), carbon monoxide (CO), methane (CH_4), and carbon dioxide (CO_2) in the outgoing gasses were quantitatively determined using an online gas chromatography (Agilent 7890A).

Table 1 – Proximate analysis of SL lignite/chars (wt%).

Sample	A_d	V_d	F_{cd}
SL-HCl	6.05	40.62	53.33
SL-300-HCl	7.27	37.90	54.84
SL-500-HCl	7.00	30.39	62.61
SL-700-HCl	13.32	14.86	71.82
SL	11.34	39.70	48.96

d, dried basis.

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