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Catalytic steam gasification of lignite with olivine as solid heat carrier

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HIGHLIGHTS

- Coal catalytic steam gasification was carried on an external circulating co-current moving bed (ECCMB) gasifier.
- Effects of gasifier temperature, steam/coal ratio (S/C), bed materials and bed height of the gasifier were studied.
- Tar content in the product was significantly reduced with calcined olivine as catalytic bed materials.

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ABSTRACT

Catalytic coal gasification was carried on a lab-scale external circulating co-current moving-bed (ECCMB) gasification system with steam as gasification agent and olivine as catalytic solid heat carrier. The system is composed of a moving-bed gasifier and a riser combustor. The effects of gasifier temperature, steam/coal mass ratio (S/C), bed materials and bed height on the product gas compositions, carbon conversion, and chemical efficiency were investigated. The result show that the gas yield and carbon conversion increase with the increase of the gasifier temperature. The highest H_2 concentration of 60.4 vol.% is produced from Dayan lignite at the gasifier temperature of 800 °C and S/C ratio of 1.0. The carbon conversion increases remarkably at the S/C range from 0 to 0.4. Olivine as the bed material has a satisfying catalytic tar cracking efficiency. Appropriate bed height favors tar destruction because of the increase in the contact time of the tar and steam with the catalytic bed material.

1. Introduction

Coal gasification technologies have been extensively studied in the past decades, and significant progress has been made in process operation, control, scaling up, and optimization; in gas quality and its utilization; and in impact on the environment [1–5].

To produce hydrogen-rich gas or syngas, steam as a gasification agent is preferred. With steam, H_2 content in the gasification raw gas can be as high as 40–60 vol.%, dry basis [6]. However, steam gasification reaction of coal is endothermic, and needs additional heat supply to the gasification system. The so-called autothermal gasification processes, in which air–steam or oxygen–steam mixtures act as the gasification agent, could be a solution to this problem and have been intensively investigated in the past decades [7–9]. These processes will result in a large amount of N_2 in the produced gas for the air–steam gasification case, or need additional cost to produce oxygen for the oxygen–steam gasification. The circulation of the solid heat carrier should be a good way to supply the energy required in steam gasification. It has actually been employed in various types of dual fluidized bed gasification (DFBG) processes such as the FERCO SilvaGas process [10], the FICFB process [11], and the others coal gasification process [12,13].

Tar production is another major problem that needs to deal with during the coal steam gasification process. Excessive tar production causes problems in the process equipment as well as the engines and turbines used in application of the product gas, and therefore, complex cleaning systems are required, which causes an increase in the external cost. Tar formation also reduces the thermal efficiency. Thus, it is important to convert tar into gaseous products such as H_2 and CO during gasification for improvement in the controllability and thermal efficiency of coal gasifiers [14].

The use of catalysts in coal gasification has been attracted increasing attention because of many advantages of the lower gasification temperature. Especially, with the help of catalytic tar destruction or in situ tar steam reforming, the tar content in the product gas could be reduced extensively. In the past decades the research of catalytic coal gasification focused mainly on finding some catalysts with high activity, low price, and low empoison and easily reclaiming or not needing reclaim [15]. However, the gasification process was based on traditional reactors, for example, fixed bed, moving bed and fluidized bed, which had some intrinsic problems [16].

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In the FICFB process, the heat needed for the steam gasification is transferred by solid heat carrier through a circulation loop of bed material created between the gasification zone and the combustion zone [11] With an active bed material olivine as the solid heat carrier, the in situ catalytic tar destruction is achieved at the same time.

Our group has developed an external circulating co-current moving bed (ECCMB) gasification system for catalytic biomass gasification [14]. It is composed of decoupled moving-bed gasifier and riser type combustor. A circulation loop of bed material, which acts as catalyst and heat carrier, is created between the gasifier and the combustor. The char circulated with the bed material and the cokes deposited on the catalyst are burned off in the combustor to regenerate the catalyst and provide the energy for the endothermic gasification reactions with steam in the gasifier. The product gas and the flue gas remain separated. The co-current movement of catalyst, coal and steam in the gasifier is in favor of cracking and steam reforming of tar to produce a high yield of hydrogen-rich gas with low tar content. The principle of the process with lignite as feeding material is shown in Fig. 1.

In this study, catalytic steam gasification of lignite in the ECCMB gasification system has been conducted. The effects of the operating parameters such as gasifier temperature, steam/coal mass ratio (S/C), type of bed material and bed height of the gasifier on the product gas quality and the gasification performance (gas yield, carbon conversion and chemical efficiency) have been investigated.

2. Experimental

2.1. Experimental set up

Experiments were carried out at ambient pressure in a lab-scale ECCMB apparatus with a maximum coal feeding rate of 1 kg/h. The apparatus had been previously described by Wei et al. [14]. It consists of a moving-bed gasifier (0.028 m i.d. and 0.8 m high) and a riser type combustor (0.028 m i.d. and 3 m high) that are made of stainless steel and are heated by seven independently controlled electrical-furnaces to ensure reaction region isothermal. Thermocouples and manometers are placed in various points along the reactor to indicate and control the temperature and pressure profiles.

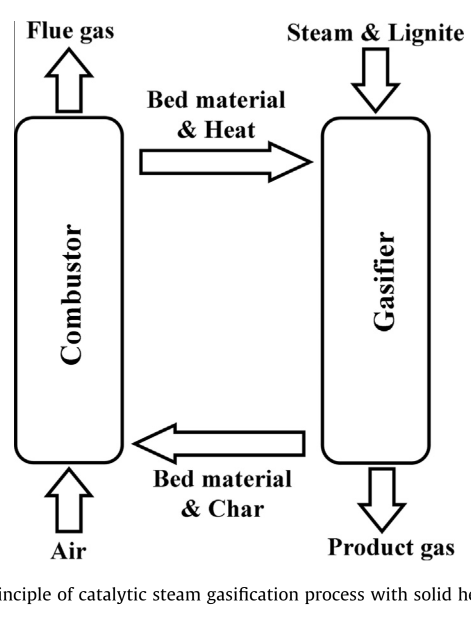

Fig. 1. Principle of catalytic steam gasification process with solid heat carrier.

Coal is fed into the gasifier by a screw feeder at a prefixed and constant flow rate from the hopper on the top of the gasifier. The coal particles from the feedstock hopper and the hot bed material (catalyst) coming from the combustor are rapidly mixed. The quantity of the hot bed material is much higher than that of coal inside the limited space of the reactor to ensure that coal was well mixed with bed material and efficient heat transfer from bed materials to coal particles is obtained. The circulating rate of the mixture of coal and bed material is controlled by feeding rate of the screw conveyor between the moving-bed reactor and the riser combustor. Steam is injected into the mixing zone from a vaporizer to take part in the gasification of the nascent chars and catalytic steam reforming of the volatiles. The reactions continue along the downwards-moving bed until the gaseous products are separated from solids by a metallic sieve at the bottom of the gasifier. The products are further cooled in the sequential ice-cooled quenchers to separate the condensable components. The remaining aerosol is captured in a filter filled with glass wool. The gaseous product is washed with water saturated with $CaCl_2$ before it is collected.

In the combustor, the bed material is fluidized by pressurized air passing through a throttle nozzle at the bottom of combustor. The char and coke deposited on the surface of catalysts are burned off and the catalysts are regenerated and heated to the desired temperature. The hot regenerated catalysts are separated from the flue gas and kept at the hopper on the top of the combustor before they are returned to the gasifier through a rotary valve. A cyclone is set at the exit to separate the ashes in the flue gas.

2.2. Coal sample and catalytic bed materials

The coal used in the experiments is Dayan lignite of particle size 0.45–0.85 mm. The analyses of the lignite are given in Table 1. The coal was dried at 110 °C for 2 h before experiments. A natural olivine was used as catalytic bed materials. The olivine was milled and sieved into size of 0.6–1.0 mm and was calcined at 900 °C for 3 h before experiments. The property of olivine is given in Table 2.

2.3. Experiment procedure

The gasification system was controlled at atmospheric pressure. At the beginning of the experimental run, the electric heaters are turned on to heat up the combustor and the bed materials within gasifier. Circulation of the bed material began with the introduction of fluidization gas (8–10 Nm^3/h) into the combustor. The circulating rate of bed material was fixed at 6 kg/h. When the gasifier temperature was reached to desired temperature (650–800 °C), the coal and steam was fed into the gasifier. The coal feed rate was fixed at 0.12 kg/h. The S/C ratio could be varied by changing the steam flow rate while keeping the coal feed rate constant. In all runs, the bed material was sampled at the top and bottom of the gasifier and analyzed to ensure that catalyst regeneration was satisfactory.

The produced gas was collected in a gasbag every 20 min and off-line analyzed by a GC-920 gas chromatograph with a TCD detector and GDX104 and 5A molecular sieve columns. The analysis was recorded by a NC-2000 chromatograph data workstation. The concentrations of H_2, CH_4, CO, CO_2, and hydrocarbons were determined. After each experiment, the tar traps were washed with tetrahydrofuran and the tar was recovered by evaporating the solvent and water at 60 °C under a vacuum. The produced char was conveyed to the combustor, where it was burned off. The char yield of the gasification process can be calculated by ash balance as described by Wei et al. [14].

The carbon conversion is defined and calculated by