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Research article

Methane-rich syngas production in an integrated fluidized bed by coupling pyrolysis and gasification of low-rank coal

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ABSTRACT

An integrated fluidized bed which combines a fluidized bed gasifier and a transport bed pyrolyzer was proposed to produce CH_4-rich syngas from coupling pyrolysis and gasification of low-rank powder coal below 10 mm. This study investigated the effects of major operating parameters on CH_4 content in the syngas produced from processing a kind of subbituminous coal in a laboratory facility. It was found that the formation of CH_4 was facilitated under the conditions of lower overall excessive oxygen ratio (ER), lower temperature of gasification (830–970 °C), higher temperature of pyrolysis (500–700 °C) and higher holdup of coal in transport bed. The CH_4 content in the produced syngas increased with elevating the operating pressure but decreased with increasing the steam to carbon ratio (S/C) for gasification. Coupling transport bed pyrolysis with fluidized bed gasification at atmospheric pressure caused the CH_4 content in the syngas to be 7.1 vol.% under the conditions of ER = 0.1, S/C = 0.1, gasification temperature of 900 °C and pyrolysis temperature of 700 °C, which was higher than 5.1 vol.% for the case without coupling coal pyrolysis. At the operating pressure of 1.4 MPa the CH_4 content reached about 11.2 vol.%, about six times higher than 2.0 vol.% for the usual fluidized bed gasification and close to 12.0 vol.% for the Lurgi gasifier.

1. Introduction

Natural gas (NG), as an eco-friendly fossil fuel, is becoming the preferred choice for the 21st century with the impending world oil shortage and the coal associated environmental impact. It is even predicted that the world will turn to gas from oil [1]. For China, the rapidly-increasing demand for NG has enlarged the gap of its domestic supply in recent years [2,3]. By 2015, NG consumption will reach 250 billion cubic meters while its production is only to be 146.5 billion cubic meters [4]. China has to rely on a flood of imports of NG to fill its domestic gas gap, and this may lead to rise in the price and a threat to energy security [1,4]. All of this has forced many studies to develop unconventional methods for the production of NG.

Coal to synthetic natural gas (SNG), as a kind of clean coal technology, provides a viable and attractive option for the steady supply of NG by utilizing the domestic abundant low-rank coal reserve in China. One of its appealing features is the SNG plant can be ideally located near coal reserve, while its product can be steadily piped to end-users via existing natural gas pipeline grid. The implementation of SNG production from coal follows four conversion steps, in order, coal gasification, gas cleaning and conditioning, methanation and fuel upgrading [5]. As the first key step, the selection of gasification technology is of vital importance to the subsequent steps. The production of SNG from syngas involves a strong exothermic methanation reaction (R7 in Table 3) to result in the reactor temperature increase of about 60–70 °C by one percent increase in CO conversion rate so that the reaction heat has to be considered because of the high CO content in the syngas [6]. In the existing SNG plant, several adiabatic fixed beds connected in series with recycle of product gas rich in CH_4 are used to dilute CO content to decrease the reaction heat [5,6]. High CH_4 content in the syngas from gasification helps to improve the SNG efficiency greatly as less reaction heat has to be removed via intermediate gas cooling in the methanation step [5]. On the other hand, the gasification efficiency can be improved by effective utilization of the reaction heat of methanation in the gasifier [7]. Consequently, such a gasification technology that can produce CH_4-rich syngas is highly expected for the coal to SNG process.

There are many commercial coal gasification processes using moving bed, fluidized bed and entrained-flow reactor. Among them, the moving bed gasifier with ascending temperature from coal feeding can make for cascade conversion of coal through combining pyrolysis

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and gasification [8]. A remarkable feature of this kind of gasification technology is its high CH_4 content in the producer gas [8,9]. The industrial practices show that nearly half of the total methane production comes directly from coal gasification step based on the typical composition of CH_4-rich syngas with CH_4 content of 8–12 vol.%, but lump coals (e.g., 6–50 mm) are required in the Lurgi pressurized moving bed gasifiers [9–11]. At present, Lurgi gasifiers have been used successfully for the commercial production of SNG in the Great Plains synfuels plant (USA) and the Datang Keqi project (China) [12]. With advancements in mechanical coal mining, the production of lump coal is immensely reduced and a large amount of powder coal (<10 mm) has to be produced. For long it is desired to develop a coal gasification technology that utilizes powder coal to produce CH_4-rich syngas.

Fluidized bed gasification has been considered as the suitable technology for direct use of powder coal (<10 mm) at temperatures of 900–1100 °C. Since the first commercialization of the Winkler gasifier with dry ash discharge, the fluidized bed gasification technology has been progressed to the ash-agglomeration gasifiers including U-gas, KRW and ICC-China. The syngas from both Winkler and agglomeration gasifiers contains about 2 vol.% CH_4. In these gasifiers, coal is converted into volatile and char, and the char is then gasified by steam and oxygen agents in the presence of volatile. The volatile–char interactions can lower the reaction rate of char gasification [13,14]. Therefore, converting the entire non-ash fraction of coal to syngas in a single reactor requires high temperature and long residence time, which is against the formation of CH_4.

If isolation of pyrolysis from gasification is realized in a fluidized bed reactor via the establishment of different temperature fields like in the Lurgi gasifier to reduce the chance of direct contact between volatile and char, not only the gasification rate increases but also the volatile including tar and CH_4-rich pyrolysis gas can be preserved as much as possible. Gomez-Barea et al. [15] found that in the oxygen and steam fluidized bed gasification the devolatilization of fuel firstly occurred at freeboard, if feeding coal on bed, to produce the similar gas composition as in pyrolysis, but the combustible gas components including CH_4, H_2, and CO from pyrolysis would be burned out with oxygen if feeding coal in bed. On the other hand, if partial gasification technology is adopted, gasification efficiency will increase in comparison with the complete gasification. Calculation results suggested that if coal was partially gasified at 80% conversion and the residues were combusted, both the reaction time and the reactor volume could be lowered to about 40% of the total time and total volume for complete conversion [16]. The formation of CH_4 can be accelerated under high pressure due to the catalysis of char on methanation reaction [17,18]. The most effective utilization of coal should be also the use of its hydrogen-rich volatile for conversion products as SNG, and the high-carbon part for energy utilization by gasification and combustion.

With the analysis above, an integrated fluidized bed (IFB) combining a bottom fluidized bed (FB) gasifier and an upper transport bed (TB) pyrolyzer has been proposed to study the fundamentals of preparation of CH_4-rich syngas from low-rank powder coal with sizes below 10 mm. FB gasification provides the endothermic heat and syngas atmosphere for TB pyrolysis. Via coupling pyrolysis and partial gasification, the high reactive volatile in coal is firstly converted into CH_4-rich syngas (although with certain tar as well) that would be the expected inlet gas for methanation in the SNG plant. The residual char after partial gasification in the bottom FB can be burned to drive utilities.

The main objective of this study is to verify the technical feasibility for preparation of CH_4-rich syngas by coupling coal pyrolysis and gasification in a laboratory IFB reactor. Experiments are conducted by varying the major parameters including coal feeding position, excess oxygen mole ratio (ER) and steam to carbon mass ratio (S/C) against entire coal, pyrolysis and gasification temperatures, holdup of coal in the TB and operating pressure. A comparison of CH_4 content in the producer gas is finally made between the proposed IFB process and major commercial gasification technologies.

2. Experimental

2.1. Fuel and apparatus

The tested fuel is a kind of subbituminous coal from Inner Mongolia, China. Table 1 shows the results of proximate and ultimate analyses for the tested coal. The coal contains relative high ash and O contents. Before each test it was crushed and sieved to desired sizes and in turn dried in an atmospheric oven of 105 °C for 2 h. Unless otherwise stated, the mixed coal particles in 0.2–0.5 mm and 1.0–2.0 mm via a mass ratio of 1:1 were used in the experiment.

Fig. 1 shows schematic diagram and the photo of the laboratory IFB apparatus. It consists mainly of an IFB reactor, a pressurized coal screw feeder, a gas supplying system, a pneumatic control back pressure valve, and a gas cleaning system. The IFB reactor is made of SUS 310S stainless steel tubes, and combines a FB gasification section on the bottom and a TB pyrolysis section on the top. The bottom FB has a conical top connecting the bottom of the upper TB via a joint flange. For the bottom FB and upper TB, their inner diameters are 79 mm and 40 mm and heights are 1.5 m and 6.0 m, respectively. There are two coal feeding inlets located 300 mm above the joint flange at the bottom of the TB and under the joint flange at the top of the FB, respectively. An inclined overflow pipe was installed at 800 mm above the bottom of the FB. The bed height for FBG gasification can be adjusted by adding Al_2O_3 ball in diameter of 2–3 mm into the bed bottom. There is a conical distributor at the bottom of FB, which has a 1.0% opening ratio and orifices of 1.0 mm in diameter. The gas supplying system consists of N_2 and O_2 cylinders, gas preheater, water pump, steam generator and gas mixer (also as the second preheater). The gas cleaning system includes a cyclone, a hot metal filter, a quench cooler, a condenser, a cotton filter and gas scrubbers.

2.2. Experimental method

The IFB reactor was externally heated to the presetting temperatures with N_2 flow through the bed by four electric furnaces. When the reactors reached their desired temperatures, the fluidizing gas consisting of N_2, O_2 and steam was preheated by two gas preheaters and induced into the bottom of the IFB. The flow rate of N_2 and O_2 were measured by mass flowmeters, and steam was obtained in a steam generator from a water flow controlled by a constant-flux pump. The N_2 and O_2 were mixed and heated in the first preheater, and then steam was further mixed into the stream in the gas mixer/preheater. The mixed gas was preheated to 700 °C before its entering the bottom FB gasifier. Once the whole system reached a steady state, coal particles were fed via a pressurized screw feeder into the IFB through either of the coal feeding inlets using N_2 as the entrainment gas. When feeding coal into the bottom of TB, the larger coal particles (1.0–2.0 mm) fell down into the FB for partial gasification, and the unreacted coarse char discharged into a lower receiver through the overflow outlet on the bed wall. Other smaller coal particles (0.2–0.5 mm) were pneumatically conveyed into the upper TB to occur fast pyrolysis and the char was separated from gas in a cyclone into an upper receiver. When coal was fed into the top of FB, all of coal particles fell down into the FB and were gasified by O_2 and steam. The produced coarse char was similarly discharged into the lower receiver, but very fine char would be carried by the producer gas to the exit of the TB and received into the upper receiver

Table 1
Proximate and ultimate analyses of the tested subbituminous coal (wt.%, dry basis).

Proximate analysis			Ultimate analysis					
VM	Ash	FC	C	H	O[a]	N	S	Cl
29.46	19.56	50.98	62.38	3.69	12.39	1.09	0.82	0.07

[a] By difference.