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Coal rapid pyrolysis in a transport bed under steam-containing syngas atmosphere relevant to the integrated fluidized bed gasification

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HIGHLIGHTS

- A transport bed was used to study the effect of steam-containing syngas on coal rapid pyrolysis.
- Steam-containing syngas combined the advantages of steam and syngas on tar production.
- Steam-containing syngas atmosphere was beneficial to CH_4 production.

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ABSTRACT

An integrated fluidized bed (IFB) consisting of an upper transport bed section and a bottom fluidized bed section was adopted to investigate the transport bed coal pyrolysis by varying its reaction temperature and reaction atmosphere adjusted to simulate steam-containing syngas produced by the bottom fluidized bed char gasification. Steam and syngas, in comparison with N_2, as the reaction atmosphere little affected the tar yield below 600 °C but significantly decreased it for the former and increased it for the latter at rather higher temperatures. The presence of H_2 in the syngas increased tar yield significantly because it could suppress polymerization and condensation reactions through providing H as radical stabilizer and hydrogenation agent. In the steam-containing syngas atmosphere, the tar yield obtained from transport bed rapid pyrolysis increased rapidly with raising temperature to a peak value of 10.5 wt.% (daf) at 600 °C, about 1.1 wt.% higher than the Gray–King assay yield, and then decreased due to the excessive secondary reactions. Analyzing tar composition further showed that steam-containing syngas combined their respective advantages that syngas improved the yields of both light and heavy tars while steam reduced the heavy tar yield, especially at temperatures above 600 °C. The steam-containing syngas atmosphere also promoted CH_4 production in comparison with syngas atmosphere below 700 °C.

1. Introduction

The ever increasing demand of natural gas in China as well as the limited domestic supply provides a strong incentive to produce synthesis natural gas (SNG) from coal, especially low-rank coal [1]. Many SNG projects have actually been planned or in demonstration in China [2,3]. The Lurgi moving bed gasifier has been widely adopted for SNG production because its producer gas contains high-content CH_4 [4]. This gasifier also produces a considerable amount of tar which is highly demanded by actual industries due to the limited availability of crude oil in China [5,6]. Because Lurgi gasifiers adopts only lump coal above 6 mm, there is a great need to treat the more abundant powder coal [7], such as below 10 mm, via a Lurgi-type gasifier.

Accordingly, an integrated fluidized bed (IFB) consisting of an upper transport bed section and a bottom fluidized bed section has been tested to process low-rank powder coal and co-produce tar and CH_4-rich syngas [8]. This IFB process was motivated to treat powder coal via the Lurgi-type process, which incorporates coal pyrolysis into char gasification. As schematically shown in Fig. 1, coal is fed into the upper transport bed section to carry out fast pyrolysis, and the formed char is circulated into the bottom fluidized bed section for gasification. The produced gas from char gasification by oxygen-steam agent, which contains syngas and unreacted steam, supplies both sensible heat and reaction atmosphere for the coal pyrolysis in the upper transport bed section. Our previous study has verified the technical feasibility of the IFB

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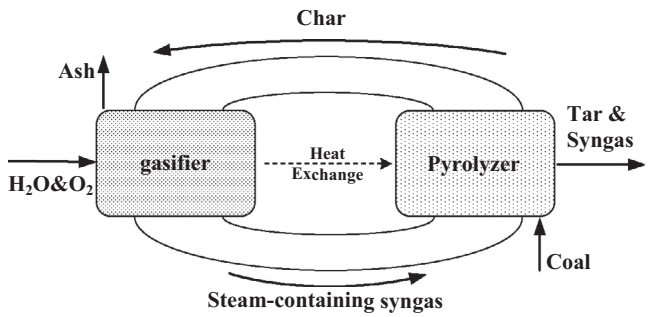

Fig. 1. Principle illustration of the integrated fluidized bed gasification process.

process for treating powder coal and producing CH_4-rich syngas [8]. At the appropriate operating conditions, 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 Luigi gasifier. This study focuses on the coal pyrolysis in the upper transport bed section which is subject to the interaction between coal and hot steam-containing syngas atmosphere from the bottom char gasification. In the Lurgi gasifer, the coal pyrolysis occurs with slow heating, as a result of its feeding at the low temperature top of gasifier and using the large coal particle sizes (lump coal). In the IFB reactor, the powder coal feeding is at the high-temperature bottom of the transport bed pyrolysis section to cause thus rapid heating of coal particles that can be around 1000 °C/s [9]. Understanding transport bed coal pyrolysis can also deliver knowledge of coal rapid pyrolysis.

Wen and Dutta [10] reported that at temperatures below 600 °C the rapid heating leads to high yield of total volatiles and high ratio of liquid to gas. Above 600 °C the secondary reactions of tar become significant, and increasing the final temperature with a particular heating rate is beneficial to total volatiles yield but decreases the product ratio of liquid to gas. In the IFB, the syngas from bottom gasification provides heat and also atmosphere for coal pyrolysis. Many studies have shown the influences of dry-syngas, coke-oven gas and pyrolysis gas on yield and quality of pyrolysis-generated tar in fixed bed and fluidized bed reactors [11–14], but there is almost no study on the transported bed rapid pyrolysis in steam-containing syngas (*in situ* wet syngas) atmosphere. Jüntgen [15] discussed the effect of H_2 on the evolution of tar during coal pyrolysis. When replacing inert gas with H_2, the pyrolysis causes additional formation of CH_4 and tar at the expense of coke. It is said radical groups are stabilized by H_2 to block radical recombination reactions and coke formation. Partial hydrogenation of polynuclear aromatics would also occur, while subsequent tar hydrocracking including demethylation, deoxygenation, hydroxylation and elimination of functional groups may lower the tar production to yield more CH_4 and light tar. Depending on operation conditions and reactor type, hydrogen would have different effects on tar production from coal pyrolysis [16–25]. The presence of CO in the atmosphere promotes tar production due to its inhibition on secondary reactions occurring to phenols [20,26]. Carbon dioxide (CO_2) can participate in reforming reactions of volatiles to affect tar formation and in char gasification to enhance production of no-condensable gases [20,21,27]. Steam in the atmosphere likely penetrates micro-pores in coal to facilitate desorption of volatile matter and recovery of tar species [28]. This is why steam is reported to enhance tar yield by its inhibiting secondary reactions at low temperature [13,28,29]. On the other hand, high temperature definitely enhances reforming reactions of nascent tar to cause tar loss [30], especially at high steam concentration [31].

The intention of this study is thus to explore the interaction of rapid coal pyrolysis and steam-containing syngas atmosphere to obtain the potential high yields of pyrolysis liquid and gas products. For this, the effects on the yield and quality of tar and production of CH_4 in the transport bed section of IFB are investigated with respect to the pyrolysis atmospheres composed of N_2, steam, simulated syngas, steam-containing simulated syngas and the major gas components containing in the syngas.

2. Experimental

Fig. 2 shows a schematic diagram of the experimental apparatus. The integrated fluidized bed (IFB) combines a transport bed pyrolysis section on the top and a fluidized bed gasification section on the bottom. In this study, only the upper transport bed with inner diameter of 40 mm and height of 6.0 m was employed as a coal pyrolyzer and the bottom fluidized bed packed with Al_2O_3 ball with diameter of 2–3 mm was used as a gas preheater. Coal feeding inlet is at 300 mm above the joint flange of the transport bed bottom. The gas supplying system was consisted of cylinders for N_2, H_2, CO, and CO_2, a gas preheater, a water pump and a steam generator for generating steam, and a gas mixer (also as the second gas preheater). When all the parts or sections of the IFB reached their desired temperatures, a gas stream mixing all components necessary was preheated in the bottom gas preheater and fed into the bottom of the IFB. The tested pyrolysis atmospheres are denoted as atmospheres of steam, simulated syngas and steam-containing simulated syngas. Their compositions are (in volume) 15% H_2O + 85% N_2, 40% H_2 + 30% CO + 15% CO_2 + 15% N_2 and 40% H_2 + 30% CO + 15% CO_2 + 15% H_2O, respectively. Other atmospheres are denoted by the names of individual gas components, like H_2, CO, CO_2 refer to the compositions of 50% N_2 + 50% individual gas (H_2, CO and CO_2).

The tested coal in this study 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 relatively high volatile matter (VM) and ash contents. The tar yield of Gray–King assay is 9.4 wt.% (daf). The coal sample was crushed to a size range of 0.3–0.4 mm and then dried in an atmospheric oven of 105 °C for 2 h before each test. The terminal velocity for coal particle with the sizes of 0.3–0.4 mm is about 1.7–2.6 m/s in the temperature range from 500 °C to 800 °C.

Coal particles were fed continuously via a screw feeder with a feeding rate of 1.2 kg/h. For each test, coal feeding was continued for about 1.0 h to ensure every parameter was at the desired stable state. The coal particles were conveyed pneumatically into the transport bed and heated by hot carrier gas to carry out rapid pyrolysis under various reaction conditions. The superficial gas velocity is about 3.1–4.1 m/s corresponding to a carrier gas flow of 80 L/min for guaranteeing coal particles to be entrained in the transport bed. The residence time of hot gases as well as coal particles is about 2 s. Volatiles released from coal together with char were conveyed by hot carrier gas out of the reactor. The char was separated from gas via a cyclone into a receiver. A metal filter was used to further separate the fine char. The quench cooler and the condenser allowed the majority of tar and steam to condense. The cotton filter was used for the removal of the remnants of fine particles. Most of the gas was scrubbed by a large amount of acetone to collect residual tar and then ventilated into the air, but a part of gas was scrubbed by several acetone traps to further remove residual tar for gas analysis.

The non-condensable gas dried by a silica gel column including N_2, H_2, CH_4, CO, CO_2, C2–C3 was metered by a wet gas flowmeter and sampled in every 4 min using gas bag. Then, the producer gas was analyzed by a micro GC (Agilent 3000A). Fig. 3 shows the outlet gas compositions as measured against time at 800 °C under syngas atmosphere. The gas compositions of N_2, H_2, CH_4, CO, CO_2, C2–C3 maintained a stable value during 1 h experiment. The average gas composition was calculated by arithmetic average method.