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Research article

Gasification of low-rank coal for hydrogen-rich gas production in a dual loop gasification system

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ABSTRACT

A dual loop gasification (DLG) system consisting of three separated reactors and two bed material circulation loops has been proposed for steam gasification of low-rank coal to obtain hydrogen-rich gas with low tar content. The system decouples the gasification process into fuel gasification, tar destruction and residual char combustion, which occur in three independent reactors correspondingly, i.e. a gasifier, a reformer and a combustor. Both the gasifier and the reformer are separately interconnected with the single combustor, forming two bed material circulation loops in parallel. In this way, both the gasifier and the reformer could be operated individually under optimized conditions. With Shenmu bituminous coal as feedstock and calcined olivine as both solid heat carrier and in-situ tar destruction catalyst, the performance of the system for the steam gasification of the coal has been investigated. It has been found that the tar was effectively removed under higher reformer temperature and in the presence of the olivine catalyst, and the coal gasification was promoted at higher gasifier temperature, S/C and ER.

1. Introduction

Coal will continue to be an important energy source contributing to the world's energy system in the foreseeable future, but its utilization has faced serious environmental issues [1,2]. To alleviate such problems, great efforts have been made to develop new technologies for clean and efficient utilization of coal. Gasification therein has been recommended to be an attractive choice as it is capable of efficiently converting coal into either clean energy, i.e. heat and electricity, or high value-added chemicals [3]. In particular gasification of low-rank coal under mild condition (atmosphere pressure and temperature lower than 1000 °C) has received great attention recently in consideration of operation cost, energy recuperation and ash-related problem [4,5].

Gasification with steam as gasification agent is a well-known process for hydrogen-rich gas production [6]. The process is highly endothermic and generally has to introduce air or oxygen as a part of gasification agent to maintain the process autothermic. It intrinsically includes a series of reactions, e.g. fuel pyrolysis, char gasification, tar destruction and combustion of carbon residues and combustible gases. In traditional gasification technologies, all the above-mentioned reactions occur in a single reactor. Hence, the reactions are closely interrelated with each other and against to be individually regulated to facilitate gasification performance, adapt fuel property and match downstream applications. Specifically, when air is used, the product gas will be seriously diluted by nitrogen introduced with the air.

In contrast, decoupled gasification separates these reactions into at least two reactors or zones, and thus provides an effective solution to promote the desired reactions or suppress those unexpected to facilitate the gasification performance [7,8]. Typically, in order to break the interaction between combustion and gasification to improve quality of product gas, the so-called dual bed gasification has been intensively investigated [9–12]. The gasification system separates the gasification and combustion reactions into two isolated reactors, i.e. a gasifier where fuel gasification occurs with steam as gasification agent, and a combustor where char combustion takes place with air as combustion agent. Bed material as solid heat carrier is circulating through the two reactors to carry the heat from the combustor to the gasifier. In this case, the flue gas from the combustor and the product gas from the gasifier are separated, and so that the hydrogen-rich product gas without dilution by nitrogen and combustion-generated carbon dioxide can be available. Typical example of the system is the so-called fast internally circulating fluidized bed (FICFB) gasification at Vienna University of Technology, Austria [13]. The system combines a bubbling fluidized bed reactor as fuel gasifier and a fast fluidized bed

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reactor as char combustor, which has been successfully demonstrated in Güssing [14] and Oberwart [15], Austria, respectively. Some similar designs can also be found in literatures [16–20].

Despite of great advantage for nitrogen-free and hydrogen-rich gas production, the system suffers from tar formation in product gas during coal gasification under mild condition, which will impose restriction on the end-use application of the gas as well as block downstream equipment [21]. In order to minimize the impact of such tar-related problems, various gas cleanup approaches have been attempted to decrease tar content in product gas [22]. Among these methods, in-bed catalytic destruction is a preferred option because of thermal integration and high tar conversion even at a low temperature [23]. Several kinds of in-bed catalysts for tar removal in dual bed gasification, e.g. natural minerals, alkali and alkali earth metals and supported transition metals, have been investigated [24–28]. Despite of inferior activity in tar removal, olivine, a naturally occurring iron-magnesium mineral with high attrition resistance and mechanical strength, is widely employed in dual fluidized bed gasifier [29].

In typical dual bed gasification system, the extent of tar destruction is limited even using in-situ tar destruction catalyst as bed material due to short residence time of volatiles in the gasifier and inadequate contact between the volatiles and the catalyst. That is essentially due to the fact that fuel gasification and tar destruction reactions are still interacted in the same space, making it impossible to enhance tar destruction for efficient tar removal. In this respect, decoupling tar destruction from the fuel gasification could provide a solution to optimize reaction condition of tar destruction. Accordingly, Xu et al. [30] proposed a two-stage dual fluidized bed gasification system, in which the single-stage bubbling fluidized bed gasifier is substituted by two stage fluidized bed gasifier. The product gas from the first stage is upgraded further in the second stage, which is favorable for increase of gasification efficiency and decrease of tar content. Göransson et al. [20] installed an in-situ reformer above the dense zone of the fluidized bed gasifier but under the hot bed material return position to intensify the contact of volatiles and catalytic bed material for efficient tar reforming. Our group [31] has developed a decoupled triple bed gasification (DTBG) system to improve tar destruction. In the system, tar destruction, fuel gasification and char combustion reactions are decoupled into three separated reactors, i.e. a reformer, a fuel reactor and a combustor, which are connected in series with the help of circulating solid heat carrier. In this way, tar destruction can be strengthened under optimized reaction conditions. Nevertheless, the coal gasification performance of the DTBG is still inferior to that of other gasification systems [13,17] in regards of gas yield, carbon conversion and cold gas efficiency due to limited coal gasification under mild condition. The key issue is that the fuel reactor temperature is restricted by the reformer temperature in one single circulation loop and then against for coal gasification.

In order to modify the DTBG, a dual loop gasification system (DLG) composed of three decoupled reactors and two bed material circulation loops has been proposed to improve both fuel gasification and tar destruction under mild condition. The two circulation loops, i.e. one char combustor-fuel gasifier loop for fuel gasification and the other char combustor-tar reformer loop for tar destruction, share the single combustor. In this way, the fuel gasifier and the tar reformer could be controlled individually, which provides a solution to strengthen both fuel gasification and tar destruction to benefit the performance of coal gasification. The system has already been employed to increase tar destruction in steam gasification of biomass [32]. This study aims to validate the feasibility of the DLG for steam gasification of low-rank coal under mild condition. With calcined olivine as both solid heat carrier and in-situ tar destruction catalyst, the effect of reaction condition on steam gasification performance of two low-rank coals has been investigated.

2. Experimental

2.1. Apparatus and procedure

The principle and lab-scale facility of DLG have been described in detail in previous publication [32]. Briefly, the system consists of three separated reactors, i.e. a gas-solid countercurrent moving bed gasifier where fuel is gasified with steam and/or oxygen, a gas-solid radial cross flow moving bed reformer where tarry product gas from the gasifier radially pass through the bed and further cracked and reformed, and a fast fluidized bed combustor where residual chars from the fuel gasifier and deposited cokes on the surface of the circulating bed material particles from the reformer are combusted with air. Two circulation loops, i.e. the char combustor-fuel gasifier loop between the gasifier and the combustor and the char combustor-tar reformer loop between the reformer and the combustor, are in parallel and share the single combustor. A cyclone following the combustor is used to separate the circulating bed material from the flue gas of the combustor. Sealing legs are separately set at the top joint zones between the cyclone and the gasifier or the reformer and the bottom between the combustor and the gasifier or the reformer to prevent the undesired gas leakage, and so that the hydrogen-rich product gas and the flue gas are isolated. The distribution of bed material into the gasifier and the reformer was achieved by three mechanical valves separately set on the top sealing leg of the gasifier and the bottom sealing legs of the gasifier and the reformer. All the reactors are made of 310S stainless steel and externally heated by independent electrical furnaces to compensate heat loss. The temperature of each reactor is recorded by a K-type thermocouple placed at the middle part of the reactor. Manometers are placed at various points of the reactors to indicate the pressure profiles. Specifically in the reformer, a differential manometer is installed to monitor the gas pressure drop through the lateral particle layer and to insure the operation to be normal. The specific operating conditions of the experiments are shown in Table 1.

2.2. Feedstock and bed materials

In this study, a Shenmu bituminous coal (SB) was adopted as feedstock, and an Inner Mongolia lignite (IL) was used for comparison (see Section 3.3). Before each experiment, the coal was crushed and sieved to an average particle size of 0.38–0.83 mm, and then dried at 105–110 °C for 3 h. Table 2 presents the results of the proximate analysis, ultimate analysis and lower heating value of the coals.

Olivine from Yichang, China, was used as the bed material and catalyst. Before test, the olivine was crushed and sieved to an average

Table 1
Operating conditions of the DLG

Total weight of bed material (kg)	7.2
Gasification circulation ratio (C/F)	10
Reforming circulation ratio (C/F)	10
Bed height in the gasifier (mm)	100
Residence time of solid in the gasifier (min)	20
Residence time of solid in the reformer (min)	45
Steam to carbon mass ratio(S/C) (kg/kg)	0.2–2.0
Coal feeding rate (kg/h)	0.2
Gasifier temperature (°C)	750–850
Reformer temperature (°C)	700–850
Combustor temperature (°C)	850
Gauge pressure in the gasifier (Pa)	0
Gauge pressure in the reformer (Pa)	− 100 to − 50
Gauge pressure in the cyclone (Pa)	0
Gauge pressure in the pre-fluidizer(Pa)	0