Research article

Biomass steam gasification for hydrogen-rich gas production in a decoupled dual loop gasification system

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A B S T R A C T

In order to improve tar destruction and hydrogen-rich gas production in steam gasification of biomass, a novel decoupled dual loop gasification system (DDLG) has been proposed. In the system, fuel pyrolysis/gasification, tar cracking/reforming and char combustion are decoupled into three reactors correspondingly, i.e. fuel reactor, reformer and combustor. Both the fuel reactor and the reformer are separately interconnected with the combustor, forming two bed material circulation loops, one for fuel pyrolysis/gasification and the other for tar cracking/ reforming. In this way, the above-mentioned reactions could be optimized separately under appropriate conditions. With pine sawdust as feedstock and calcined olivine as both solid heat carrier and in-situ tar cracking/ reforming catalyst, the steam gasification performance of the system has been investigated. It has been indicated that the configuration of the system provides an effective way to strengthen the tar cracking/reforming reaction for efficient tar removal. Specifically, a product gas with hydrogen concentration of 40.8 vol%, tar content as low as 14.1 g/Nm^3 and dry gas yield of 1.0 Nm^3/kg daf was obtained at the fuel reactor temperature 800 °C, reformer temperature 850 °C, steam to carbon mass ratio 1.2 and circulation ratio 10.

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1. Introduction

In view of depletion of fossil fuels and environmental problems, biomass as a clean and renewable alternative energy for fossil fuels has drawn great concern in recent years [1,2]. Among all thermo-chemical conversion processes, gasification is considered to be one of the most promising routes to convert biomass into versatile syngas due to its high conversion efficiency and wide application of the gas, which can be used as either gaseous fuels for heat and power generation or feedstock for chemical synthesis and hydrogen production [3].

Gasification intrinsically involves a series of reactions including fuel pyrolysis, char gasification, carbon residues combustion and tar cracking/reforming. Commercial gasification technologies such as moving bed gasification, fluidized bed gasification and entrained flow gasification, despite of their relative maturity, fail to manipulate the above-mentioned reactions individually to match feedstock property and downstream applications since all of these reactions are mutually interactive to occur in a single space. Specifically, in the circumstance when air is used as gasification agent, nitrogen introduced with air and carbon dioxide generated in combustion will dilute the product gas.

Alternatively, decoupled gasification characterized by isolating and then reorganizing one or some of such reactions has the potential to strengthen the desired reactions and suppress those unexpected to

facilitate the gasification performance [4–6]. Typically, the so-called dual bed gasification has been developed by researchers [7–16]. In these systems gasification and combustion reactions take place in two isolated reactors, i.e. a fuel gasifier and a char combustor, respectively. Solid heat carrier is circulating between the two reactors, while the flue gas from the combustor and the product gas from the gasifier are separated. By this way, nitrogen-free and hydrogen-rich gas can be generated using air as combustion agent and steam as gasification agent. It also provides possibilities to optimize the both reactions under suitable operating conditions respectively. For example, Hofbauer et al. [17] from the Vienna University of Technology has developed a fast internally circulating fluidized bed (FICFB) gasification process to produce high quality product gas for heat and power generation as well as fuels for transportation and synthetic chemicals from biomass. It consists of a bubbling fluidized bed gasification zone, where biomass steam gasification takes place, and a fast fluidized bed combustion zone, where the residual char from the gasification zone is combusted with air to generate heat for the gasification. The process has been successfully demonstrated at an industrial scale of 8 MW combined heat and power plant in Güssing, Austria since 2001. Xu et al. [18] from the Institute of Process Engineering of Chinese Academy of Science, China, have developed a dual fluidized bed gasification (DFBG) system. The system combines a dense low-velocity fluidized bed fuel gasifier and a high-velocity pneumatic riser char combustor, which has already been employed for gasification of dried coffee grounds to produce medium heating value gas free of nitrogen in a 5.0 kg/h pilot facility. Similarly, Matsuoka et al.

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[19] from Energy Technology Research Institute, National Institute of Advanced Industrial Science and Technology, Japan, has proposed a new circulating dual bubbling fluidized bed (CDBFB) gasification system for woody biomass gasification to increase calorific value of gaseous product. The system consisted of two bubbling fluidized beds, one serving as a gasifier and the other as a combustor. Compared to the riser combustor, the bubbling fluidized bed combustor has longer residence time of solid for sufficient combustion. Nevertheless, in normal dual bed gasification system, residence time of gaseous product in bubbling fluidized bed gasifier is limited and insufficient for tar destruction.

In order to improve tar destruction, considerable efforts have been tried to increase the residence time of gaseous product in the gasifier of dual fluidized bed gasification system. Hofbauer et al. [20] have developed a novel dual fluidized bed gasification system in which the classical bubbling fluidized bed gasifier is substituted by a countercurrent reactor with multi-staged zones of solid accumulation operated in turbulent fluidized bed regime. In this case, significant improvement in gas-solid contact and residence time of gas and solid can be achieved, which is beneficial for tar cracking/reforming reaction. Xu et al. [21] adopted two-stage fluidized bed gasifier to replace the single-stage bubbling fluidized bed gasifier in a normal dual fluidized bed gasification system. 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. [22] installed an in-situ reformer (upper fluidized bed) in the freeboard region of the fluidized bed gasifier to intensify the contact of volatiles and catalytic bed material for efficient tar reforming. Our group has adopted co-current or countercurrent moving bed gasifier [23,24] to improve the quality of the product gas.

In addition, tar produced during gasification also can be converted by using in-situ catalytic bed material (catalyst) inside the gasifier [25–34], which is proved to be effective for tar removal and meanwhile hydrogen-rich gas production. Despite of inferior activity in tar elimination, natural minerals are suitable as the in-situ catalysts in the perspective of low cost and wide availability. Specifically, a natural iron-bearing olivine with advantages of high attrition resistance and mechanical strength is a preferred candidate in dual fluidized bed gasifier [35–37]. It has already been successfully employed in the FICFB commercial plant located at Güssing, Austria [38].

Still, in the dual bed gasification system even with in-situ catalyst, the tar destruction is unsatisfactory. That is essentially due to the fact that pyrolysis/gasification and tar cracking/reforming reaction are still mutually interactive in the same space, which makes it difficult to strengthen the tar cracking/reforming reaction to realize a deep conversion of the tar. In this sense, we have proposed a gasification system composed of triple isolated reactors, i.e. a fuel reactor, a tar reformer and a char combustor. Especially, the system has adopted a so-called radial cross flow moving bed reformer with the feature of longer residence time of volatiles and excellent contact between the tar and the catalyst to enhance the tar conversion [39,40]. Nevertheless, it is still impossible to realize independent regulation and control of the fuel pyrolysis/gasification and the tar cracking/reforming due to the fact that the reformer and the fuel reactor line in a single circulation loop. In particular, the fuel reactor temperature is restricted by the reformer temperature.

To overcome the shortcomings of the triple bed gasification system, a novel decoupled dual loop gasification system (DDLG) has been proposed. In the system, the fuel reactor and the reformer share the single combustor and are separated into two circulation loops, i.e. the char combustor-fuel reactor loop and the char combustor-tar reformer loop, and so that the fuel reactor and the tar reformer could be controlled individually. The fuel reactor could be operated in either pyrolysis mode (named as pyrolyzer) or gasification mode (gasifier) to match variety of feedstocks. The tar generated in the pyrolyzer/gasifier is further cracked/reformed in the reformer. The circulating bed material could be inert as heat carrier or catalytic to strength the tar cracking/reforming. This study reports some preliminary experimental results of the system at laboratory scale where the fuel reactor is operated in gasification mode. With pine sawdust as feedstock, the influence of reaction conditions including fuel reactor temperature, reformer temperature, steam to carbon mass ratio (S/C) and type of bed materials on the steam gasification performance of the DDLG has been investigated.
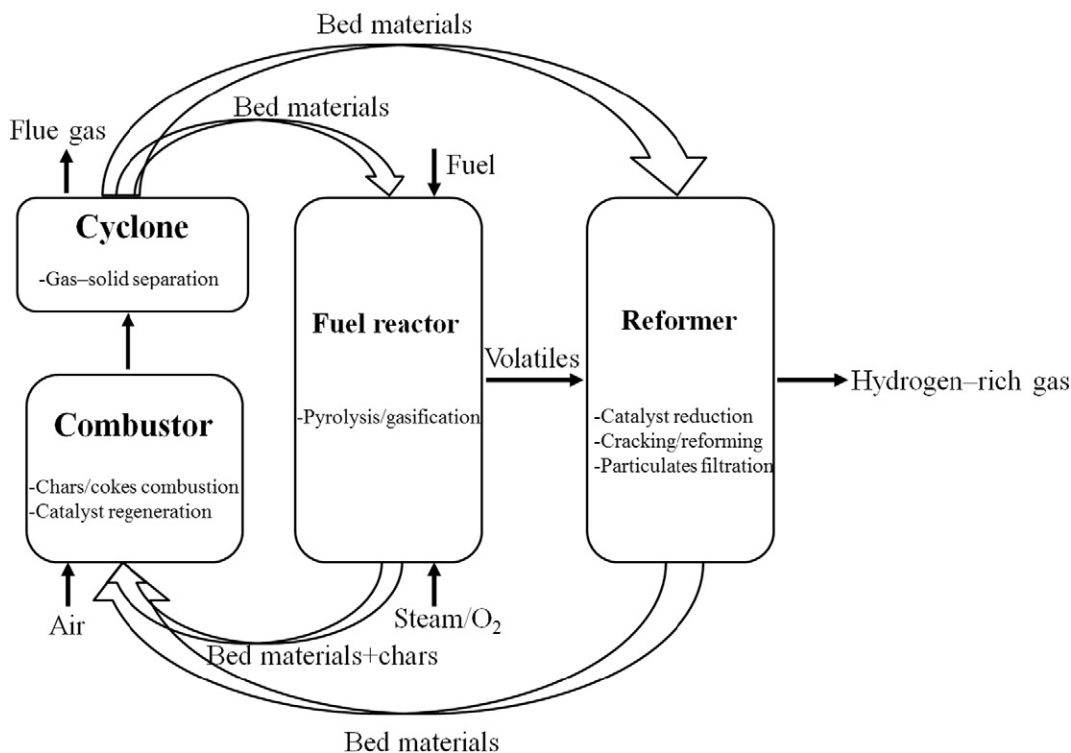

Fig. 1. Principle of DDLG.

2. Experimental

2.1. Experimental apparatus and procedure

The principle of DDLG is shown in Fig. 1. It consists mainly of three separated reactors, i.e. a fuel reactor where fuel pyrolysis/gasification takes place, a reformer where tar/hydrocarbons in volatiles from the fuel reactor is further cracked/reformed, and a combustor where residual chars from the fuel reactor and deposited cokes on the surface of the circulating bed material particles from the reformer are combusted with air. Both the fuel reactor and the reformer are in parallel and separately interconnected with the combustor, forming two circulation loops. One is the char combustor-fuel reactor loop, and the other is the char combustor-tar reformer loop. Bed material as both solid heat carrier and in-situ tar cracking/reforming catalyst is circulating among these reactors. Following the combustor, there is a cyclone to separate the circulating bed material from the flue gas of the combustor, and so that the hydrogen-rich gas of the reformer and fuel reactor are isolated from the flue gas of the combustor. As solid heat carrier, the circulating bed material transfers heat generated from the combustor to both the reformer and the fuel reactor to provide the energy needed for the cracking/reforming and pyrolysis/gasification reactions. When catalytic bed material (catalyst) is used, it also acts as in-situ tar cracking/reforming catalyst, which undergoes continuously regeneration and reaction along with the circulation loops. In the case a metal oxide catalyst is used as bed material, e.g. olivine, it will undergo reduction in the reformer by contacting with the hydrogen-rich gas. During the reduction, Fe_2O_3 on the surface of the calcined olivine is reduced to $FeO_{1.5-x}$ ($0 < x \leq 1.5$) [37], which has been proved to be active for tar cracking/reforming, and part of the hydrogen-rich gas simultaneously is oxidized to H_2O and CO_2. The fuel reactor could be operated in either pyrolysis mode (named as pyrolyzer) or gasification mode (gasifier) to match variety of feedstocks. To increase the contact between tar-containing volatiles and catalyst for tar destruction at a smaller pressure drop, a radial cross flow moving bed reformer is adopted. It also acts as particulates filter to capture dust in the product gas.

Fig. 2 is the schematic diagram of DDLG facility. The fuel reactor, herein as a gasifier, is a gas-solid countercurrent moving bed reactor with an 80 mm i.d. and a 200 mm height. The reformer is a gas–solid radial cross flow moving bed reactor with a 136 mm i.d. and a 400 mm height, and has an annular bed with an i.d. of 20 mm and an o.d. 90 mm in the center. A horizontal connection pipe between the top of the fuel reactor and that of the reformer allows for flow of pyrolysis gas from the fuel reactor into the central gas channel of the reformer. The combustor is a fast fluidized bed riser with a 26 mm i.d. and a 2500 mm height. Sealing legs are separately set at the top and bottom joint zones between the combustor and the fuel reactor or the reformer to prevent the undesired gas leakage. All the reactors are constructed by 310S stainless steel and externally heated by independent electrical furnaces to compensate heat loss. The mean temperature of each reactor is monitored by a K-type thermocouple placed at the middle part of the reactor. Manometers are used to indicate the pressure profiles of each reactor and ensure the gas streams from each reactor are separated. 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.

Prior to test, about 7.2 kg of bed material as well as catalyst were added into the gasification system. Then all of the reactors were
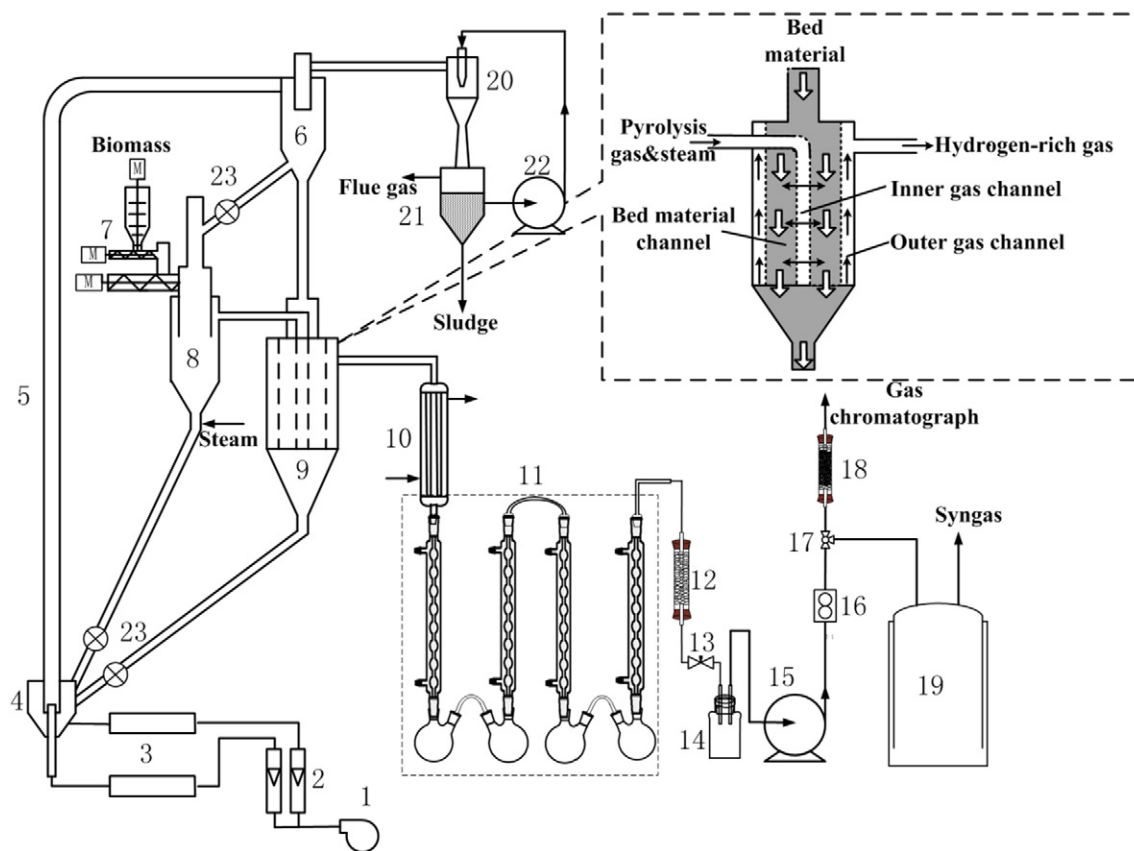

Fig. 2. Schematic diagram of DDLG facility. 1-Air compressor, 2-Gas flow meter, 3-Air pre-heater, 4-Pre-fluidizer, 5-Riser combustor, 6- Cyclone separator/bunker, 7-Screw conveyor, 8-Fuel reactor, 9-Reformer, 10-Pipe bundle condenser, 11-Condenser, 12-Cotton wool filter, 13-Needle valve, 14-Surge flask, 15-Vacuum pump, 16-Gas meter, 17-Three way valve, 18-Silica gel filter, 19-Gas holder, 20- Venturi gas scrubber, 21-Water tank, 22-Circulating water pump, 23-Mechanical valve.

Table 1
Operating conditions of the DDLG.

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

electrically heated under nitrogen purging. A primary fluidization air about 3.8 Nm³/h and a secondary fluidization air about 2.5 Nm³/h were introduced into the combustor to fluidize the bed material in it and to ensure the circulation of the bed material among reactors. The circulating rate of bed material in each circulating loop was controlled to 2.0 kg/h by three mechanical valves separately set on the top sealing leg of the fuel reactor and the bottom sealing legs of the fuel reactor and the reformer.

When all the reactors reached to the desired temperatures, the biomass particles were fed into the fuel reactor at a feeding rate of 200 g/h, meanwhile steam was injected into the bottom of the fuel reactor with a certain flow rate. With the help of a vacuum pump, the product gas was extracted from the reformer and subsequently cooled down in sequential ice-cooled and glycol-cooled quenchers to capture the condensable components in it. The condensed liquids were collected in tar traps. The aerosol in the gas was further captured in a filter filled with cotton wool. The dry product gas volume was determined by the gas flow meter. Through a venturi gas scrubber, the gas from the cyclone was cooled down and the dust in it was scrubbed by water spray. The detailed operating conditions were summarized in Table 1.

2.2. Feedstock and bed materials

Pine sawdust with a particle size of 0.38–0.83 mm was used as biomass feedstock. Prior to test, the pine sawdust was dried at temperature of 105–110 °C for 3 h. The properties of pine sawdust are given in Table 2.

Olivine of 0.38–0.83 mm from the Institute of Olivine Research, Yichang, China, was used as catalytically active bed materials. Before

Table 2
Properties of pine sawdust.

Proximate analysis[a] (wt%, ad)	
Moisture	9.0
Ash	0.6
Volatile	77.8
Fixed carbon	12.6
Ultimate analysis[b] (wt%, daf)	
Carbon	47.75
Hydrogen	6.98
Oxygen[d]	44.84
Nitrogen	0.07
Sulfur	0.36
LHV[c](MJ/kg, d.b.)	18.73

[a] Conducted by ASTM D 3172.
[b] Performed by a vario EL III elemental analyzer.
[c] Calculated by Dulong's formula.
[d] Calculated by difference.

experiments, the olivine was calcined at 900 °C for 4 h. The chemical composition of the olivine analyzed by X-ray Fluorescence (XRF) is presented in Table 3. Silica sand with the same particle size as that of the olivine was used as inert bed material in blank test for comparison.

2.3. Sampling and analysis

About 1 h after the start of gasification, when the gasification system reached a steady state, the product gas was sampled with gas bag every 15 min. The composition of the gas sample was analyzed by a gas chromatograph GC7900 equipped with a thermal conductivity detector (TCD) and a flame ionization detector (FID). The compositions of dry gas excluding nitrogen presented in this paper are averaged values of at least three samplings with respect to time on stream. After each test, liquid products including tar and water were washed with solvent tetrahydrofuran. The solvent was removed by evaporation at 40 °C under reduced pressure. The water was separated from the liquid mixture using ethyl acetate extraction, and then the tar was obtained by evaporating off the ethyl acetate solvent at 45 °C under reduced pressure. Specification of the tar sampling and analysis can also be found in detail elsewhere [39,40]. To evaluate the performance of the process, the parameters are defined by the following formula:

$$
\text{Dry gas yield} \left(Nm^3/kgdaf \right)
$$
$$
= \frac{\text{Volume of the gas dry product gas} \left(Nm^3 \right)}{\text{Mass of biomass of dry ash-free basis fed into the system} \left(kg \right)} \quad (1)
$$

$$
\text{Tar yield} \left(\% \right) = \frac{\text{Volume of the gas dry product gas} \left(Nm^3 \right)}{\text{Mass of biomass of dry ash-free basis fed into the system} \left(g \right)} \times 100 \quad (2)
$$

$$
\text{Tar content of product gas} \left(g/Nm^3 \right)
$$
$$
= \frac{\text{Mass of tar collected in the test} \left(g \right)}{\text{Volume of the gas dry product gas} \left(Nm^3 \right)} \quad (3)
$$

$$
\text{Cold gas efficiency} \left(\% \right)
$$
$$
= \frac{\text{Lower heat value of product gas} \left(kJ/Nm^3 \right) \times \text{Gas yield} \left(Nm^3/kJ \right)}{\text{Lower heat value of biomass fed into the system} \left(kJ/kg \right)}
$$
$$
\times 100 \quad (4)
$$

$$
\text{Carbon conversion} \left(\% \right)
$$
$$
= \frac{\text{Mass of carbon in the product gas} \left(kg \right)}{\text{Mass of carbon in biomass fed into the system} \left(kg \right)} \times 100 \quad (5)
$$

$$
\text{Water conversion} \left(\% \right) \quad (6)
$$
$$
= \frac{\text{Mass of water introduced into the} \left(g \right) - \text{Mass of water collected in the test} \left(g \right)}{\text{Mass of water introduced into system} \left(g \right)} \times 100
$$

$$
\text{Steam to carbon mass ratio} \left(S/C \right)
$$
$$
= \frac{\text{Mass of steam introduced into the system} \left(kg \right)}{\text{Mass of carbon in biomass fed into the system} \left(kg \right)} \quad (7)
$$

$$
\text{Circulation ration} \left(C/F \right) \quad (8)
$$
$$
= \frac{\text{Mass of bed material into the fuel reactor or reformer per hour} \left(kg/h \right)}{\text{Mass of biomass fed into the system per hour} \left(kg/h \right)}
$$

Table 3
Chemical composition of olivine by XRF analysis (wt%).

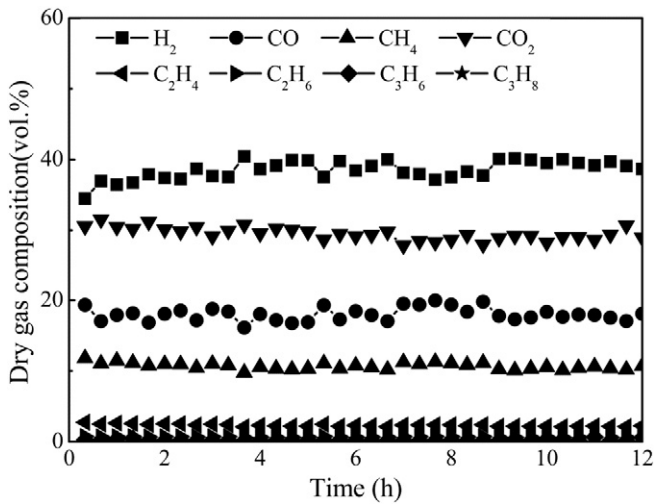| MgO	SiO₂	Fe₂O₃	Al₂O₃	Cr₂O₃	CaO	NiO
51.80	36.50	9.14	0.88	0.60	0.37	0.36

Fig. 3. Variation of dry gas composition with time on stream (biomass feeding rate 120 g/h, fuel reactor temperature of 800 °C, reformer temperature 850 °C, S/C 1.2 and circulation ratio 10).

3. Results and discussion

3.1. Stability test

In order to validate the reliability of the system, a 12 h long-term operation of the system for biomass steam gasification was examined at the biomass feeding rate 120 g/h, fuel reactor temperature 800 °C, reformer temperature 850 °C, S/C 1.2 and circulation ratio 10. The variation of dry gas composition with time on stream is shown in Fig. 3. H_2, CO, CO_2 and CH_4 are the main components in the product gas. The H_2 and CO_2 concentrations gradually increase whereas those of the CO and CH_4 decrease at the initial 40 min. There is no significant change on dry gas composition with the time on stream during the following 11 h. The concentrations of the H_2, CO, CO_2 and CH_4 in the dry gas keep steadily at 38.5 vol%, 18.0 vol%, 29.5 vol% and 10.7 vol%, respectively. Other trace gases, i.e. C_2H_4, C_2H_6, C_3H_6 and C_3H_8, are 2.3 vol%, 0.7 vol%, 0.3 vol% and 0.03 vol%, respectively. The results demonstrate that the system is of good stability and reliability for biomass steam gasification. In this case, all of the experiments in this study were kept for 2 h and the product gas was sampled at a steady state.

3.2. Effect of fuel reactor temperature

In the configuration of DDLG, the volatiles from the fuel reactor are further cracked/steam-reformed in the reformer. In order to investigate the effect of fuel reactor temperature on biomass steam gasification, experiments were conducted by varying the fuel reactor temperature from 700 °C to 850 °C, keeping the reformer temperature constant at 700 °C, S/C 1.2 and circulation ratio 10. As shown in Fig. 4, with the increase of the fuel reactor temperature, the dry gas yield, cold gas efficiency, carbon conversion and water conversion increase remarkably, although the water conversion is limited under a low level especially at lower temperature. The concentration of H_2 in product gas increase sharply, and the concentrations of CO, CH_4 and CO_2 show a little decrease. It indicates that the devolatilization (pyrolysis) of biomass, the steam gasification of char and the Boudouard reaction have been promoted at the higher fuel reactor temperature. The contribution of the water gas shift reaction leads to the decrease of CO and the increase of H_2/CO ratio. With the increase of the fuel reactor temperature, the tar
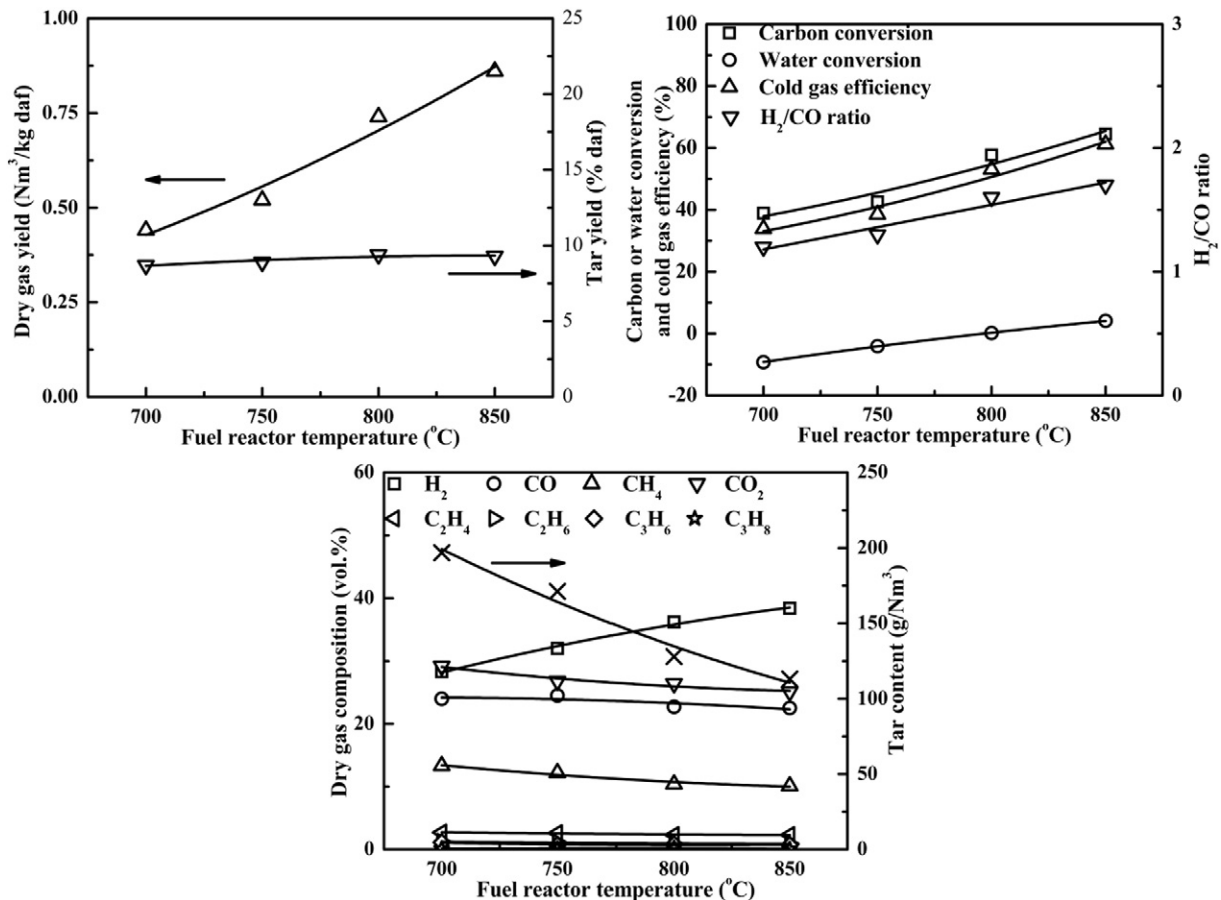

Fig. 4. Effect of fuel reactor temperature on biomass steam gasification (reformer temperature 700 °C, S/C 1.2 and circulation ratio 10).
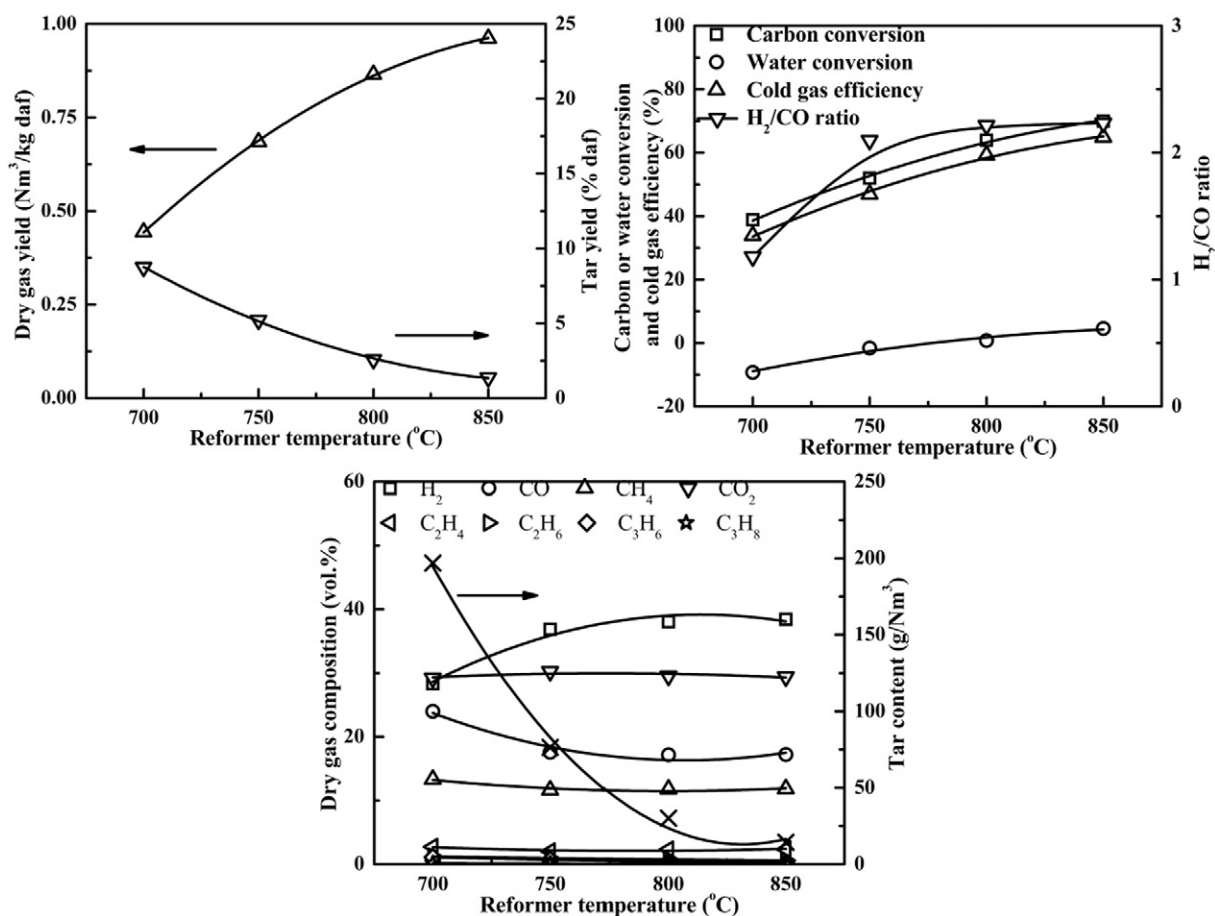
Fig. 5. Effect of reformer temperature on biomass steam gasification (fuel reactor temperature 700 °C, S/C 1.2 and circulation ratio 10).

content in product gas decreases obviously but the tar yield exhibits a little increase. It suggests that the tar cracking/reforming reaction is restricted because of the insufficient residence time of the volatiles from biomass pyrolysis in the fuel reactor.

As discussed above, biomass devolatilization/gasification makes a great contribution to the biomass gasification, and there still remains a large amount of tar in the gas from the fuel reactor. In order to increase the gas yield and decrease the tar in it, the tar cracking/reforming should be strengthened. In the DDLG, these reactions would be enhanced under optimized reformer temperature and S/C, and in presence of catalytic bed materials.

3.3. Effect of reformer temperature

Biomass steam gasification as a function of varied reformer temperature was investigated at fuel reactor temperature 700 °C, S/C 1.2 and circulation ratio 10. The results are shown in Fig. 5. Higher reformer temperature gives rise to dramatic increase of the dry gas yield and decrease of the tar yield. The carbon conversion and cold gas efficiency meanwhile increase significantly. Water conversion is at a low level but still increases clearly with the reformer temperature. In product gas, H_2 concentration increases while the concentrations of CO_2 and CO decrease slightly. It confirms that endothermic reactions such as cracking and steam/dry reforming of tar have been strengthened with the increase of reformer temperature, which leads to the increase of H_2 at the expense of tar, H_2O and CO_2. The downward tendency of the CO concentration exhibits that the water gas shift reaction plays an important role in this reformer temperature range, which was also found by Koppatz et al. [41] and Franco et al. [42].

3.4. Effect of S/C

The effect of S/C on biomass steam gasification was investigated in the S/C range of 0.2 to 2.0, fixing the fuel reactor temperature at 800 °C, reformer temperature 850 °C and circulation ratio 10. As shown in Fig. 6, with the increase of S/C, the dry gas yield, carbon conversion, water conversion and cold gas efficiency show a significant increase. H_2 concentration in the product gas ascends gradually while those of CO and CH_4 descend slowly, which results in H_2/CO ratio rising linearly with the S/C. The tar yield and tar content in product gas display a little increase with the increase of S/C. It is reasonable to believe that the gas yield improvement and gas composition variation are mainly caused by the char steam gasification and water gas shift reaction promoted by steam addition. Nevertheless, the unreacted steam and the incremental gas yield decrease the residence time of the volatiles in both the fuel reactor and the reformer, which restrains the tar cracking/reforming to some extent. For a practical application, excessive steam addition should be avoided because it will consume a large amount of energy and deteriorate the gasification condition.

3.5. Effect of bed materials

Bed materials, especially the catalytically active materials, may have a significant impact on tar cracking/reforming. The behavior of olivine as bed material compared with that of silica sand for biomass steam gasification was investigated at the fuel reactor temperature 800 °C, reformer temperature 850 °C, S/C 1.2 and circulation ratio 10. The results are shown in Table 4. Compared with the silica sand, the olivine as bed material shows a superior gasification performance in terms of
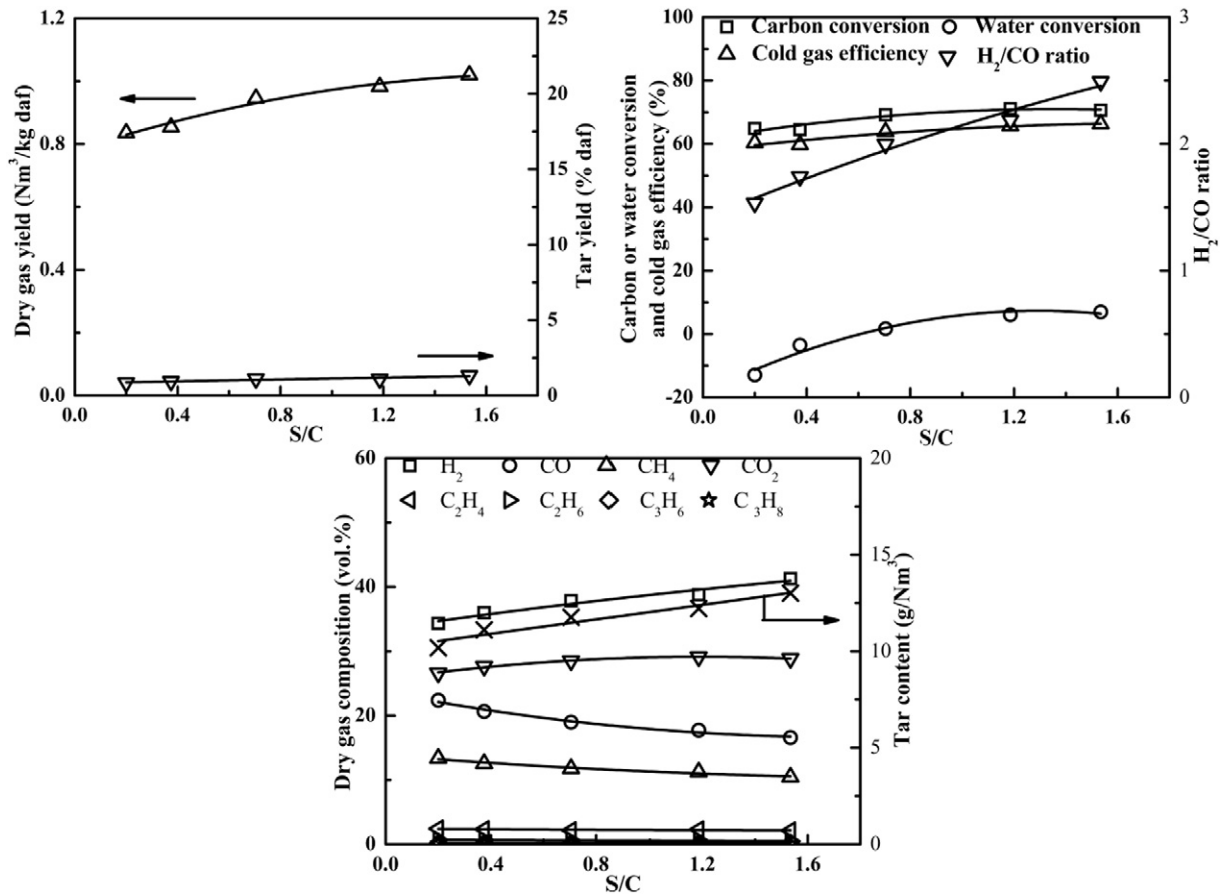
Fig. 6. Effect of S/C on biomass steam gasification (fuel reactor temperature 800 °C, reformer temperature 850 °C and circulation ratio 10).

the dry gas yield, carbon conversion, water conversion, cold gas efficiency and tar yield. The concentration of H_2 in the gas is higher and that of light hydrocarbons are lower. Especially, the dry gas yield increases by 17% and the tar yield decreases by 73%. It indicates that the olivine has a superior catalytic activity for tar cracking/reforming, which is ascribed to the FeO_x species on the olivine. The CO is lower and CO_2 is higher, suggesting that water gas shift reaction has also been catalyzed by the olivine.

As discussed above, in the DDLG, the separation of the fuel reactor and the reformer makes them possible to be run under optimized condition. Especially the tar from the pyrolysis/gasification could be further cracked/reformed in the reformer at suitable temperature and with extended residence time of the volatiles and excellent contact between the volatiles and the olivine catalyst. As a result, a hydrogen-rich product gas with low tar content could be obtained. The gasification performance of the DDLG at similar operating conditions but relative lower S/C is comparable to that of the typical dual bed gasification process, e.g. FICFB [41], in regards of H_2 concentration, tar content, dry gas yield, carbon conversion, water conversion and cold gas efficiency.

A comparison of the DDLG to the other similar three-bed decoupling gasification systems, e.g. T-DFBG [21], MIUN [22] and DTBG [39], is also made in Table 4. In terms of hydrogen concentration and tar content in the gas, the performance of DDLG with olivine as bed material is at the same level as that of the other processes with olivine as bed material under similar operating condition but superior to that of the T-DFBG with inert silica sand as bed material.

Nevertheless, similar to other steam gasification systems [17,43], the water conversion in the DDLG is still poor. Specifically, negative water conversion values were obtained under mild reaction conditions, i.e. the lower fuel reactor or reformer temperature and S/C. That could be attributed partially to the water generation from the pyrolysis of pine sawdust.

In addition, the reduction of the catalytic bed material olivine contributes greatly to the water formation. In the reformer, the reduction of the olivine from the combustor yields water at the cost of hydrogen in the product gas, which is going to be studied in the future.

Table 4
Gasification performances of DDLG and other similar gasification systems with silica sand and olivine as bed material.

Gasification system	DDLG		FICFB	DTBG	MIUN	T-DFBG
Bed material	Silica sand	Olivine	Olivine	Olivine	Olivine	Silica sand
Fuel reactor temperature (°C)	800	800	850	600	850	820
Reformer temperature (°C)	850	850	–	800	850	820
S/C	1.2	1.2	1.7	1.3	1.2	1.1
C/F	10	10	–	22	–	–
Dry gas composition (vol%)						
H_2	35.0	40.8	40.0	40.4	32.9	23.7
CO	22.0	17.1	26.0	12.5	36.1	36.6
CH_4	13.2	10.8	10.0	10.3	11.8	15.8
CO_2	25.2	28.6	18.0	34.0	14.5	12.4
C_2H_4	3.7	2.0	2.0	2.0	4.2	7.6
C_2H_6	0.7	0.5	0.3	0.5	0.3	3.8
C_3H_6	0.3	0.2	<0.1	0.2	<0.1	0.1
C_3H_8	<0.1	<0.1	0.5	<0.1	<0.1	<0.1
H_2/CO ratio	1.6	2.4	1.5	3.2	0.9	0.6
Dry gas yield (Nm³/kg daf)	0.87	1.02	1.10	0.84	–	–
Tar yield (%)	5.2	1.4	–	–	–	–
Tar content (g/Nm³)	59.8	14.1	7.5	11.1	26.0	32.0
Carbon conversion (%)	68.1	73.3	67.0	54.0	–	70.0
Water conversion (%)	2.2	6.6	6.0–7.0	–	–	–
Cold gas efficiency (%)	66.6	68.9	70	53.0	–	80.0

4. Conclusions

The rationality of the decoupled dual loop gasification system for biomass steam gasification has been validated with pine sawdust as feedstock and calcined olivine as the circulating bed material. The pyrolysis/gasification of pine sawdust is improved markedly by increasing the fuel reactor temperature. In comparison, the reformer temperature plays more effect on tar cracking/reforming. The dry gas yield increases and tar yield decreases evidently at the higher reformer temperature. The concentration of H_2 increases and that of CO decreases with the S/C, which makes it possible to flexibly adjust H_2/CO ratio. Compared with silica sand as bed material, olivine shows a superior catalytic activity for tar cracking/reforming, which favors tar removal and hydrogen-rich gas production.

Acknowledgments

References

[1] G. Berndes, M. Hoogwijk, R. van den Broek, The contribution of biomass in the future global energy supply: a review of 17 studies, Biomass Bioenergy 25 (2003) 1–28.
[2] P. McKendry, Energy production from biomass (part 1): overview of biomass, Bioresour. Technol. 83 (2002) 37–46.
[3] A.V. Bridgwater, Renewable fuels and chemicals by thermal processing of biomass, Chem. Eng. J. 91 (2003) 87–102.
[4] J.W. Zhang, Y. Wang, L. Dong, S.Q. Gao, G.W. Xu, Decoupling gasification: approach principle and technology justification, Energy Fuel 24 (2010) 6223–6232.
[5] J.W. Zhang, R.C. Wu, G.Y. Zhang, C.B. Yao, Y.M. Zhang, Y. Wang, G.W. Xu, Recent studies on chemical engineering fundamentals for fuel pyrolysis and gasification in dual fluidized bed, Ind. Eng. Chem. Res. 52 (2013) 6283–6302.
[6] C.Z. Li, G.W. Xu, Decoupled thermochemical conversion-preface, Fuel 112 (2013) 607–608.
[7] H.J. Ge, W.J. Guo, L.H. Shen, T. Song, J. Xiao, Biomass gasification using chemical looping in a 25 kWth reactor with natural hematite as oxygen carrier, Chem. Eng. J. 286 (2016) 174–183.
[8] T. Song, J. Wu, L.H. Shen, J. Xiao, Experimental investigation on hydrogen production from biomass gasification in interconnected fluidized beds, Biomass Bioenergy 36 (2012) 258–267.
[9] G.Q. Guan, C. Fushimi, A. Tsutsumi, Prediction of flow behavior of the riser in a novel high solids flux circulating fluidized bed for steam gasification of coal or biomass, Chem. Eng. J. 164 (2010) 221–229.
[10] L.G. Wei, S.P. Xu, J.G. Liu, C.L. Lu, S.Q. Liu, C.H. Liu, A novel process of biomass gasification for hydrogen-rich gas with solid heat carrier: preliminary experimental results, Energy Fuel 20 (2006) 2266–2273.
[11] K. Göransson, U. Söderlind, W. Zhang, Experimental test on a novel dual fluidized bed biomass gasifier for synthetic fuel production, Fuel 90 (2011) 1340–1349.
[12] K. Göransson, U. Söderlind, P. Engstrand, W. Zhang, An experimental study on catalytic bed materials in a biomass dual fluidised bed gasifier, Renew. Energy 81 (2015) 251–261.
[13] W. Saw, H. McKinnon, I. Gilmour, S.S. Pang, Production of hydrogen-rich syngas from steam gasification of blend of biosolids and wood using a dual fluidized bed gasifier, Fuel 93 (2012) 473–478.
[14] A. Larsson, M. Seemann, D. Neves, H. Thunman, Evaluation of performance of industrial-scale dual fluidized bed gasifiers using the Chalmers 2−4-MWth gasifier, Energy Fuel 27 (2013) 6665–6680.
[15] K. Göransson, U. Söderlind, J. He, W.N. Zhang, Review of syngas production via biomass DFBGs, Renew. Sust. Energ. Rev. 15 (2011) 482–492.
[16] J. Corella, J.M. Toledo, G. Molina, A review on dual fluidized-bed biomass gasifiers, Ind. Eng. Chem. Res. 46 (2007) 6831–6839.
[17] C. Pfeifer, R. Rauch, H. Hofbauer, In-bed catalytic tar reduction in a dual fluidized bed biomass steam gasifier, Ind. Eng. Chem. Res. 43 (2004) 1634–1640.
[18] G.W. Xu, T. Murakami, T. Suda, Y. Matsuzawa, H. Tani, Gasification of coffee grounds in dual fluidized bed: performance evaluation and parametric investigation, Energy Fuel 20 (2006) 2695–2704.
[19] K. Matsuoka, K. Kuramoto, T. Murakami, Y. Suzuki, Steam gasification of Woody biomass in a circulating dual bubbling fluidized bed system, Energy Fuel 22 (2008) 1980–1985.
[20] C. Pfeifer, J.C. Schmid, T. Pröll, H. Hofbauer, Next Generation Biomass Gasifier, 19th European Biomass Conference and Exhibition, Germany, Berlin, 2011.
[21] G.W. Xu, T. Murakami, T. Suda, Y. Matsuzaw, H. Taniet, Two-stage dual fluidized bed gasification: Its conception and application to biomass, Fuel Process. Technol. 90 (2009) 137–144.
[22] K. Göransson, U. Söderlind, T. Henschel, P. Engstrand, W.N. Zhang, Internal tar/CH_4 reforming in a biomass dual fluidised bed gasifier biomass, Conserv. Bioref. 5 (2015) 355–366.
[23] L.G. Wei, S.P. Xu, J.G. Liu, C.H. Liu, S.Q. Liu, Hydrogen production in steam gasification of biomass with CaO as a CO_2 absorbent, Energy Fuel 22 (2008) 1997–2004.
[24] W.J. Zou, C.C. Song, S.P. Xu, C. Lu, Y. Tursun, Biomass gasification in an external circulating countercurrent moving bed gasifier, Fuel 112 (2013) 635–640.
[25] J. Han, H. Kim, The reduction and control technology of tar during biomass gasification/pyrolysis: an overview, Renew. Sust. Energ. Rev. 12 (2008) 397–416.
[26] S. Anis, Z.A. Zainal, Tar reduction in biomass product gas via mechanical, catalytic and thermal methods: a review, Renew. Sust. Energ. Rev. 5 (2011) 2355–2377.
[27] Y.F. Shen, K. Yoshikawa, Recent progresses in catalytic tar elimination during biomass gasification or pyrolysis–a review, Renew. Sust. Energ. Rev. 21 (2013) 371–392.
[28] D. Sutton, B. Kelleher, J.R.H. Ross, Review of literature on catalysts for biomass gasification, Fuel Process. Technol. 73 (2001) 155–173.
[29] Z. Abu El-Rub, E.A. Bramer, G. Brem, Review of catalysts for tar elimination in biomass gasification processes, Ind. Eng. Chem. Res. 43 (2004) 6911–6919.
[30] J. Corella, J.M. Toledo, R. Padilla, Olivine or dolomite as in-bed additive in biomass gasification with air in a fluidized bed: which is better? Energy Fuel 18 (2004) 713–720.
[31] C. Courson, E. Makaga, C. Petit, A. Kiennemann, Development of Ni catalysts for gas production from biomass gasification. Reactivity in steam- and dry-reforming, Catal. Today 63 (2000) 427–437.
[32] C. Pfeifer, R. Rauch, H. Hofbauer, In-bed catalytic tar reduction in a dual fluidized bed biomass steam gasifier, Ind. Eng. Chem. Res. 43 (2004) 1634–1640.
[33] S. Hosokai, M. Sugawa, K. Norinaga, C.Z. Li, J.I. Hayashi, Activity of mesoporous alumina particles for biomass steam reforming in a fluidized-bed reactor and its application to a dual-gas-flow two-stage reactor system, Ind. Eng. Chem. Res. 47 (2008) 5346–5352.
[34] S. Hosokai, J.I. Hayashi, T. Shimadai, Y. Kobayashi, K. Kuramoto, C.Z. Li, T. Chiba, Spontaneous generation of tar decomposition promoter in a biomass steam reformer, Chem. Eng. Res. Des. 83 (2005) 1093–1102.
[35] S. Rapagn, N. Jand, A. Kiennemann, P.U. Foscolo, Steam-gasification of biomass in a fluidized-bed of olivine particles, Biomass Bioenergy 19 (2000) 187–197.
[36] L. Devi, K.J. Ptasinski, F.J.J.G. Janssen, Pretreated olivine as tar removal catalyst for biomass gasifiers: investigation using naphthalene as model biomass tar, Fuel Process. Technol. 86 (2005) 707–730.
[37] J. Marinkovic, H. Thunman, P. Knutsson, M. Seemann, Characteristics of olivine as a bed material in an indirect biomass gasifier, Chem. Eng. J. 279 (2015) 555–566.
[38] F. Kirnbauer, H. Hofbauer, Investigations on bed material changes in a dual fluidized bed steam gasification plant in Güssing, Austria, Energy Fuel 25 (2011) 3793–3798.
[39] Y. Tursun, S.P. Xu, C. Wang, Y.H. Xiao, G.Y. Wang, Steam co-gasification of biomass and coal in decoupled reactors, Fuel Process. Technol. 141 (2016) 61–67.
[40] Y.H. Xiao, S.P. Xu, Y. Tursun, C. Wang, G.Y. Wang, Catalytic steam gasification of lignite for hydrogen-rich gas production in decoupled triple bed reaction system, Fuel 189 (2017) 57–65.
[41] S. Koppatz, C. Pfeifer, H. Hofbauer, Comparison of the performance behaviour of silica sand and olivine in a dual fluidized bed reactor system for steam gasification of biomass at pilot plant scale, Chem. Eng. J. 175 (2011) 468–483.
[42] C. Franco, F. Pinto, I. Gulyurtlu, I. Cabritaet, The study of reactions influencing the biomass steam gasification process, Fuel 82 (2003) 835–842.
[43] J. Corella, J.M. Toledo, G. Molina, Biomass gasification with pure steam in fluidised bed: 12 variables that affect the effectiveness of the biomass gasifier, Int. J. Oil Gas Coal Technol. 1 (2008) 194–207.