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Study on the enhancement of hydrogen generation via biomass gasification in fluidized-bed reactors

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Abstract. In this study, solid biomass is gasified in fluidized-bed reactors, to investigate the effect of various means on syngas composition, especially for enhancing hydrogen content in the production gas. Conventionally, air is supplied to the reactor as gasification medium, which inevitably results in a high nitrogen content in the syngas. Alternatively, steam or oxygen-rich gas can be supplied to improve the syngas characteristics. On the other hand, a so-called “indirect gasification technology” realizes the whole conversion processes in dual reactors, for combustion and gasification, respectively; moreover, solid materials are circulated through two reactors, while gaseous streams in between are separated from each other. Hence, this system features the advantage of producing near nitrogen-free syngas in the gasifier, with air as oxidant in the combustor. Baseline experiments with various operating parameters, including air equivalence ratio (ER) and temperature, were firstly performed in a 30 kW_{th} bubbling fluidized-bed gasifier; then, trial tests were conducted with the aforementioned operational and constructional factors. The preliminary test data show positive trends for the enhancement of hydrogen generation via biomass gasification. Further efforts will be pursued to establish a data base, which would be beneficial to extensive researches on clean energy and carbon abatement technologies.

1 Introduction

With the ambitious goal set by the “Paris Agreement,” which aims at a level even below 2DS (two-degree scenario), a large amount emission of greenhouse gas (GHG), predominantly carbon dioxide (CO_2), caused by anthropogenic impacts has been an environmental issue around the globe for the climate change. However, increased concentrations of CO_2 in the environment are inevitable unless energy systems reduce the carbon emissions to the atmosphere. Decarbonizing the energy systems is a daunting challenge in the 2DS. It is essential to pursue follow-up efforts for energy utilization with

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lower carbon and pollutant emissions, as well as mitigate GHG emissions from the viewpoints of sustainability, which requires various portfolios. Furthermore, biomass can further reduce the carbon dioxide emission, due to the feature of carbon neutral. Hence, clean utilization of biomass via gasification technology is developed in this work.

Gasification technology is a kind of thermo-chemical process for converting carbonaceous feedstock to syngas, mainly consisting of CO, H_2, CO_2, H_2O, CH_4, etc., which can be further utilized in various application processes to meet the multiple energy and resource demands from the industry [1]. Gas generated from gasification process, dominantly CO and H_2, can be further converted to chemical products and liquid fuel after clean-up processes; while some carbon could be fixed in the chemical products or separated in the processes as requested from end applications, of which the feature results in lower CO_2 emission. Syngas could also be delivered to combined-cycle for generating electricity, so that the system efficiency is increased and pollutant emission is decreased.

Fluidized bed is one of the major platforms of biomass gasification. It utilizes gasification-medium (air, steam, CO_2 etc.) through the solid particles and makes them behave like fluid flow. Furthermore, there are some advantages when implementing fluidized bed for gasification, e.g., lower pollution, high efficiency and flexible feedstock [2-4]. Conventionally, air is supplied to the reactor as gasification medium, which inevitably results in high nitrogen content in the syngas. Alternatively, steam or oxygen-rich gas can be supplied to improve the syngas characteristics.

On the other hand, a so-called “indirect gasification technology” realizes the whole conversion processes in dual reactors, for combustion and gasification, respectively; moreover, solid materials are circulated through two reactors, while gaseous streams in between are separated from each other (Fig. 1). Hence, this system features the advantage of producing near nitrogen-free syngas in the gasifier, with air as oxidant in the combustor.

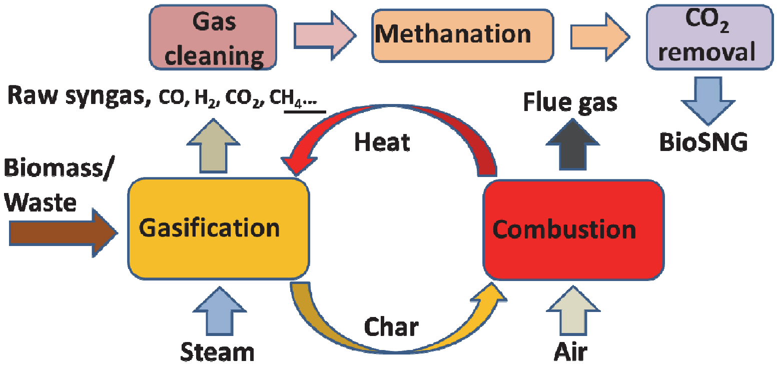

Fig. 1. Conceptual sketch of indirect gasification technology and application.

A comprehensive review on dual fluidized-bed (DFB) biomass gasifiers has been published in the literature [5]. A description of the gasifiers operated today is given, e.g., in Europe (TU Wien and Güssing in Austria [6] and ECN in The Netherlands [7]), and in Japan (IHI Co. [8], EBARA [9]). Biomass gasification with pure steam in a fluidized bed may generate a gasification gas with 60 vol % H_2 (dry basis). Thus, a gasification gas that is very rich in H_2 and with relatively low tar content, for which steam reacts with tar compounds, currently can be obtained, via biomass gasification with pure steam. This concept is also suitable for Bio-SNG production [10].

Interconnected fluidized bed (IFB), regarded as an alternative type of reactor for circulating operation, configures the reactor vessel and associated piping into integrated compartments, which facilitate solid circulation through the beds and enhance the system performance. With higher solid circulating rates and less particle attrition, IFB has been developed as a new application for many particle operations on physical and chemical processes.

Originally, Kunii proposed a conceptual design of a new compact fluidized-bed reactor combined with gasification and combustion processes 1980 [11]. In 1990's, the concept to develop the applications of the interconnected fluidized bed, such as the regenerative desulfurization process [12 - 15], has been realized at Delft University of Technology (DUT) in the Netherlands. In 2004, Wu et al. at Industrial Technology Research Institute (ITRI), Taiwan constructed a 200 kW_{th} interconnected fluidized bed gasification test facility to carry out gasification of refuse derived fuel (RDF) to obtain the performance information for future development [16].

Figure 2 shows the configuration of an interconnected fluidized bed [16]. The IFB system consists of four fluidization compartments, for which two dense-phase beds and two lean-phase beds are alternatively connected by weirs and orifices, respectively. The two lean-phase beds denote gasification zone and combustion zone, respectively. The two dense-phase beds denote the piping and feature to prevent the gas penetration from one lean-phase bed to another one. Hence, the gas in the gasification zone and combustion zone is individually isolated. The solids within the dense-phase bed flow with a lower velocity through the orifice into the lean-phase bed, and then ascend with a higher velocity over the weir into another dense-phase bed.

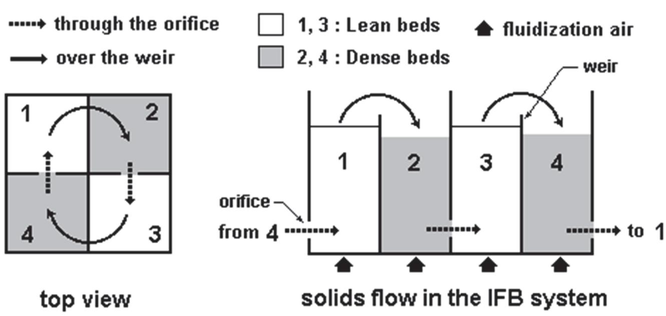

Fig. 2. Configuration of an interconnected fluidized bed.

2 Experimental

In recent years, research on mitigating greenhouse gas emissions and sustainable clean coal technologies has been undertaken at the Institute of Nuclear Energy Research (INER) in Taiwan, together with cooperative research teams at National Chung Hsing University (NCHU). In summary, this work acquires preliminary results for the gasification of Eucalyptus, and further efforts will be pursued to establish a data base for gasification reaction performance and optimal operating parameters, which would be beneficial to extensive researches on clean energy and carbon abatement technologies.

2.1 Feedstock

In this study, there are two types of feedstock used in gasification, i. e., Eucalyptus wood chips and mixed wood pellet. The feedstock size is smaller than 7 mm. Other feedstock properties are shown in Table 1.

Table 1. Feedstock property analysis.

Ultimate analysis (wt. %, daf *)：					
	C	H	O	N	S
Eucalyptus	49.07	4.92	44.94	0.27	0.16
Mixed Wood Pellet	49.52	6.26	43.50	0.72	0.0
Proximate analysis (wt. %, a.r.*)：					
	Moisture		Ash		Combustible
Eucalyptus	11.48		0.56		85.9
Mixed Wood Pellet	9.95		1.02		88.29
Heating value (HHV, MJ/kg, a.r.)					
Eucalyptus	17.34				
Mixed Wood Pellet	17.69				

*daf: dry ash free   a.r.: as received

2.2 Platform

As a start-up effort, a 30 kW_{th} bubbling fluidized-bed (BFB) gasification system has been commissioned; then, the facilities is extended to circulating fluidized-bed (CFB) mode. Later, construction efforts of various dual fluidized-bed (DFB) reactors are in progress.

2.2.1 Solo fluidized bed

Figure 3 shows the 30 kWth bubbling fluidized-bed gasification system, consisting of the feeding system, gasification chamber, air supply system, syngas cleaning system, tar sampling unit and syngas analysis system [17]. This gasifier is made of SUS310 stainless steel with an internal diameter of 6.2 cm in the bed region and total height of 580 cm, enclosed by insulating material of ceramic fibre to shield the heat loss and supplemented by an electric heating system for controlling experimental condition. Silica sand with 2.6 g/cm^3 in density and 505μm in average particle size were used as bed material.

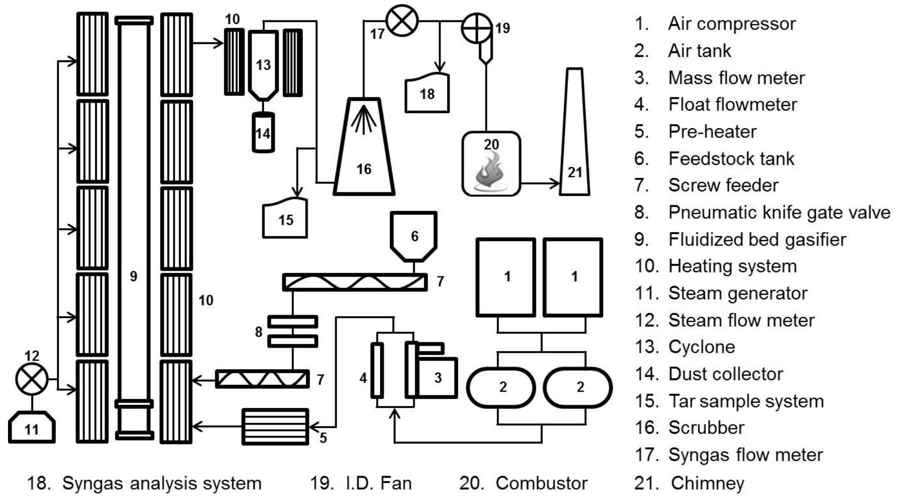

Fig. 3. The 30 kW_{th} bubbling fluidized-bed gasification system.

Syngas analysis system includes online analysis and batch system. The online analysis system is manufactured by Siemens, showing the content of CO, CO_2, O_2, H_2 and CH_4 during the experiment, which displays the timing for sampling the tar and syngas.

2.2.2 Interconnected fluidized beds

Both the cold model and hot model of IFB were commissioned. The former is to investigate hydrodynamic behavior [18], while the latter is for characteristics of thermos-chemical conversion [19]. Figure 4 presents the schematic of a 20 kW_{th} IFB gasification system. Steam generator will be equipped for further study of steam gasification in the future. Two bed materials were adopted in the study: the first one is silica sand, which is for operation in typical gasification; while the other one is ilmenite, which can release oxygen to investigate the effect on syngas compositions..

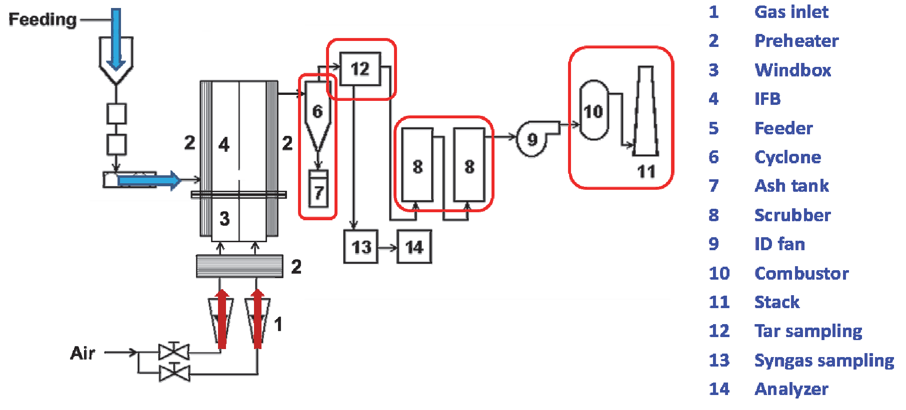

Fig. 4. The 20 kW_{th} IFB gasification system.

2.2.3 Dual fluidized beds

Indirect gasification can achieve nearly nitrogen-free syngas without using air separation unit (ASU) for generating pure oxygen or oxygen-rich stream. At present, a cold flow model was designed by referring to the actual size of the 100 kW_{th} dual fluidized beds, and operating based on dimensionless analysis. The apparatus of the cold model has thirteen pressure gauges which located on the two fluidized-bed reactor as well as the upper and the lower loop seals (Fig 5(a)). The test results were recorded by the instrument equipped. The main body of the cold model was constructed by acrylic material (Fig 5(b)). The characteristics of cold model provide the building blocks for the DFB hot model currently under commissioning at INER.

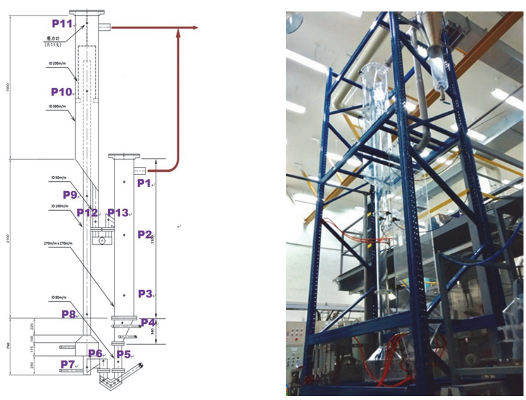

Fig. 5. The cold model of DFB gasification system: (a) Sketch of the cold model and the location of the pressure transmitters (P1~P13), and (b) the photo of the test apparatus.

The physical property of the bed material (olivine) and of the exhaust gas/product syngas in the hot mode reactor are listed in Table 2, at the CRH (combustion reactor) and GRH (gasification reactor) columns, respectively. Farrel's proposed that dimensionless group, e.g., De, Ar, Fl, Gs*, Fr and Rep, were could be calculated using these parameters in combination with the size of the cold model [20]. However, due to two degrees of freedom (i.e., U and Gs), only two dimensionless group values (Fl and Gs*) can be the same as (or close to) the hot model. Hence, the glass beat was selected as bed material in cold model.

Further analysis of the characteristics of the system, the average pressure distribution pattern is similar to that of the published literature [6]. The pressure distribution data are under analysis processes, and will be presented in the further study.

Both IFB and DFB have key features of two independent reaction zones with the gases being isolated. It is easy to adopt steam as gasification medium to generate nitrogen-free syngas. Steam is generally used in industry and is relatively cheaper compared with oxygen as gasification medium. But, steam-blown gasification reactions are endothermic and result in a requirement of an external heat source. The external heat is provided by the bed material that is circulating between the two beds. Adopting IFB to apply in steam gasification is a new concept and is still in the early stage of development. Thus, the hot model of IFB is using air as gasification medium in the first stage, to simplify the experimental procedure. The experimental data are used as the base case to make comparison with those of steam gasification in the further study. On the other hand, DFB

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has been well employed in the steam gasification, and is introduced in the study to make comparison with IFB in the future.

Table 2. The results of dimensionless analysis.

		CR_H	CR_C	GR_H	GR_C
Bed Material		Olivine	Glass	Olivine	Glass
η_G	Pa s	4.50E-05	1.83E-05	3.59E-05	1.83E-05
ρ_G	kg m^{-3}	0.29	1.15	0.20	1.15
U	m s^{-1}	9.20	3.06	0.66	0.19
ρ_P	g cm^{-3}	2.85	2.42	2.85	2.42
d_P	µm	520.00	200.00	520.00	200.00
Φ	-	0.80	0.80	0.80	0.80
D	mm	102.30	102.30	270x270	270x270
G_s	kg m^{-2} s^{-1}	42.24	11.85		
De	ρ_P/ρ_G	9796.98	2095.53	14103.04	2095.53
Ar	dp3*ρg*(ρs-ρg)*g/μ2	564.21	652.06	616.49	652.06
FI	U/u_{mf}	120.45	130.00	6.88	8.00
G_S*	$G_S \rho_p^{-1} U^{-1}$	1.61E-03	1.60E-03		
Fr	$U^2/g\, d_p$	16608.26	4780.76	85.32	18.10
Re_p		30.93	38.58	1.93	2.37
$Ar^{1/3}$		8.26	8.67	8.51	8.67
U*	$Re_p/Ar^{1/3}$	3.74	4.45	0.23	0.27
Q_o	m^3/h	272.22	90.58	173.05	49.44
m_s	kg h^{-1}	1250.00	350.72		

3 Results and Discussion

3.1 Baseline performance characteristics

The baseline gasification experiments were performed in the 30 kW_{th} bubbling fluidized-bed gasification system as shown in Figure 3, to investigate the effect of various operating parameters, including air equivalence ratio (ER) and temperature. The equivalent ratio (ER) is defined as the actual air-fuel ratio to stoichiometric air-fuel ratio. There are three equivalent ratio (ER=0.2, 0.3 and 0.4) and three experimental temperature (700, 800 and 900°C) as the experimental conditions in this study.

The effect of ER on CO and H_2 content in the syngas of Eucalyptus wood chips gasification are presented as contours in Figure 6. It is shown that CO and H_2 increased as ER decreased. As ER increasing, more air is supplied to the gasifier; according to Le Chatelier's Principle, whole gasification would tend to oxidation. Also it enhances the burning of char to generate CO_2 with the compensation of product gas such as H_2, CO and CH_4. As a result, there are more CO_2 while less H_2 and CO when the ER is increased. The trends are all consistent with previous research [21 - 23].

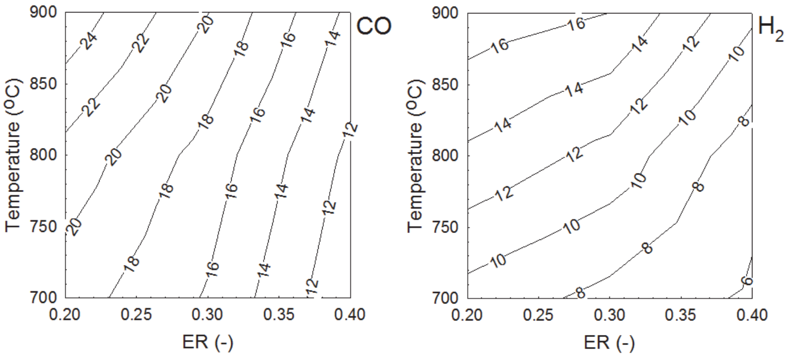

Fig. 6. Contours of CO and H_2 [%] in the syngas of Eucalyptus gasification.

Temperature is another important parameter in gasification. When the temperature ascends, CO and H_2 increase, which is consistent with the results of previous studies [24, 25]. Higher temperature promotes some endothermic reaction, like Boudouard reaction($C+CO_2 \rightarrow 2CO$) and water gas reaction ($C+H_2O \rightarrow CO+H_2$, $C+2H_2O \rightarrow CO_2+2H_2$), to produce more CO and H_2. On the other hand, CO/CO_2 ratio will be higher as temperature increases. When temperature is higher than 830°C, the Boudouard reaction will have more effect than the water gas reaction [26].

3.2 Influence of gasification medium

As mentioned before, air is usually supplied to the reactor as gasification medium for simplicity and cost reasons, which in turn results in high nitrogen content in the syngas. From operational viewpoints, alternative media, e.g., steam or oxygen-rich gas, can be implemented to improve the syngas characteristics.

Two trial tests were conducted to verify the above scenarios. The first case is to address the influence of steam. The reference case in Fig. 7 was carried out under the following operating conditions: feedstock mass flow rate 2 (kg/h), temperature 900°C and ER 0.3. For comparison, an additional stream of steam to fuel ratio (kg/kg) SF = 0.4 was injected to the reactor. Although the steam temperature was substantially lower, due to equipment limitation, than that in the reactor, it is clearly seen that hydrogen generation is enhanced.

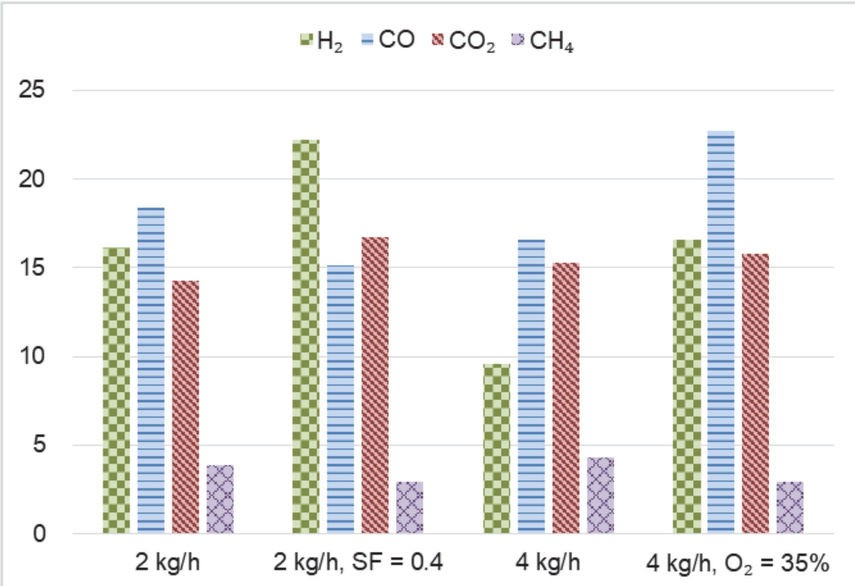

Fig. 7. Comparison of syngas composition with various gasification media.

Oxygen-rich gas is another candidate for enhancing hydrogen generation. Again in Fig. 9, the reference case was carried out under the following operating conditions: feedstock mass flow rate 4 (kg/h), temperature 900°C and ER 0.3. Then, the oxygen content was increased to 35%; as a consequence, hydrogen generation is enhanced as well.

3.3 Effect of indirect gasification

Figure 8 presents the effect of indirect gasification on syngas composition. The experiments were carried out under the following operating conditions: feedstock mass flow rate 2 (kg/h), temperature 900°C and ER 0.2. Since IFB features nearly zero N_2 environment in the gasifier when steam is used as gasification medium, the composition of syngas is presented as N_2-free condition for the cases of air as gasification medium from the current test rig in Fig. 4, to make convenience for comparing with steam counterpart in the further study. For fair comparison, the BFB data were adjusted to the same condition. It is seen that IFB exhibits potential for enhancing hydrogen generation, based on the preliminary trial.

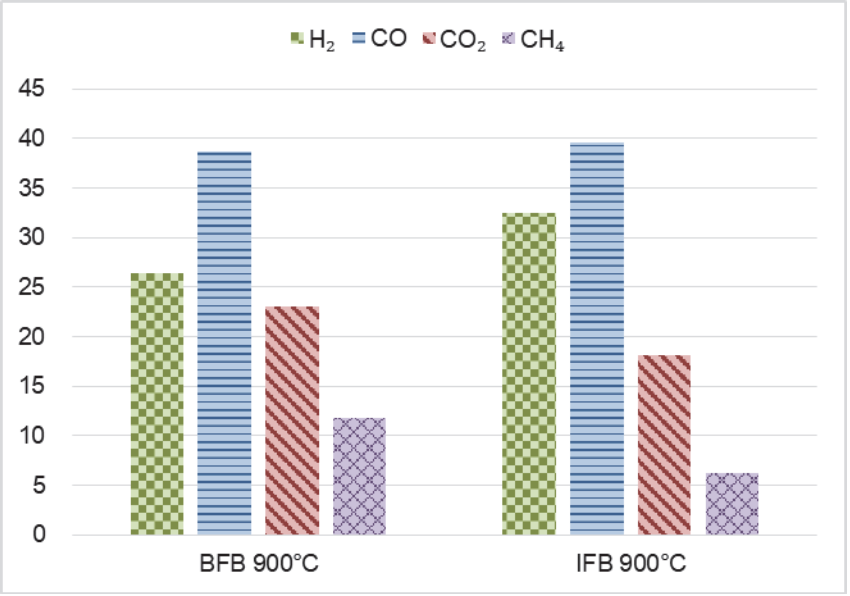

Fig. 8. Effect of indirect gasification on syngas composition (N_2-free): BFB vs. IFB.

3.4 Effect of chemical looping gasification

On the other hand, chemical looping process (CLP) is an enabling technology for advanced CO_2 separation option with great potential, which realizes the looping of solid materials through two reactors, while gaseous streams in between are separated from each other. Thus, the IFB, as a new CLP reactor, can be employed to reduce particle attrition, increase solid circulating rates, and separate CO_2 simultaneously. Hence, oxygen carrier (OC), ilmenite imported from Australia, was utilized to assist the gasification; as shown in Fig. 9, hydrogen generation is enhanced.

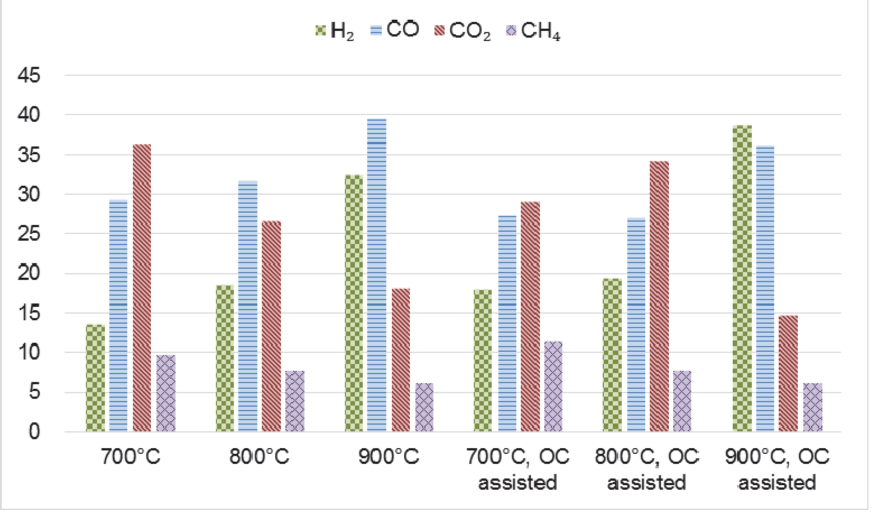

Fig. 9. Effect of chemical looping gasification on syngas composition (N_2-free): IFB.

4 Conclusions

Gasification experiments of biomass were performed in various fluidized-bed reactors to investigate the potential means for the enhancement of hydrogen generation. The preliminary test data show positive trends for the experiments conducted with the aforementioned operational and constructional factors. Further efforts will be pursued to establish a data base for gasification reaction performance and optimal operating parameters. The outcome would be beneficial to extensive researches on clean energy and carbon abatement technologies.

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