



# CaO-based chemical looping gasification of biomass for hydrogen-enriched gas production with in situ CO<sub>2</sub> capture and tar reduction

Jakkapong Udomsirichakorn<sup>a</sup>, Prabir Basu<sup>b</sup>, P. Abdul Salam<sup>a,\*</sup>, Bishnu Acharya<sup>c</sup>

<sup>a</sup> Energy Field of Study, Asian Institute of Technology, P.O. Box 4, Khlong Luang, Pathumthani 12120, Thailand

<sup>b</sup> Mechanical Engineering Department, Dalhousie University, Halifax B3J 1Z1, Canada

<sup>c</sup> Greenfield Research Incorporated, P.O. Box 25018, Halifax B3M 1N8, Canada

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## ABSTRACT

Steam gasification of biomass undergoes the problem of undesirable CO<sub>2</sub> and tar formation. Calcium oxide (CaO), when added to the gasification, could play the dual role of tar reforming catalyst and CO<sub>2</sub> sorbent, and thereby produce more hydrogen. However, the deactivation of CaO after carbonation reaction is challenging for continuous hydrogen production and economical perspective. The concept of CaO-based chemical looping gasification (CaO-CLG) plays a key role in overcoming such a challenge. This work primarily aims at studying steam gasification of biomass with the presence of CaO in a uniquely designed chemical looping gasification (CLG) system for hydrogen production with in situ CO<sub>2</sub> capture and tar reduction. The effect of solid circulation rates on gas and tar production is studied. A comparison of CaO-CLG, sand-based chemical looping gasification (Sand-CLG) and CaO-based bubbling fluidized bed gasification (CaO-BFBG) is presented mainly focusing on gas and tar production. The maximum H<sub>2</sub> and minimum CO<sub>2</sub> concentrations as well as maximum H<sub>2</sub> yields of 78%, 4.98% and 451.11 ml (STP)/g of biomass, respectively, are obtained at the solid circulation rate of 1.04 kg/m<sup>2</sup>s. At the same point, the maximum total gas yield was 578.38 ml (STP)/g of biomass and the tar content of 2.48 g/Nm<sup>3</sup> was the lowest. 30% higher concentration of H<sub>2</sub> and triple yield of H<sub>2</sub> were found in CaO-CLG compared to Sand-CLG. Compared to CaO-BFBG, CaO-CLG resulted in 15% higher concentration of H<sub>2</sub> and almost double yield of H<sub>2</sub>. Moreover, the lowest tar content of 2.48 g/Nm<sup>3</sup> was obtained for CaO-CLG while the tar content was 68.5 g/Nm<sup>3</sup> for Sand-CLG and 26.71 g/Nm<sup>3</sup> for CaO-BFBG. CO<sub>2</sub> concentration obtained for CaO-CLG also significantly reduced by 13–17% as compared to both Sand-CLG and CaO-BFBG.

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## 1. Introduction

The continued use of fossil based energy is increasingly contributing to global warming, climate change and energy security issues. As a consequence, many researches and developments worldwide are realized to encourage the use of renewable and sustainable energy sources. Hydrogen being a pollution-free energy carrier is expected to be the most promising source to replace fossil fuel employed both in power generation and transportation sectors [1–3]. However, it is not naturally available in sufficient quantities and also needs to be synthetically produced. Currently, there are a number of energy sources and technologies to produce hydrogen. But about 96% of the hydrogen for commercial use is produced from fossil fuels, nearly 50% of which is contributed by natural gas, primarily via steam methane reforming [4], which is a fossil fuel based process. Biomass is considered as the potential substitute for

the depleting fossil fuels [5]. It is also accepted as the greatest promise due to its availability everywhere in the world [2].

The technologies available for conversion of biomass into hydrogen-rich gas can be classified into biological and thermo-chemical methods [6–10]. Biomass steam gasification, as one of the thermo-chemical methods, has been perceived as an attractive process for producing syn-gas rich in hydrogen [11–16]. However, the process unavoidably suffers from the problem of unpleasant tar and CO<sub>2</sub> formed within the process. The use of calcium oxide (CaO) dually acknowledged as a tar reforming catalyst and a CO<sub>2</sub> sorbent has currently gained lots of attention due to its cheapness and abundance [17]. Its role in catalytic reforming of tar not only reduces the tar amount in the product gas but also enhances the total gas and hydrogen yields [1,17,18]. Similarly, another role in removing CO<sub>2</sub> from the gasification reaction as soon as it is formed alters the equilibrium composition of the produced gas and promotes the production of gas rich in hydrogen [3,17]. However, the deactivation of CaO after capturing CO<sub>2</sub> seems to be a major obstacle to continuous hydrogen production. Also, if the frequent replacement of CaO sorbent is needed, the process may not be economically attractive [19]. To overcome such

\* Corresponding author. Tel.: +66 2 524 5420; fax: +66 2 524 5439.  
E-mail address: [salam@ait.ac.th](mailto:salam@ait.ac.th) (P. Abdul Salam).

challenges, the concept of CaO-based chemical looping gasification (CaO-CLG), basically aimed for hydrogen production with in situ CO<sub>2</sub> capture, is playing an important role.

The CaO-CLG concept was initiated through the CO<sub>2</sub> acceptor process and has currently been developed through the HyPr-RING process, the ZECA process, the ALSTOM process, and the AGC process [20]. Nevertheless, some of these processes are not considered as environmentally sustainable hydrogen production processes due to their dependence on fossil fuel. Moreover, the processes were designed to operate at high pressures and/or temperatures and some of them were designed with multiple-loop configuration that causes high operational complexity as well as high capital cost. Although these demerits are challenging for the operation of chemical looping process for continuous hydrogen production, no formation of CO<sub>2</sub> and tar observed in some previous processes, such as the HyPr-RING, seems to be advantageous. This shows that each technology has its own pros and cons. To facilitate such challenge of high operational complexity of the past technologies and encourage environment-friendly hydrogen production, recent efforts [21,22] have been devoted to developing the simpler CaO-CLG system with the single-loop and atmospheric operation as well as utilization of biomass and biowaste. A real CaO-CLG system which mainly consists of regenerator, gasifier and loopseal as shown in Fig. 1 was uniquely designed and developed by Acharya et al. [21,22] based on the mature circulating fluidized bed (CFB) technology and was experimentally used in the present work as well. Acharya et al. [21] studied the influence of temperature and in-bed CaO on hydrogen and CO<sub>2</sub> production in a gasifier and also the regeneration of calcium carbonate (CaCO<sub>3</sub>) in a regenerator. Recently, Acharya et al. [22] tested the influence of three different calcination media that are necessary and used for supplying to a regenerator, whereas Udomsirichakorn et al. [17] investigated the influence of temperature and steam-to-biomass ratio (S/B) as well as

in-bed CaO on syngas and tar produced from a gasifier. Moreover, the capability of the CaO sorbent as a bed material undergoing calcination-carbonation cycles is also studied by Acharya et al. [22]. These previous experimental studies have been conducted using the same CaO-CLG system but separately focused on individual component and sub-process of the system. Overall investigation on the whole looping system is still lacking not only in this CaO-CLG system but also in other CaO-CLG systems that are scarcely found in open literature. This provides an opportunity for the present study to fulfill the knowledge gap on the CaO-CLG system which is currently available only in R&D stage [23].

This paper presents an experimental study of catalytic steam gasification of biomass in the CaO-CLG system with the main focus of studying the influence of solid circulation rate on the combined role of CaO as a tar reforming catalyst and CO<sub>2</sub> sorbent to enhance hydrogen production. The comparative study of the production of syngas and tar obtained from CaO-CLG and sand-based chemical looping gasification (Sand-CLG) as well as CaO-based bubbling fluidized bed gasification (CaO-BFBG) is also presented here.

## 2. Experimental

### 2.1. Biomass feedstock

Pine wood sawdust with particle size of 0.425–0.5 mm was used as the feedstock. The proximate analysis of the sawdust was conducted following ASTM standard test methods. The higher heating value (HHV) analysis was performed according to the British Standard No. BS4379 using a bomb calorimeter (Parr 6100). The carbon, hydrogen, nitrogen and sulfur contents (C, H, N and S) in the sawdust were determined using an elemental analyzer (Leco CHN-1000 and Leco SC-432) and

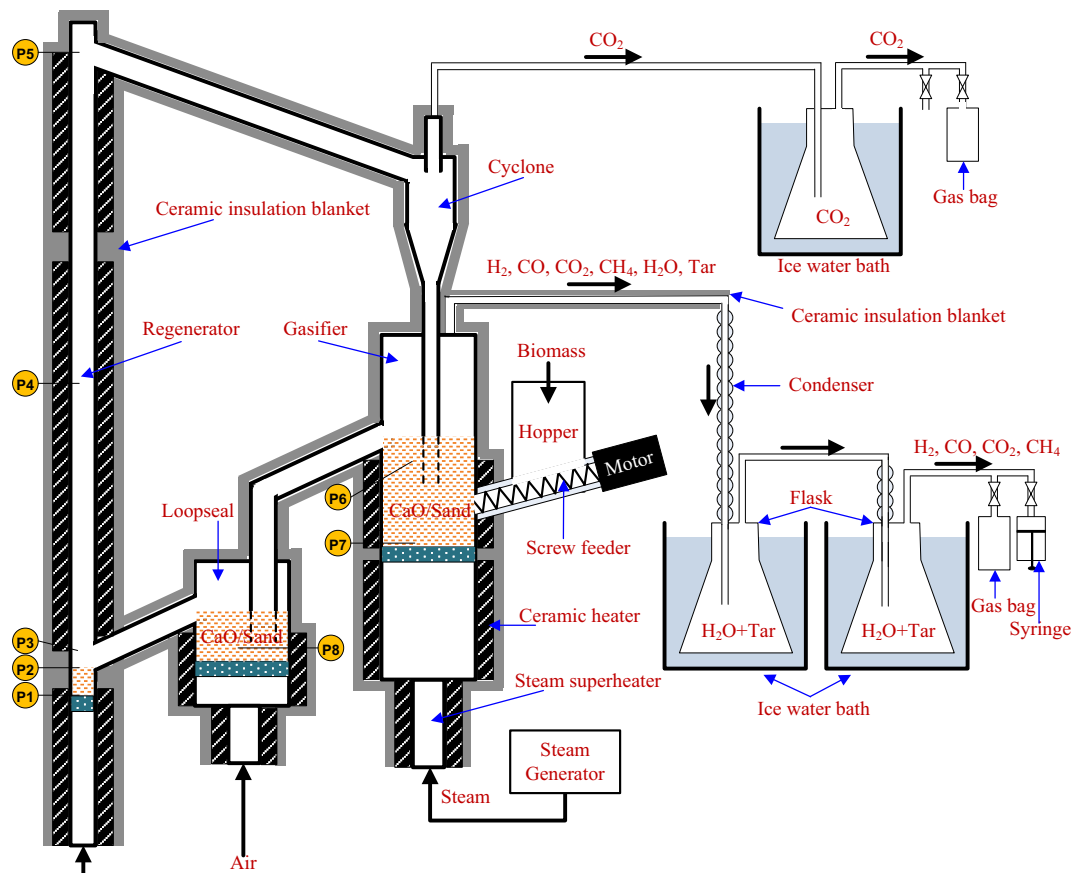


Fig. 1. Experimental setup.

**Table 1**  
Ultimate and proximate analysis of biomass.

Feedstock	Ultimate analysis (wt.%, dry ash-free basis)					HHV (MJ/kg)	Proximate analysis (wt.%, as-received basis)			
	C	H	N	S	O		M	VM	Ash	FC
Pine sawdust	53.28	5.55	0.01	0.003	41.15	18.70	6.16	85.42	0.53	7.89

oxygen (O) was then calculated by difference to 100%. The ultimate and proximate analysis results are presented in Table 1.

## 2.2. Bed materials

Silica sand (mean particle size 0.25–0.3 mm) with apparent density of 2300 kg/m<sup>3</sup> and calcined limestone (mean particle size 0.25–0.3 mm) were used as bed materials for the Sand/CaO–CLG system. Prior to starting each experiment, the sized limestone (apparent density 2600 kg/m<sup>3</sup>) was calcined in a muffle oven at 900 °C for 3 h and then gradually cooled down by purging the atmosphere in the oven with nitrogen gas to avoid a carbonation reaction of calcined limestone. The chemical composition of calcined limestone is presented in Table 2. The change in particle size of limestone after calcination was found to be very small, but its weight loss through calcination was about 42.7%.

## 2.3. Facilities and procedures

Fig. 1 shows the schematic drawing of the chemical looping gasification system developed by Acharya et al. [21,22] and used in this experimental study. The unique tailor-made configuration of this system mainly consists of a bubbling fluidized-bed gasifier with a height of 450 mm, a circulating fluidized-bed regenerator with a height of 1500 mm and a bubbling fluidized-bed loopseal with a height of 135 mm. Electric heaters with a total capacity of 12 kW were mounted alongside these reactors and used to externally heat the system. All experiments were carried out at atmospheric pressure. Before starting each run, about 1.5–2 kg of either fresh CaO (calcined limestone) or sand with a mean particle size of 0.275 mm was manually fed into the bed of the reactors for use as bed material. The biomass feedstock was loaded into the hopper. After loading the gasifier, all the heaters as well as the electric steam generator and steam/air superheater/preheater were turned on. For initial heating up of the system, air was supplied through a 30-μm porous ceramic distributor plate of each reactor to fluidize the bed material. Moreover, the temperature and pressure of all reactors were measured by K-type thermocouples and pressure transducers. It took around 3–4 h to reach a bed temperature of 650 °C in the gasifier, of 900 °C in the regenerator and of 350 °C in the loopseal. Once the bed attained these temperatures, the air flow through the bed of gasifier was replaced with superheated steam supplied from the electric steam generator and superheated by the electric steam superheater installed below the gasifier. But the fluidization in the regenerator and in the loopseal was kept supplying with air but increased to higher velocities.

Table 3 shows the flow rate and superficial velocity of different fluidization media supplied to each reactor based on theoretical calculation and practical considerations for suitable fluidizing regime. Unlike other reactors, the aeration provided to the loopseal for all experiments was sufficient to give a circulation of the solid from one to another reactor as a looping process and this was also varied to allow different desired solid circulation rates. Once the system was stabilized at the desired temperature in a circulating mode of the solid, the biomass feeding was

started from the hopper through a water cooled screw pipe. Thus, biomass nearly at room temperature mechanically moved into the bed at the desired constant feeding rate that was regulated by the variable speed drive. Inside the system, unburned biomass char obtained from gasification leaves the gasifier together with the bed material through an inclined, gasifier-loopseal connecting the pipe towards the loopseal operated in a bubbling mode and then go through an inclined, loopseal-regenerator connecting the pipe towards the riser serving as a regenerator. Inside the riser-regenerator, the heat internally generated from char combustion was transferred to the solid and also used to regenerate calcium carbonate (CaCO<sub>3</sub>) produced in and circulated from the gasifier. The air supplied to the bed in the regenerator causes a fast fluidization so that the solid gets entrained and separated in the cyclone. Once separated, the hot bed material falling through the standpipe will start filling the gasifier and simultaneously providing heat to the gasification process. Despite being heated by the circulating hot solid, the gasification process was still being heated by electric heaters to maintain the bed temperature constant at 650 °C for all experiments. Once the gasification took place steadily, that typically occurred 10–15 min after feeding the biomass, the hot syngas from the gasifier flowing through the sampling line, insulated with ceramic insulation blanket, was condensed in cold traps where the condensed steam and tar as well as solid particles were collected in flasks. The dry and clean gas then passed through the sampling point where a Tedlar gas bag was connected and the sample was taken every 5 min for about 1 h. The gas flow rate was further measured and recorded at some particular moments of the experiment. After the end of the experiment, the gas samples were analyzed in a gas chromatograph (VARIAN Micro GC-490) and the condensate collected in the flasks was taken for tar analysis. The liquid sample of tar, collected after each experimental run, was paper-filtered, homogenized and then used for quantitative analysis of gravimetric tar. The concentration of gravimetric tar was determined by evaporating the homogenized liquid sample in a standard rotary evaporator, according to the Tar Protocol CEN/TS 15439:2006 [24].

## 3. Results and discussion

### 3.1. Hydrodynamics and pressure distribution

Fig. 2 shows the average pressure distribution in the present CLG system to verify the circulating fluidization regime in this CFB based looping system. This is similar to what one would expect in a CFB system [22,25]. The average voidage in the bubbling fluidized-bed gasifier was found to be 0.61. The average voidage in the turbulent bed section of the riser-regenerator was 0.82, which increases to 0.96 around its exit. The pressure balance around the loop shows that the pressure inside the gasifier is much higher than that at the top of the loopseal so that no air going into the loopseal could flow into the gasifier. This was also verified by the negligible amount of air composition detected in the product gas measured at the outlet of the gasifier. Moreover, the pressure at the base of the loopseal is much higher than that in the regenerator so that no gas can escape the regenerator through the loopseal.

Fig. 3 presents the average suspension density profile in the riser-regenerator calculated from the pressure measured along its height. The nature of this suspension profile is similar to the one observed in CFB boilers [25]. These two characteristics of a nature of a CFB regime found in this study can confirm that the system was operated in suitable CFB-based looping process.

**Table 2**  
Chemical composition of calcined limestone in wt.%.

CaO	MgO	SiO <sub>2</sub>	Fe <sub>2</sub> O <sub>3</sub>	Al <sub>2</sub> O <sub>3</sub>	CaCO <sub>3</sub>
95.5	0.9	1.3	0.2	0.4	1.1

**Table 3**  
Flow streams in different fluidizing reactors.

Reactor	Fluidizing medium	Fluidization regime	Total flow rate (Nm <sup>3</sup> /s)	Fluidizing velocity (m/s)
Gasifier	Steam	Bubbling bed	$7.40 \times 10^{-4}$	0.09
Loopseal				
at $G_s^a = 0.91 \text{ kg/m}^2\text{s}$	Air	Bubbling bed	$2.33 \times 10^{-4}$	0.029
at $G_s = 1.04 \text{ kg/m}^2\text{s}$	Air	Bubbling bed	$2.70 \times 10^{-4}$	0.033
at $G_s = 1.14 \text{ kg/m}^2\text{s}$	Air	Bubbling bed	$3.25 \times 10^{-4}$	0.04
Regenerator <sup>b</sup>				
at $G_s = 0.91 \text{ kg/m}^2\text{s}$	Air	Fast bed	$1.10 \times 10^{-3}$	2.17
at $G_s = 1.04 \text{ kg/m}^2\text{s}$	Air	Fast bed	$1.17 \times 10^{-3}$	2.30
at $G_s = 1.14 \text{ kg/m}^2\text{s}$	Air	Fast bed	$1.27 \times 10^{-3}$	2.51

<sup>a</sup>  $G_s$  = Solid circulation rate.

<sup>b</sup> Total flow rate of air to regenerator = Fixed flow rate of air to regenerator + Varied flow rate of air to loopseal.

### 3.2. Influences of solid circulation rate on production of syngas and tar

Fig. 4 presents the influence of solid circulation rate on product gas composition and H<sub>2</sub> yield obtained at identical operating conditions (S/B = 3.41 and gasification temperature = 650 °C). Fig. 5 shows its effect on the tar and product gas yields. Experimental results (Fig. 4) show the concentration and yield of H<sub>2</sub> increase with the increment of circulation rate from 0.91 to 1.04 kg/m<sup>2</sup>s and then decrease with further increase of circulation rate from 1.04 to 1.14 kg/m<sup>2</sup>s. The similar trend is found for the total gas yield but the opposite is observed for the tar yield (Fig. 5).

Fig. 4 shows that as the solid circulation rate increased from 0.91 to 1.04 kg/m<sup>2</sup>s the volumetric concentration of H<sub>2</sub> sharply increased from 71.93% to 78% and its yield also increased over 40 ml/g of biomass. The main reason for this phenomenon is probably attributed to the influence of the circulation rate on the quantity of CaO derived from regenerating CaCO<sub>3</sub> via a calcination reaction (Eq. (1)) in the regenerator and then circulated to the gasifier, in which CO<sub>2</sub> and tar are produced. This can be also implied that a higher circulation rate allows a higher amount of regenerated CaO in the gasifier. As a consequence, more sites are available for higher CO<sub>2</sub> absorption by CaO via a carbonation reaction (Eq. (2)). This conjecture was also reported by Udomsirichakorn et al. [17] and could be supported by the decrease in CO<sub>2</sub> concentration from 8.07% to 4.98% observed for the present study. The higher absorption of CO<sub>2</sub> consequently lowers CO<sub>2</sub> partial pressure and then enhances

water–gas shift reaction (Eq. (3)) to move in forward direction to produce more H<sub>2</sub> [9,26,27]. This promotion of water–gas shift reaction (Eq. (3)) could be also evidenced by the observed decrease of CO concentration from 9.04% to 5.93%, which is consistent with the increase of H<sub>2</sub> concentration.

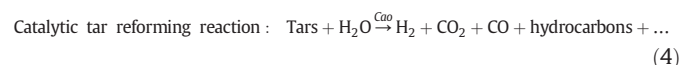
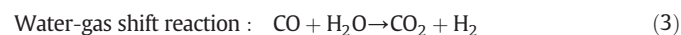
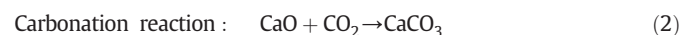


Fig. 5 shows a decrease of tar content from 2.95 to 2.48 g/Nm<sup>3</sup> with an increase of circulation rate from 0.91 to 1.04 kg/m<sup>2</sup>s. The similar trend was reported by Pfeifer et al. [28]. Moreover, the present observation of tar amount can also support the above conjecture of more availability of CaO in the gasifier due to higher circulation rate of solid. As a result of more CaO in the gasifier, the reduction of tar content can be attributed to the enhancement of catalytic reforming of tar with CaO

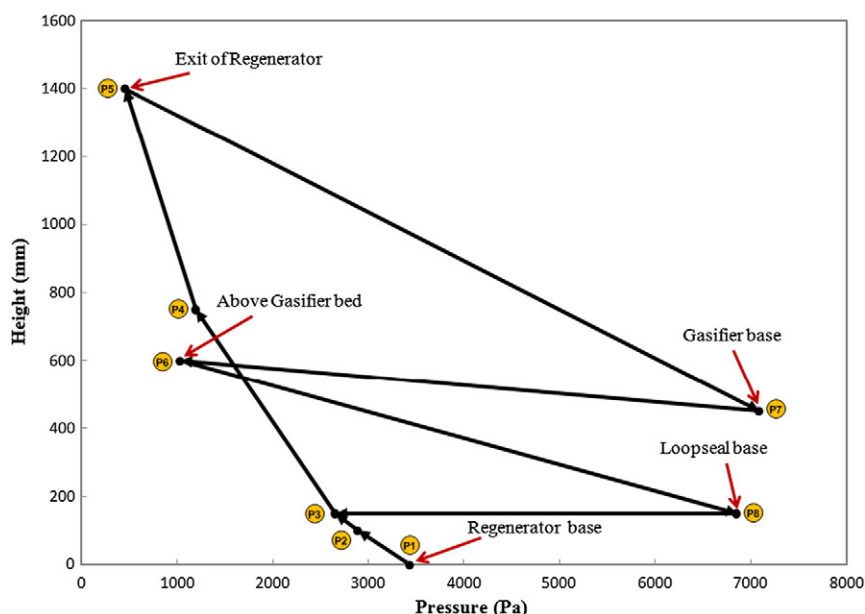


Fig. 2. Pressure distribution in the CaO-CLG system.



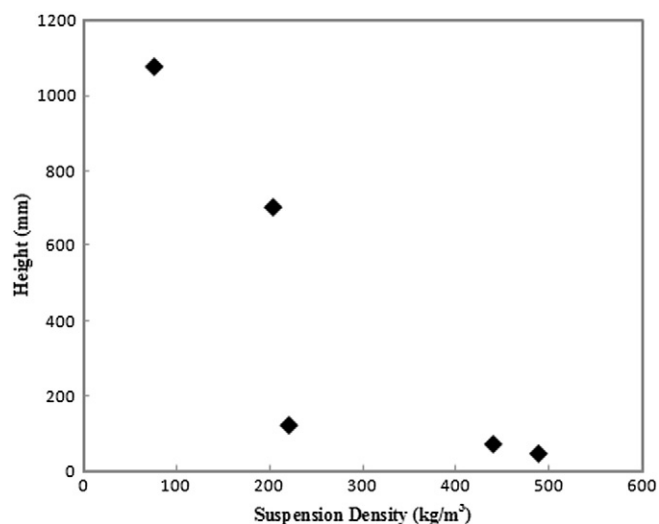


Fig. 3. Suspension density profile along the height of the riser-regenerator.

(Eq. (4)) resulting in the consistent increase of the yields of  $H_2$  (Fig. 4) and total gas (Fig. 5) as reaction products. This also shows that CaO can simultaneously play another role in reforming tar into gas apart from  $CO_2$  absorption [9,17,26,27,29]. In-depth analysis of this is well discussed elsewhere [17].

However for further increase of circulation rate beyond  $1.04 \text{ kg/m}^2\text{s}$ , a sudden decrease in  $H_2$  concentration to 73.08% with consistent increase in  $CO_2$  concentration to 10.06% was noted (Fig. 4). This could be attributed to a short residence time of  $CaCO_3$  in the regenerator along with low calcination level of it, which consequently results in less amount of reactivated CaO in the gasifier. This speculation is also supported by the increase of tar content that is consistent with the decrease of total gas yield within the range of circulation rate of  $1.04\text{--}1.14 \text{ kg/m}^2\text{s}$  (Fig. 5).

Overall, it can be concluded that even though a higher circulation rate of bed material allows positive effect on production of  $H_2$ ,  $CO_2$  and tar, too high circulation rate can adversely affect the calcination process of  $CaCO_3$ , which consequently results in negative production of syngas and tar. However, due to unavailability of this information in CaO-CLG system, more studies are encouraged to validate the present results.

### 3.3. Influences of CaO-CLG on production of syngas and tar

The main focus of this experimental study is steam gasification of biomass in the CaO-CLG system. To point out its superiority in terms of gas and tar production, two baseline studies such as Sand-CLG as well as CaO-BFBG were conducted for comparison. The experimental results obtained for different gasification systems but at identical operating conditions ( $S/B = 3.41$  and gasification temperature =  $650^\circ\text{C}$  for

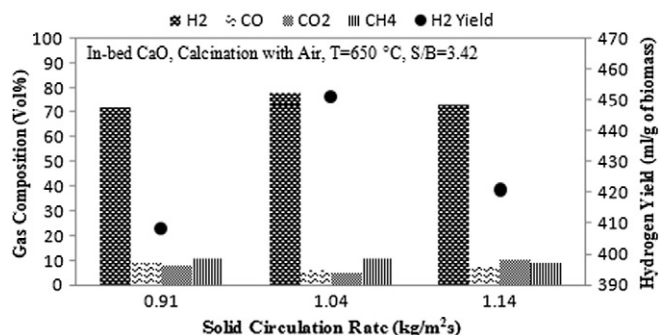


Fig. 4. Effect of solid circulation rate on gas composition and hydrogen.

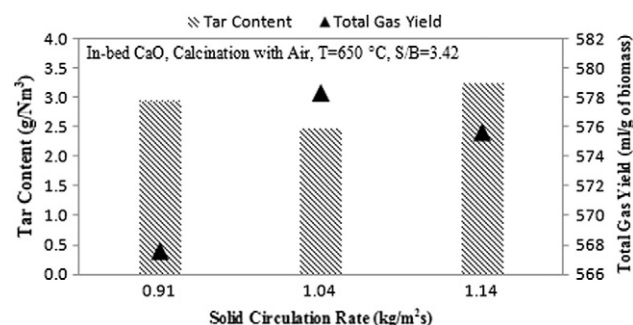


Fig. 5. Effect of solid circulation rate on yields of tar and gas.

all the systems, and solid circulation rate =  $1.04 \text{ kg/m}^2\text{s}$  for both CLG systems) are compared in Fig. 6 for gas composition and  $H_2$  yield and in Fig. 7 for tar and total gas yields. It is evidenced that by using CaO-CLG, one can produce the product gas with the highest concentration and yield of  $H_2$  and the highest yield of total gas as well as lowest contents of  $CO_2$  and tar.

As the influence of CaO is superior to sand as a bed material, using CaO-CLG can increase  $H_2$  concentration from 48.07% obtained for Sand-CLG to 78% and reduce  $CO_2$  concentration from 21.16% obtained for the same baseline system to almost zero (Fig. 6). The plausible reason of this CaO-based phenomenon could be explained by the correlation between the partial pressure reduction of  $CO_2$  due to carbonation reaction (Eq. (2)) and the enhancement of water-gas shift reaction (Eq. (3)). More details of this are previously described in Section 3.2. In addition, the significant decrease of tar content consistent with the increase of total gas yield was also observed in CaO-CLG as compared to Sand-CLG (Fig. 7). This finding can be explained with catalytic tar reforming reaction of CaO (Eq. (4)). As observed in the present study, similar results of the influence of CaO on the production of  $H_2$ ,  $CO_2$  and tar were also reported by many authors [9,29–31] but those results were obtained from different gasification systems. Thus, to verify the superiority of CLG system, the following discussion comparatively presents the results obtained for CaO-CLG and CaO-BFBG, both of which were identically operated with CaO as a bed material.

Compared to CaO-BFBG, the CaO-CLG system is better with higher concentration and yields of  $H_2$  and lower contents of  $CO_2$  and tar (Figs. 6 and 7). This may be because of more availability of CaO in the gasifier of CaO-CLG. In CaO-CLG, the gasifier is continuously filled with regenerated CaO that is obtained from calcination of deactivated CaO (i.e.,  $CaCO_3$ ) in the regenerator, unlike CaO-BFBG in which deactivated CaO cannot be regenerated. This can contribute to the findings above.

## 4. Conclusions

This work investigated steam gasification of biomass with the presence of CaO in a uniquely designed CLG system for hydrogen production with in situ  $CO_2$  capture and tar reduction. The effect of solid circulation rates on production of gas and tar was examined. Comparative

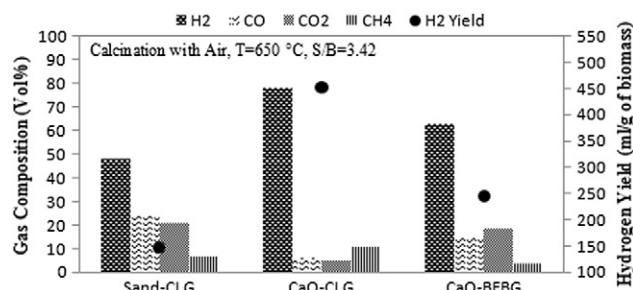


Fig. 6. Effect of CaO-CLG on gas composition and hydrogen.

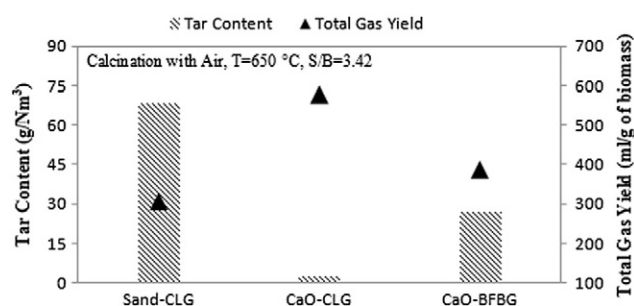


Fig. 7. Effect of CaO-CLG on yields of tar and gas.

study on gas and tar production from CaO-CLG, Sand-CLG and CaO-BFBG was also conducted. Important conclusions drawn from the present study are as follows:

- By increasing the solid circulation rate from 0.91 to 1.14 kg/m<sup>2</sup>s, the concentration and yield of H<sub>2</sub> obtained increased to 78% and 451.11 ml/g of biomass respectively at the circulation rate of 1.04 kg/m<sup>2</sup>s. Also at this point, the lowest CO<sub>2</sub> concentration of 4.98% and the lowest tar content of 2.48 g/Nm<sup>3</sup> were obtained. Higher circulation rate of solid can enhance hydrogen production with simultaneous CO<sub>2</sub> and tar reduction due to more sites available for carbonation reaction and catalytic tar reforming reaction between intermediate products (CO<sub>2</sub> and tar) and regenerated CaO. However, too high circulation rate can cause inverse results due to a short residence time of CaCO<sub>3</sub> undergoing calcination reaction in the regenerator. This study suggests the solid circulation rate of 1.04 kg/m<sup>2</sup>s to be optimal for the system.
- A comparative study of three different gasification systems shows that H<sub>2</sub> concentration and yield were highest in CaO-CLG, while the CO<sub>2</sub> concentration and tar content were lowest. Compared to Sand-CLG, CaO-CLG gave 30% higher concentration of H<sub>2</sub> and triple yield of H<sub>2</sub>. Once compared to CaO-BFBG, it gave 15% higher concentration of H<sub>2</sub> and almost double yield of H<sub>2</sub>. Besides, tar content obtained for CaO-CLG significantly reduced from 68.5 g/Nm<sup>3</sup> for Sand-CLG and 26.71 g/Nm<sup>3</sup> for CaO-BFBG to 2.48 g/Nm<sup>3</sup>, while CO<sub>2</sub> concentration obtained for CaO-CLG also drastically reduced to nearly 0%. This study can indicate that the CaO-CLG system is a promising technology for hydrogen production with in situ CO<sub>2</sub> and tar reduction.

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