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CaO-based chemical looping gasification of biomass for hydrogen-enriched gas production with in situ CO_2 capture and tar reduction

Jakkapong Udomsirichakorn [a], Prabir Basu [b], P. Abdul Salam [a,*], Bishnu Acharya [c]

[a] Energy Field of Study, Asian Institute of Technology, P.O. Box 4, Khlong Luang, Pathumthani 12120, Thailand
[b] Mechanical Engineering Department, Dalhousie University, Halifax B3J 1Z1, Canada
[c] 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_2 and tar formation. Calcium oxide (CaO), when added to the gasification, could play the dual role of tar reforming catalyst and CO_2 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_2 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_2 and minimum CO_2 concentrations as well as maximum H_2 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^{2}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^3 was the lowest. 30% higher concentration of H_2 and triple yield of H_2 were found in CaO-CLG compared to Sand-CLG. Compared to CaO-BFBG, CaO-CLG resulted in 15% higher concentration of H_2 and almost double yield of H_2. Moreover, the lowest tar content of 2.48 g/Nm^3 was obtained for CaO-CLG while the tar content was 68.5 g/Nm^3 for Sand-CLG and 26.71 g/Nm^3 for CaO-BFBG. CO_2 concentration obtained for CaO-CLG also significantly reduced by 13–17% as compared to both Sand-CLG and CaO-BFBG.

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 syngas rich in hydrogen [11–16]. However, the process unavoidably suffers from the problem of unpleasant tar and CO_2 formed within the process. The use of calcium oxide (CaO) dually acknowledged as a tar reforming catalyst and a CO_2 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_2 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_2 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

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

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challenges, the concept of CaO-based chemical looping gasification (CaO-CLG), basically aimed for hydrogen production with in situ CO_2 capture, is playing an important role.

The CaO-CLG concept was initiated through the CO_2 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_2 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_2 production in a gasifier and also the regeneration of calcium carbonate ($CaCO_3$) 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_2 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

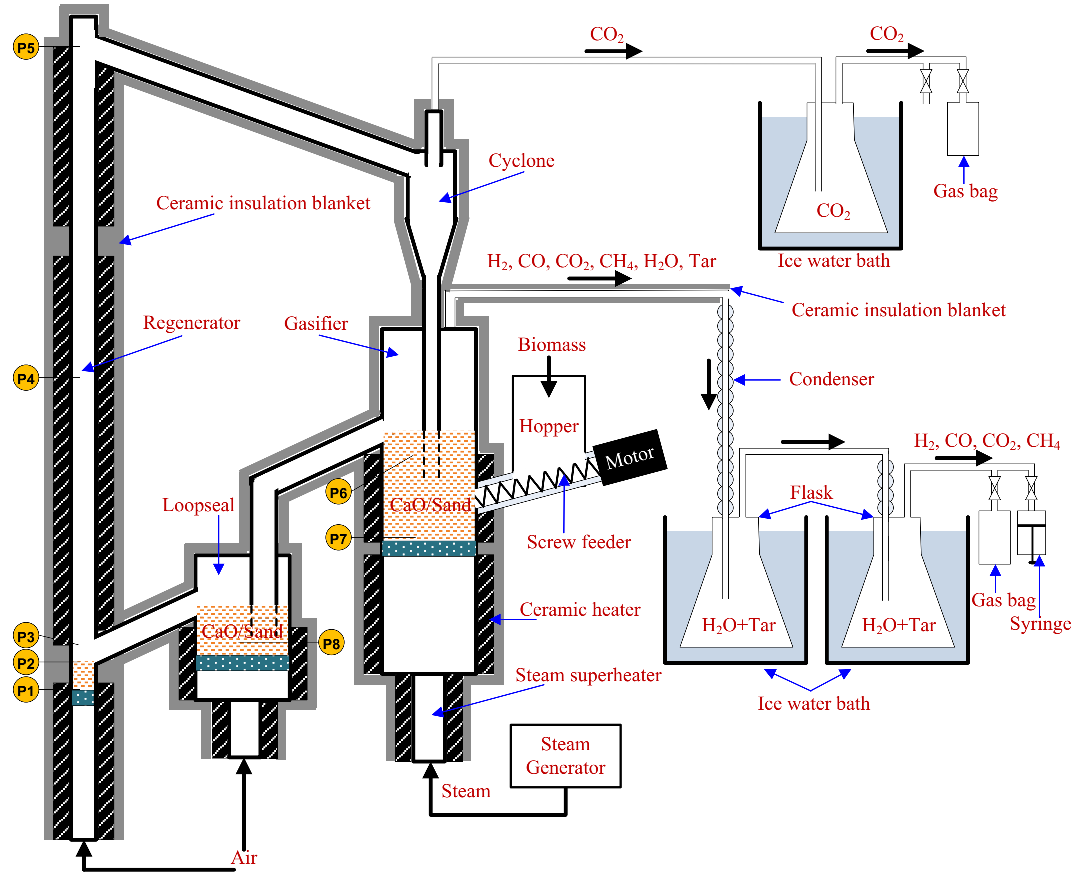

Fig. 1. Experimental setup.