



# Performance evaluation of sorption enhanced chemical-looping reforming for hydrogen production from biomass with modification of catalyst and sorbent regeneration



Trirat Udomchoke<sup>a</sup>, Suwimol Wongsakulphasatch<sup>b</sup>, Worapon Kiatkittipong<sup>b</sup>, Amornchai Arpornwichanop<sup>c</sup>, Watcharapong Khaodee<sup>a</sup>, Jonathan Powell<sup>d</sup>, Jinlong Gong<sup>e</sup>, Suttichai Assabumrungrat<sup>a,f,\*</sup>

<sup>a</sup> Center of Excellence in Catalysis and Catalytic Reaction Engineering, Department of Chemical Engineering, Faculty of Engineering, Chulalongkorn University, Bangkok 10330, Thailand

<sup>b</sup> Department of Chemical Engineering, Faculty of Engineering and Industrial Technology, Silpakorn University, Nakhon Pathom 73000, Thailand

<sup>c</sup> Computational Process Engineering Research Unit, Department of Chemical Engineering, Faculty of Engineering, Chulalongkorn University, Bangkok 10330, Thailand

<sup>d</sup> School of Chemical Engineering and Advanced Materials, Newcastle University, UK

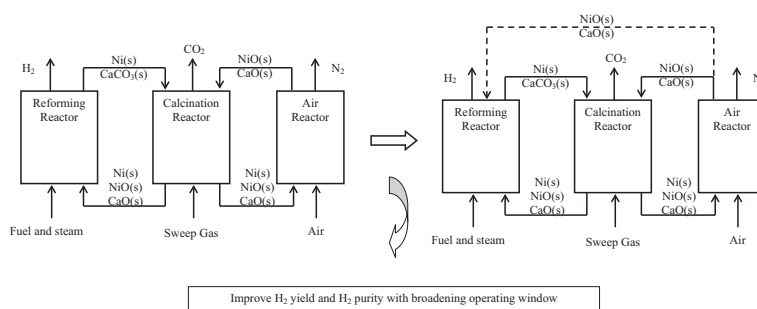
<sup>e</sup> Key Laboratory for Green Chemical Technology of Ministry of Education, School of Chemical Engineering and Technology, Tianjin University, Collaborative Innovation Center of Chemical Science and Engineering, Tianjin 300072, China

<sup>f</sup> PTT Group Frontier Research Center, PTT Public Company Limited, Bangkok 10900, Thailand

## HIGHLIGHTS

- Sorption enhanced chemical-looping (SE-CLR) for hydrogen production was modified.
- Corn stover was used to convert into bio-oil by pyrolysis prior to SE-CLR reforming.
- Splitting a fraction of NiO & CaO to feed directly back to reformer was examined.
- Fractioning solid streams flow directly to reformer can improve H<sub>2</sub> yield and purity.
- Splitting solid circulation streams can broaden process operating window.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Process simulation of sorption enhanced chemical-looping reforming for hydrogen production from biomass was investigated. Corn stover was converted to bio-oil via pyrolysis prior to sorption enhanced chemical-looping reforming (SE-CLR). NiO was used as a catalyst and oxidizing agent and CaO was used as a CO<sub>2</sub> sorbent which are cycled between the reduction and oxidation reactors. Modification of the process to incorporate a step for catalyst and sorbent regeneration was examined and compared with conventional sorption enhanced chemical-looping reforming process. By modifying the SE-CLR process with the recirculating of solids from the air reactor directly to the reformer, a maximum bio-oil conversion rate of 92%, a maximum hydrogen production yield of 153.4 g H<sub>2</sub>/kg corn stover, and a maximum hydrogen purity of 77% v/v can be obtained with a circulated NiO to bio-oil molar ratio (CNB) of 12.3; a circulated CaO to bio-oil molar ratio (CCB) of 125.2; and where the ratio of solids recovered by the air reactor from the calcination reactor ( $\alpha$ ) was set as 1, and the ratio of solids flowing directly from the air reactor to the

\* Corresponding author at: Center of Excellence in Catalysis and Catalytic Reaction Engineering, Department of Chemical Engineering, Faculty of Engineering, Chulalongkorn University, Bangkok 10330, Thailand.

E-mail address: [suttichai.a@chula.ac.th](mailto:suttichai.a@chula.ac.th) (S. Assabumrungrat).

reformer ( $\beta$ ) was set as 0.02. The split of a fraction of solids streams to feed directly back to the reformer offers a great impact process control as operating conditions can be broadened.

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## 1. Introduction

Fossil fuels have been widely used as a source of energy since the industrial evolution. However, as a result of increasingly high global demand and their non-renewable nature, it is becoming increasingly important to substitute fossil fuels with alternatives. Moreover, the use of petroleum resources also results in the emissions of environmental pollutants including greenhouse gases and acidification gases such as carbon dioxide ( $\text{CO}_2$ ), carbon monoxide (CO), sulfur oxide ( $\text{SO}_x$ ), etc. As a result of these concerns, alternative energy carriers have been considered and hydrogen is the one that draws much attention. Hydrogen can be produced from a variety of feedstocks such as natural gas [1], fuel oil [2], or biomass [3], electrolysis of water. Among these, biomass is emerged as a remarkable source for hydrogen production since it is renewable and considered as a relatively green process because biomasses consume atmospheric  $\text{CO}_2$  during growth which results in a smaller net  $\text{CO}_2$  impact compared to fossil fuels [4]. For typical hydrogen production from biomass, the process begins by pyrolysis of biomass into bio-oil and followed by the autothermal reforming (ATR) reaction, the process whereby steam reforming and partial oxidation occur simultaneously. The obtained gaseous products are sent to the water-gas shift reactor and then separation unit to produce high purity hydrogen gas [5]. The process of pyrolysis followed by autothermal steam reforming offers higher potential in both the production of hydrogen yield and energy ratio (heating value of hydrogen with respect to the biomass feed) when compared to those obtained from gasification followed by water-gas shift process [5]. However, limitations for this typical reforming process, are a result of the necessary oxygen separation unit, used to obtain the high oxygen purity gas, required for the partial oxidation process. These units have high operating costs due to high energy demands [6]. For these reasons, opportunities still exist for improving the sustainability and decreasing the cost of producing hydrogen gas.

A hydrogen production technique which has recently received a lot of interest is the technique called sorption enhanced chemical-looping method, which combines the sorption enhanced hydrogen production method with chemical-looping. An integration of these two techniques can potentially improve both hydrogen production and energy consumption as higher purity hydrogen and higher production yields can be obtained without the need of an air or hydrogen gas separation unit [7]. Hydrogen production via sorption enhanced reforming is accomplished by including the adsorption of  $\text{CO}_2$  in the reforming unit in order to shift reaction forward to hydrogen production, hence high purity of hydrogen could be obtained. Adsorbents used in this process to simultaneously capture  $\text{CO}_2$  from product stream could be calcium oxide (CaO), dolomite (CaO/MgO), or hydrotalcite (HTC) [8]. NiO is usually used as oxygen transfer material, which in its reduced form also performs as catalyst to accelerate the reforming reaction. The reaction between the metal oxide and fuel occurs in the reduction reactor, where metal oxide is converted to metal. After that, the metal and air are fed into the oxidation reactor where the metal is converted back to the metal oxide before being fed back to the reduction reactor. As a result of this technique, an expensive and energy intensive gas purification unit is not required for either the separation of hydrogen from nitrogen or for separation of oxygen from nitrogen.

Pimenidou et al. [9] investigated hydrogen production via sorption-enhanced chemical looping reforming of waste cooking oil in a packed bed reactor which composed of dolomite as  $\text{CO}_2$  sorbent and NiO as oxygen carrier. High purity hydrogen (>95%) could be achieved at 600 °C and 1 atm with steam to carbon ratio of 4 without obvious deterioration in fuel and steam conversion up to 6 cycles. Rydén and Ramos [7] investigated sorption enhanced chemical-looping reforming for hydrogen production from methane with the use of NiO as oxygen carrier and CaO as  $\text{CO}_2$  adsorbent. Their main process involved the use of 3 fluidized bed reactors: a reforming reactor for partially oxidizing the methane with oxygen, a calcination reactor for regeneration of the adsorption solids, and the oxidation reactor for oxidation of the oxygen carrier. Thermodynamic results show that more than 2.8 mol of  $\text{H}_2$  can be produced with purity >98 vol% for each mole of  $\text{CH}_4$  fed at a temperature of 600 °C and a pressure 1 bar.

In this work, we investigated the production of  $\text{H}_2$  from corn stover via sorption enhanced chemical-looping (SE-CLR) using Aspen Plus software with the aim to maximize the bio-oil conversion, hydrogen yield and hydrogen purity and minimize energy consumption. The possible operation of a reforming process without an oxygen separation unit and with fewer preheating and cooling units is the subject of investigation and is achieved by varying the amount of solids circulated directly between the oxidation and reduction reactors.

## 2. Simulation

Fig. 1 shows the block flow diagram of a sorption enhanced chemical-looping process (SE-CLR; without dashed line) taken from Rydén et al. [7] and the modification to the SE-CLR process, indicated by the dashed line, which provides a direct supply of NiO and CaO from the oxidation reactor to the reduction reactor, thereby providing more heat for the reduction reaction.

A schematic diagram of our modified sorption enhanced chemical-looping (SE-CLR) process is shown in Fig. 2 and the

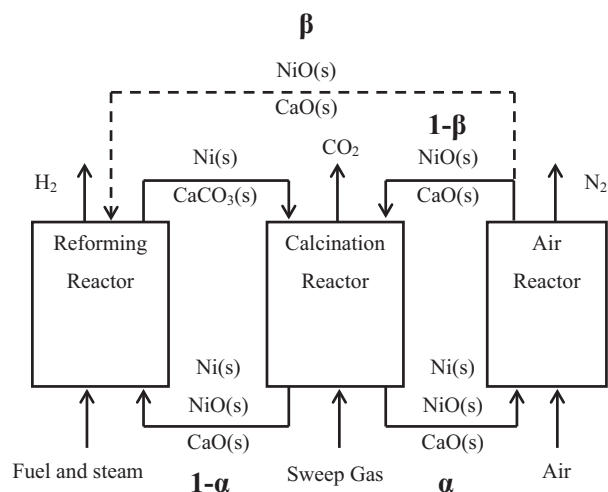


Fig. 1. Block flow diagram of sorption enhanced chemical-looping for hydrogen production.

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