



Full Length Article

High-purity hydrogen production with in situ CO₂ capture based on biomass gasification



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ABSTRACT

Tar formation and CO₂ emission represent the strongest barrier for use of gasification technology for biomass conversion, whereas sufficing for both is only possible with expensive physical methods and further chemical processing. The use of CaO as a CO₂ sorbent within an advanced high-temperature gasification system is able to achieve efficient cracking of the tars to the primary syngas with low emissions. The present work aims to propose a semi-kinetic model on the basis of an Aspen Plus model to describe specific catalytic behavior of calcium oxide on the gasification of rice husk. There has also been an attempt to validate the developed model by means of an experimental study and explore the influence of minerals within ash on gasification characteristics, since kinetic data are scarce in the literature. Effects of some critical parameters such as gasification temperature, equivalence ratio (ER), and steam/biomass ratio (S/B) on hydrogen yield and CO₂ absorption ratio have been studied. Results showed that CO₂ absorption ratio ascends as CaO loading ratio increases, then to continuously decrease due to a significant reduction in endothermic nature of the Boudouard reaction. When ER ascended from 0.15 to 0.25, syngas yield and hydrogen yield did the same thing, increasing from 2.1 to 2.45 Nm³/kg biomass and 37–41 g/kg biomass respectively. In the second stage, ER undertook a rise from 0.25 to 0.3 where gas caloric value and hydrogen yield were decreased.

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1. Introduction

There is an increasing interest in biomass energy as a result of its low cost and high potentials for the production of bio-oil and other chemicals. In addition, due to the environmental considerations as well as the decreasing of fossil fuels reserves, more attention has been paid to utilize the biomass feedstocks for energy production [1]. Among various ways of bioenergy use (such as gasification, combustion and devolatilization), gasification is more economical in the long term [2]. Biomass gasification offers a wide range of applications for synthesis (syngas) and hydrogen production which can be used as a feedstock when producing chemicals such as methanol and other useful products [1,2]. The produced syngas can also be used as a fuel to generate power and electricity in various power generation systems like boilers, gas turbines, and Proton Exchange Membrane (PEM) fuel cells.

One of the most important uses of biomass gasification is the production of high-purity hydrogen. Several pathways have been suggested to researchers in order to produce hydrogen gas, some of which include water electrolysis, steam gasification, biological methods, and nuclear production of hydrogen. However, due to safety issues, economic aspects, and low environmental impacts, other methods are not as popular as biomass gasification [3]. The use of biomass as a raw material to produce hydrogen is not only pro higher hydrogen-rich gas production but also more economical than other fuels such as coal, crude oil, and municipal solid wastes [2]. To date, the potential of biomass gasification for hydrogen production in Iran has attracted much attention, owing to its availability throughout the year, especially in the form of agricultural residues such as rice husk. The largest producer of rice in Middle East, Iran achieves a production rate of more than 2.0 million tons annually [4].

Several experimental and modeling studies in the scientific literature [5–8] have dealt with conventional methods of energy production from hydrogen gas, which are based on integrated

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systems. Beheshti et al. [5] developed an Aspen Plus model to produce methanol from hydrogen rich gas, produced from biomass gasification in a Circulating Fluidized Bed (CFB). Galeno et al. [6] evaluated the potential of an Integrated Plasma Gasification/Fuel Cell (IPGFC) system to produce power with their results showing the system able to generate a net power of 4.2 MW/kg of Refuse Derived Fuel (RDF). Din and Zainal [7] reviewed the literatures on the combination of biomass gasification with fuel cells for Clean Energy Production (CEP). They attempted to provide a good understanding of biomass characteristics, the factors affecting the tar concentration, and the thermochemical conversion in gasifiers. Adefeso et al. [8] proposed an Aspen Plus model to study the influence of some critical parameters like equivalence ratio, metal catalysts, temperature variations, and gasification agents on hydrogen yield and cell voltage. Han et al. [9] carried out an experimental work in a self-design bubbling fluidized bed gasifier, aiming to explore the influence of $[CaO]/[C]$ and $[H_2O]/[C]$ on hydrogen production. They also developed an equilibrium model and compared the modeling study with the experimental measurements.

Various chemical and mechanical methods have been taken into consideration so that the emission of CO_2 from thermal processes like combustion and gasification can be decreased with the aim of preventing the rise of atmospheric greenhouse gases that may cause harmful climate changes [9,10]. Currently there are two chemical techniques as the most conventional ones to capture carbon dioxide from biomass gasification/combustion that have sufficiently high efficiency: (1) calcium looping which provides a gas stream with a relatively low CO_2 concentration [11] and (2) chemical absorption via monoethanolamine (MEA), widely used as a commercial method on an industrial scale [12]. Between the two, calcium looping is the most promising owing to its low cost and high reactivity. In this reaction, CO_2 reacts with CaO in the exothermic carbonation reaction ($CO_2 + CaO \rightarrow CaCO_3$) and produces $CaCO_3$. From a given sorbent with a constant value of steam/fuel ratio, it was found by other authors that the carbonation temperature and the cycle number are the parameters that directly affect the reactivity and absorbed CO_2 concentration [13]. Li et al. [14] conducted several experimental tests to clarify the role of atmosphere in coal combustion and gasification at the presence of CaO as an absorptive substance. Hejazi et al. [15] simulated an integrated biomass gasification and cyclic CO_2 capture in a Dual Fluidized Bed (DFB) to evaluate the influence of CO_2 capture on steam gasification of wood residue. Van Dyk et al. [16] conducted a thermodynamic study in order to identify the role of Ca-containing mineral components in CO_2 adsorption during gasification process and to explore the system's mechanism by means of a high temperature X-ray setup. Hu et al. [17] used a series of potential organic acids to improve the catalytic behavior of the natural CaO in an acidification-decomposition process and found the best performance when the system operates under oxy-fuel atmosphere.

From the above discussion, it was easily discovered that few studies referred to hydrogen production from biomass gasification in presence of CaO using a computer-based model. Therefore, the present work aims to propose a semi-kinetic model on the basis

of an Aspen Plus model to describe specific catalytic behavior of calcium on the gasification of rice husk. There has also been an attempt to validate the developed model by means of an experimental study and explore the influence of minerals within ash on gasification characteristics, since kinetic data are scarce in the literature.

2. Aspen Plus model

2.1. Model description

A semi-kinetic quasi-steady state model is proposed to simulate steam gasification of rice husk, using Aspen Plus simulator. The following conditions have been taken into consideration when developing the model: (1) steady-state operation; (2) atmospheric pressure condition; (3) fast devolatilization due to high temperature operation; (4) spherical geometry of particles; (5) negligible heat losses in process equipment; and (6) perfect mixing in the reactor. To improve the accuracy of the modeling results, a series of FORTRAN files have been inserted to Aspen Plus simulator to simulate the hydrodynamic properties, the rate of reactions, and chemical properties of the conventional components, where the rate of involved reactions can be found elsewhere [18–21]. The biomass stream “BIOMASS” is modeled by elemental compositions of rice husk obtained from elemental analysis, given in Table 1. The volatile matter “VM” and solids “SOLIDS” are introduced to the semi-kinetic reactor “RYEILD 1”, which is divided into separate zones, each simulating a specific process that occur in the reactor such as tar cracking and char oxidation. Two different separators namely separator column “SEPCOLUMN” and cyclone “CYCL” have been used to separate solid particles and tars from the gas stream. More details about the Aspen Plus reactor blocks can be found in Fig 1.

2.2. Hydrodynamic model

To improve the accuracy of the hydrodynamic model, fluidized bed has been divided into two regions: the lower region (bed) and the upper region (freeboard). It is also assumed that the fluidization state in the former is maintained within the bubbling phase. We have used a zero-dimensional correlation to predict the minimum fluidization velocity (U_{min}) in the bed region, which is as follows [22]

$$U_{min} = \frac{33.7\mu}{\rho_g d_p} (\sqrt{1 + 3.59 \times 10^{-5} Ar} - 1) \quad (2.1)$$

$$Ar = \frac{d_p^3 \rho_g (\rho_s - \rho_g) g}{\mu^3} \quad (2.2)$$

where μ , ρ_g , d_p , and Ar are viscosity (kg/ms), gas density (kg/m^3), particle diameter (m), and Archimedes number (–), respectively. To calculate the molar fraction occupied by bubbles in the fluidization regime, the following equation has been used which is strongly depended on the particle size (d_p) and U_{min} [22]

$$B = 1.0 + \frac{10.978(U - U_{mf})^{0.738} \rho_s^{0.376} d_p^{1.006}}{U_{mf}^{0.937} \rho_g^{0.126}} \quad (2.3)$$

$$\varepsilon_b = 1 - 1/B \quad (2.4)$$

Here U and ε_b are superficial gas velocity and volume fraction occupied by the bubbles respectively. Note that the minimum and superficial gas velocity is variable due to a significant difference in the rate of reactions. Kunii and Levenspiel [23] introduced

Table 1
Fuel properties.

Proximate analysis (in dry basis except moisture)		Ultimate analysis (in dry basis)	
Moisture	12.10%	C	39.78%
Fixed carbon	15.07%	H	04.97%
Volatile	70.36%	O	40.02%
Ash	14.57%	N	00.46%
LHV (MJ/kg)	14144.47	S	00.20%

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