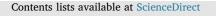
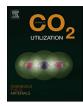
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Journal of CO₂ Utilization



journal homepage: www.elsevier.com/locate/jcou

Numerical investigation of CO_2 valorization via the steam gasification of biomass for producing syngas with flexible H_2 to CO ratio



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ARTICLE INFO

Keywords: CO₂ utilization H₂/CO ratio Syngas Steam gasification Equilibrium modelling

ABSTRACT

This work presents a numerical investigation into CO_2 valorization via the gasification of poplar wood. It is aimed at determining an optimum gasification condition for enhancing the production of syngas with a flexible H₂ to CO molar ratio (H₂/CO), which is essential in many petrochemical processes. The research is performed by simulation in ASPEN Plus, where steam and CO_2 are fed as co-gasifying agents. The equilibrium concentrations of the product gas are obtained, and the H₂/CO as well as the heating value of the resulting syngas are quantified. It is found that the inclusion of CO_2 as a co-gasifying agent promotes CO evolution through the Boudouard reaction. However, it reduces H₂ concentration, and consequently decreases the H₂/CO. Furthermore, the effects of some process parameters have been studied in this work. It is observed that H₂/CO reduces with a rise in temperature, increases with increasing CO₂ to biomass ratio (CBR), and shows no significant change with pressure. Results further show that methanol synthesis from syngas can be achieved at temperatures close to 660 °C, while oxo-synthesis requires a higher temperature. A CBR of around 0.6 in the present work would be an optimum value for Fischer-Tropsch synthesis to achieve a H₂/CO of 2:1, but the CBR should be lower for processes requiring a lower H₂/CO like acetic acid formation and oxo-synthesis.

1. Introduction

One of the critical issues facing mankind is the ever-increasing energy demand. To satisfy the demand, a significant portion of energy is generated from fossil fuels, which leads to CO₂ emissions. Due to the increasing concerns about global warming and climate change by these emissions, the use of alternative sources of energy has gained considerable encouragement in recent times. Reduction in the emissions may be achieved with the application of carbon-neutral energy sources and by carbon capture techniques [1,2]. One of the promising technologies in use is the conversion of biomass, which is carbon neutral, into combustible gaseous fuels via gasification. Although renewable energy sources enable carbon neutrality, it is increasingly becoming obvious that the problem of carbon emissions cannot be fully solved with just renewable energy resources. A recent report by the World Meteorological Organization (WMO) buttresses this point [3]. It was revealed that the global emission of CO2 reached a new record rise of 3.3 ppm from 2015 to 2016, culminating in a record high average of 403.3 ppm in 2016. This value was further reported to be around 145% of the pre-industrial era emission. A process that enables not just carbon neutrality but also negativity would be a pivotal step in solving the carbon emission problem. One of the possible means of achieving carbon negativity is by utilizing the captured CO₂ from industrial renewable energy plants. The CO₂ can be reused in many processes like in enhanced oil recovery, mineral carbonation, as a diluent, and as a feedstock for some chemicals and fuels, thus enhancing economic and environmental sustainability [4-6]. This enhancement makes carbon capture and utilization (CCU) a more beneficial and attractive technique than carbon capture and storage (CCS). The use of the captured carbon as an oxidizer in the gasification of a renewable fuel (biomass) will enable carbon negativity in gasification processes. In other words, CO₂ gasification or co-gasification of biomass is an important step in realizing sustainable energy generation and promoting carbon negativity. For this reason and due to the anxieties over CO₂ mitigation, the utilization of CO2 as an oxidizing medium in thermochemical conversion processes has become an important area of study. Some recent investigations have been reported with CO₂ as a gasifying/co-gasifying agent in biomass gasification [7-10]. A summary of some studies with CO_2 as a co-gasifying agent is presented in Table 1.

It is important to further explore the possible utilization of CO_2 in

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https://doi.org/10.1016/j.jcou.2018.07.003

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Received 15 February 2018; Received in revised form 11 June 2018; Accepted 5 July 2018 2212-9820/@ 2018 Published by Elsevier Ltd.

Nomeno	clature	n _{total}	То
		Р	Pre
As	Total number of atoms of the sth element	R	Un
a _{is}	Number of atom of the sth element in the molecules of a	SBR	Ste
	specie j	SCR	Ste
CBR	CO_2 to biomass ratio	Т	Te
G^t	Total Gibbs free energy, kJ	y _i	Mo
G^o_{fj}	Standard Gibbs free energy of formation of a specie j, kJ/	z	То
	kmol		
HHV	Higher heating value, MJ/Nm ³	Greek letters	
H_2/CO	H ₂ to CO molar ratio		
f_j	Fugacity of a specie <i>j</i> , atm	μ	Ch
Ĺ	Lagrange function	φ	Fu
Ν	Total number of species in the mixture	λ_s	La
n _i	Number of moles of a specie <i>j</i>		

Universal gas constant, 8.314 kJ/kmol. K

Total number of elements in the system

Steam to biomass ratio Steam to CO_2 ratio Temperature, K

Mole fraction of a specie *j*

Chemical potential Fugacity coefficient Lagrange multiplier Journal of CO₂ Utilization 27 (2018) 32-41

free energy is utilized in the gasification process. The main objective of this work is to study the adjustment of H₂/CO ratio in the resulting syngas after the inclusion of CO₂ in the oxidizer stream of biomass steam gasification. This study will be helpful in the optimal design of a biomass gasifier with the use of both steam and CO₂ as the gasifying agents. It will also aid the generation of syngas that can be directly used for many downstream applications without further adjustment, reducing operational and investment costs. The H₂/CO ratio is a critical factor in various processes including the Fischer-Tropsch's gas-to-liquid (GTL) process and methanol production, with both requiring a ratio of 2:1, acetic acid formation and oxo-synthesis with ratios of 1:1. This work aims to generate syngas for these processes and therefore, a technique of lowering the H₂/CO ratios to values between 1 and 2 on a molar basis is numerically investigated. CO₂ conversion in terms of CO/ CO₂ molar ratio is also evaluated. Additionally, the energy content in terms of higher heating value and the composition of the resulting syngas after CO₂ inclusion in the gasification are discussed in detail.

2. Methodology

2.1. Assumptions

To simplify the gasification simulation in this work, the following assumptions are made:

- 1 The gasification system is operated under isothermal, isobaric and steady-state conditions, and all the unit operations are operated at 1 atm.
- 2 The product gas stream is comprised of only CO, H₂O, H₂, CO₂, N₂, and CH₄, neglecting higher hydrocarbons.
- 3 All the reactions in the gasifier are assumed to have attained thermodynamic equilibrium.
- 4 All the gases obey the ideal gas law.
- 5 The biomass and CO_2 are supplied at room temperature and atmospheric pressure, while steam is supplied at 150 °C and 1 atm.
- 6 Heat loss and pressure drop are not considered in the gasification.
- 7 Char contains only carbon and ash, ash is inert and tar formation is neglected.

2.2. Gibbs free energy equilibrium model

The model in this work is based on the minimization of Gibbs free energy. This model is a non-stoichiometric equilibrium model, and in this case, there is no need to specify chemical reactions and the equilibrium constant. The model is an efficient way of finding the equilibrium gas composition from the gasification of biomass [23]. The mathematical models developed in previous studies [24–26] illustrate the Gibbs free energy equilibrium model. This is described in the

gasification. In this way, a direct conversion of a greenhouse gas (CO_2) to gaseous fuel can be achieved. Thus, CO2 gasification can generate a useful fuel gas (carbon monoxide) for many petrochemical applications. Interestingly, CO is an important gas in the production of many chemicals like organic acids, methanol synthesis, agricultural chemicals, polycarbonates, and dimethyl ether (DME) [16-19]. Thus, these processes require syngas with a low H₂/CO ratio. An effective way of reducing emissions can be achieved by enhancing the Boudouard reaction in steam gasification with the use of the captured CO₂ as a co-gas agent. As a result, attention may be further shifted from CCS to CCU. It is well known that steam gasification of biomass is an important process due to the formation of hydrogen-enriched gas under a reducing atmosphere. Promotion of the Boudouard reaction in this process may also be important, especially if the goal of the process is to adjust the H₂ to CO molar ratio (H₂/CO) of the syngas. It is expected that the inclusion of CO_2 in a steam gasifier will enhance CO evolution and reduce H_2 concentration, lowering the H_2/CO ratio. The flexibility of this ratio with CO₂ as a gasifying agent makes the syngas readily available for use in the Fischer-Tropsch's gas-to-liquid (GTL) process for the production of liquid fuel for mobile fuel cell applications, which can be more easily transported than hydrogen. Other processes requiring a low H₂/CO ratio include methanol production, acetic acid formation, and oxosynthesis with H₂/CO ratios of 2:1, 1:1 and 1:1 respectively [20-22]. The elemental composition of biomass is also an influential factor on the H₂/CO ratio of syngas. The results reported by Cao et al. [21] showed that H₂/CO ratio is more dependent on the elemental compositions (H/C ratio) than it is on the reactivity of the fuel. This has been validated in the study by Ravaghi-Ardebili et al. [22], where the authors claimed that due to the higher carbon content in lignin-based biomass, it showed a higher H₂/CO with increasing temperature than cellulose-based biomass. The H₂/CO ratio varied from 0.6 - 0.78 and 0.81-1.02 for the cellulose- and lignin-based respectively when temperature was increased from 550 to 800 K. The aforementioned studies have shown that in addition to reducing the release of greenhouse gas emissions and the cost of carbon sequestration, feeding of CO₂ as a gasifying agent in a steam gasifier aids in the adjustment of the H₂/CO ratio of the produced gas. Consequently, the ratio can be controlled with the aid of CO₂ variation in the gas agent stream, this can further be tailored to an application of interest. Only a few experimental studies on steam/CO₂ gasification are available in the literature. To the best of our knowledge, numerical simulation of woody biomass gasification in a mixed atmosphere of steam and CO₂ is not available; especially for the production of H₂/CO ratio suitable for various chemical processes. This suggests a need to numerically investigate the gasification in a mixture of steam and CO₂. In this work, a numerical investigation into the valorisation of CO₂

In this work, a numerical investigation into the valorisation of CO_2 through its utilization as a co-oxidizing medium in the steam gasification of a biomass (poplar wood) is performed. A model based on Gibbs

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