Evaluation of a Green Methanol Production System Using the Integration of Water Electrolysis and Biomass Gasification

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For a long time, the development of green shipping has been highly valued by countries and organizations. Biomass gasification-based green methanol is seen as a long-term alternative to conventional shipping fuel to reduce greenhouse gas emissions in the maritime sector. While the operational benefits of renewable methanol as a marine fuel are well-known, its cost and environmental performance depend largely on the production method. In this study, a green methanol production system based on the integration of biomass gasification and water electrolysis is proposed and evaluated via the parametric and thermodynamic analysis methods. The water electrolysis is used to increase the hydrogen content in syngas, thereby increasing the production of methanol. The results show that as the S/C ratio increases, the mass flow rate and the calorific value of product gas, the mole flow rate of methanol decreases. The enhancement of the H_2/CO ratio can increase the mole fraction of H_2, thereby increasing the methanol yield. The mole flow rate of methanol dramatically increases from 925.0 kmol/h to 3725.2 kmol/h. Additionally, the mole flow rate of methanol in the proposed system is 10776.0 kmol/h, larger than the traditional system of 3603.4 kmol/h. The carbon element conversion rate of the proposed system is 94.6%, higher than the 31.5% of the traditional system. This system can significantly provide an efficient green methanol production method for the shipping sector, while also helping to find a feasible solution for the consumption of renewable energy.

Key words*: biomass gasification, green methanol, shipping fuel, water electrolysis*

1. Introduction

Methanol is a significant chemical product that is used in the production of many other chemicals, including paints, plastics, and building supplies, as well as formaldehyde, dimethyl ether, methyl tertbutyl ether, and acetic acid [1, 2]. Nowadays, fossil fuels like coal and natural gas are used to make a significant amount of methanol, which increases energy consumption and creates numerous

environmental issues [3, 4]. Due to its coal-based energy structure, coal gasification technology produces over 65% of methanol in China. [5]. Nevertheless, the manufacture of methanol from coal has a higher carbon emission intensity because of the high carbon content of coal [6]. Besides, China has pledged to reduce carbon emissions per GDP unit by 60–65% from 2005 levels by 2030 and achieve carbon neutrality by 2060 to address the climate catastrophe. [7]. Therefore, action must be taken to achieve net zero carbon output in the methanol business given the pressing need to reduce CO_2 emissions.

Alternatives to fossil fuels that are sustainable and renewable include biomass, sun, wind, hydropower, and hydropower [8, 9]. Biomass, a renewable resource, has steadily come under pressure to be used in industrial production in an effort to lessen the strain on the environment and energy sources of today [10]. One of the most promising commodities and fuel products of the future is green methanol made from biomass. [1].

The methanol synthesis process is the subject of several investigations aimed at producing alternative green fuels. Blanco et al. [11] evaluated how methanol and ammonia are converted to electricity. Based on the findings of the technical investigation, the methanol process had the highest efficiency of 38%. Sollai et al. [4] created a green methanol plant that captures carbon dioxide and uses hydrogen electrolysis. The findings demonstrated that although green methanol was not economically competitive, the technique was predicted to become viable in the future. Wang et al. [12] suggested a revolutionary method of producing methanol with almost negligible carbon emissions, and the suggested system escaped the need for a water-gas shift unit to lower CO_2 output at the source. The new process has an energy efficiency of 130.8 % and a carbon utilization efficiency of 82.9 % compared to the traditional plant. Sun et al. [3] examined the direct chemical looping processes' thermodynamic and techno-economic outcomes for the two suggested approaches for producing methanol from biomass. According to the results, the second configuration of the biomass chemical looping hydrogen production system was more economically viable, while the syngas conditioning process in the first configuration of the biomass chemical looping gasification system had higher exergy destruction. Shahabuddin et al. [13] evaluated the co-gasification performance using a fluidized bed gasifier at different ratios of pine bark biomass and bituminous coal. Grosso et al. [14] examined the technique for producing methanol via biomass gasification. Aspen Plus was used to mimic various processing processes, including gasification, methanol production, and energy conversion. Qin et al. [5] demonstrated the impact of adding hydrogen-rich sources from solar-based hydrogen and biomass co-gasification on the environmental, economic, and technological performance of methanol plants. Li et al. [15] assessed and replicated the process of producing methanol via a biomass gasification system. According to the findings of the sensitivity analysis, an overall exergetic efficiency of 30.5% was attained at mass ratios of 0.14 for O_2-rich gas and 0.26 for steam relative to biomass. Bai et al. [16, 17] examined the thermodynamic and financial results of producing power from a solar-biomass gasification system using liquid fuel methanol. According to the results, the intended methanol production capacity is 51.2×10^3 tons/year, while the power capacity is 32.7 MW. The energy efficiency of 51.9 % and the energy efficiency of 51.2 % were attained.

We seek as much as possible to transform biomass—which contains valuable carbon—into green fuel. Thus, adding green hydrogen throughout the green methanol synthesis process can enhance the pace at which carbon elements are converted and raise the production of green fuels. Clausen et al. [18] devised a renewable energy-based methanol production process architecture, utilizing

subterranean gas storage for hydrogen and oxygen in conjunction with electrolysis. According to the results, the suggested plant had methanol exergy efficiencies of 59~72 %. The best results come from a setup that produces syngas by electrolyzing water and automatically thermally reforming biogas. Gu et al. [7] centered on a green methanol plant that included a CO_2-based methanol synthesis system downstream and an upstream green hydrogen-electricity generating system. To determine the most cost-effective operating mode for the suggested system, a few configurations were taken into consideration. According to the results, the green methanol plant with oxygen sales and a hydrogen production system driven by solar energy and wind had the lowest levelized cost. Zhao et al. [19] suggested a method of converting coal into methanol with the use of green hydrogen produced by three different methods of electrolyzing water using solar, wind, or hybrid solar and solar electricity. The final approach showed significant viability in terms of economy, environment, and technology, according to the results.

In this study, to increase the carbon elements in biomass, a novel methanol synthesis system coupled with biomass gasification and water electrolysis is investigated and compared with the traditional methanol synthesis plant. The hydrogen generated from the water electrolysis is considered to mix with the syngas from the gasification process to increase the ratio of H_2/CO. Moreover, the performance of the proposed system is parametrically analyzed under different ratios of steam to carbon (S/C).

2. Methodology

2.1. Process modeling

As a benchmark case, the conventional methanol synthesis process comprises five main parts. The biomass should first react with the oxygen in the air and be gasified to generate crude syngas in the gasifier. To meet the requirement of the H_2/CO ratio, a portion of the crude syngas is split into the water gas shift (WGS) unit. After that, the crude syngas is delivered to the acid gas removal (AGR) unit to be treated for acid gas (sulfur). The synthesis and distillation of methanol can be done with the refined syngas.

Since biomass contains important carbon, as much of it as possible should be turned into green fuel. The schematic diagram of the proposed green methanol synthesis process is shown in Fig. 1. In the gasifier, the biomass initially combines with oxygen from the electrolysis unit to create crude syngas. One benefit of employing pure oxygen for oxidation in a gasifier rather than air is that it can lower investment and equipment size. Crude syngas can be directly transported to the AGR unit for removing sulfur without involving the WGS unit. The generated green hydrogen is mixed with the crude syngas to increase the ratio of H_2/CO. The high concentration of H_2 in syngas contributes to increased production of green methanol and enhanced rate of conversion of carbon components in biomass to green fuel.

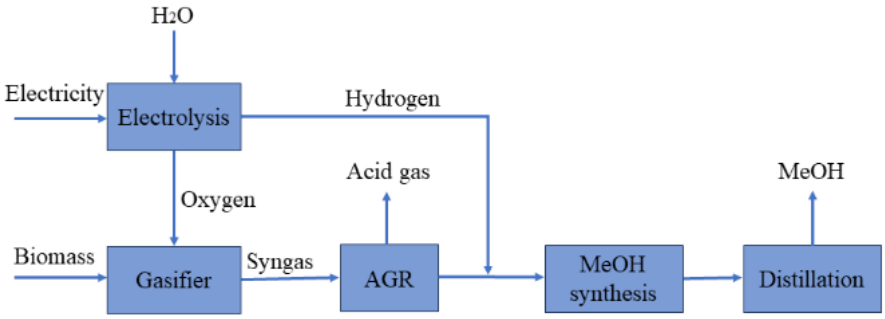

Figure 1. Schematic diagram of the proposed green methanol synthesis process

The associated processes and their linkages are modeled and implemented in the generic simulation software for this proposed biomass gasification and water electrolysis-based green methanol production system. The following are the primary presumptions that the modeling took into account:

(1) The chemical reactions are in a state of thermodynamic equilibrium and the fluid is in a stable flow condition.

(2) It is acceptable to overlook the heat loss from the system to the surroundings, pressure drop, and gas flow resistance.

(3) The system components are examined using the thermodynamic model, and the thermodynamic parameters have a uniform distribution.

(4) At atmospheric conditions (101.3 kPa and 25 °C), air (composition by volume: 79% nitrogen, 21% oxygen) enters the system.

2.2. Water gas shift process

The hydrogen to carbon ratio of synthesis gas $M = n(\mathrm{H_2})/n(\mathrm{CO}+1.5\mathrm{CO_2})$ is too low, indicating that the CO content in the syngas is relatively high, which does not meet the requirement of hydrogen to carbon ratio M=2.1~2.15 for methanol synthesis. The WGS process is required to convert the excess CO into H_2 and CO_2, and the methanol distillation process can be used to eliminate the excess CO_2. The reaction can be expressed as follows:

$$\mathrm{CO}+\mathrm{H_2O} \rightarrow \mathrm{CO_2}+\mathrm{H_2} \quad \Delta H = -38.4\ \mathrm{kJ/mol} \tag{1}$$

The process flowsheet of the WGS unit is shown in Fig. 2. The composition between reference data and simulated results in the WGS unit is listed in Table 1, moreover, the specific parameters for each stream are listed in Appendix A. It indicates that the model and the reference results are in very good agreement.

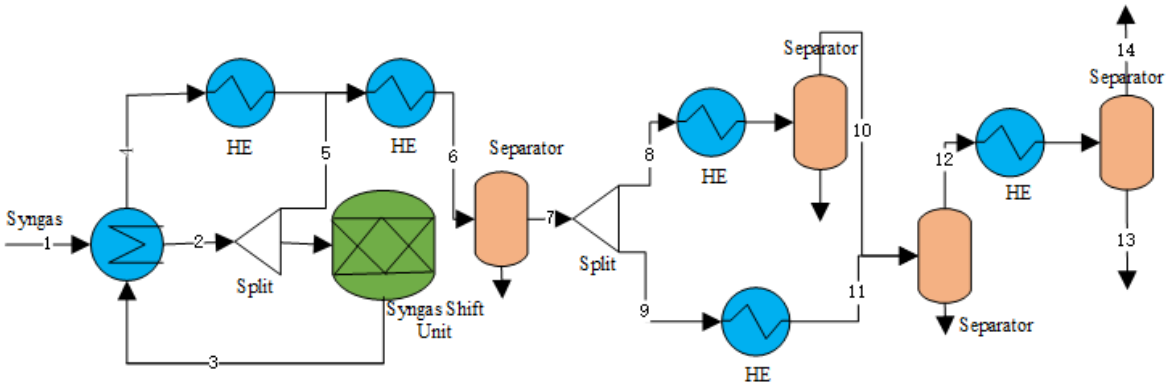

Figure 2. Process flowsheet of the WGS unit

Table 1. The composition between reference data and simulated results in WGS unit

Components	Stream 1		Stream 14	
	Ref. [24]	Sim.	Ref. [24]	Sim.
CO (%)	24.9	24.9	22.9	22.5
CO_2 (%)	1.64	1.64	30.7	31.0
H_2 (%)	9.2	9.2	46.1	46.2
H_2O (%)	64.3	64.3	0.31	0.26

2.3. Methanol synthesis and distillation

The methanol reactor receives the generated syngas. Low temperatures and high pressures are ideal for the reaction [21]. To ensure that the catalysts are active and to efficiently use the reaction heat, the ideal conditions for methanol synthesis are typically in the range of 30~80 bar and 200~350°C. [8]. For the equilibrium methanol synthesis, the Gibbs free energy minimization processes (2) and (3) are utilized to compute the equilibrium, and the equilibrium reactors are thought to be adiabatic [8].

$$CO+2H_2 \rightarrow CH_3OH \quad \Delta H=-90.7\ kJ/mol \tag{2}$$

$$CO_2+3H_2 \rightarrow CH_3OH+H_2O \quad \Delta H=-40.9\ kJ/mol \tag{3}$$

Based on a dual-site adsorption mechanism, the methanol synthesis reaction kinetics are expressed as follows for the reaction rates [22,23]:

$$r_{CO_2}=\frac{k_1\cdot K_2\cdot f_{CO_2}\cdot f_{H_2}^{1.5}\left(1-\frac{f_{MeOH}\cdot f_{H_2O}}{f_{CO_2}\cdot f_{H_2}^{3}\cdot K_{eq,1}}\right)}{(1+K_1\cdot f_{CO}+K_2\cdot f_{CO_2})\cdot(f_{H_2}^{0.5}+K_3\cdot f_{H_2O})} \tag{4}$$

$$r_{rWGS}=\frac{k_2\cdot K_2\cdot f_{CO_2}\cdot f_{H_2}\left(1-\frac{f_{CO}\cdot f_{H_2O}}{f_{CO_2}\cdot f_{H_2}\cdot K_{eq,2}}\right)}{(1+K_1\cdot f_{CO}+K_2\cdot f_{CO_2})\cdot(f_{H_2}^{0.5}+K_3\cdot f_{H_2O})} \tag{5}$$

where *fi* stands for the fugacity of component *i* (Pa); The revised water-gas shift reaction (rWGS) and CO_2 hydrogenation have reaction rate constants of k_1 and k_2, respectively; K_1, K_2 and K_3 are the constants of adsorption for CO, CO_2 and H_2O, respectively; $K_{eq,1}$ and $K_{eq,2}$ stand for the equilibrium constants for the reverse water-gas shift process and CO_2 hydrogenation, respectively. The reaction parameters are directly taken from the literature [23].

The compressed syngas from the water electrolysis is combined with H2 in the suggested setup. An external direct current source facilitates the flow of electrons while anions (O^{2-}) or cations (H^+) are exchanged between electrodes during the electrolysis process. At the electrodes, ions and electrons recombine to generate oxygen and hydrogen.

The process flowsheet of the methanol synthesis and distillation unit is shown in Fig. 3. The composition between reference and simulated results in the methanol synthesis and distillation unit is listed in Table 2, moreover, the specific parameters for each stream are listed in Appendix B. The reference data and the methanol synthesis and distillation simulation results were compared. It is evident that the reference data and the simulated findings of methanol production and distillation agree.

Pre-distillation columns are taken into consideration for the methanol distillation process in order to eliminate light components like dimethyl ether that are present in the crude methanol product. In order to obtain approved methanol products, water and contaminants with a high boiling point are removed using pressurized and atmospheric distillation columns. At the top of both the pressurized and atmospheric distillations, the methanol product is eventually collected. The purity of the methanol product is 99.9%.

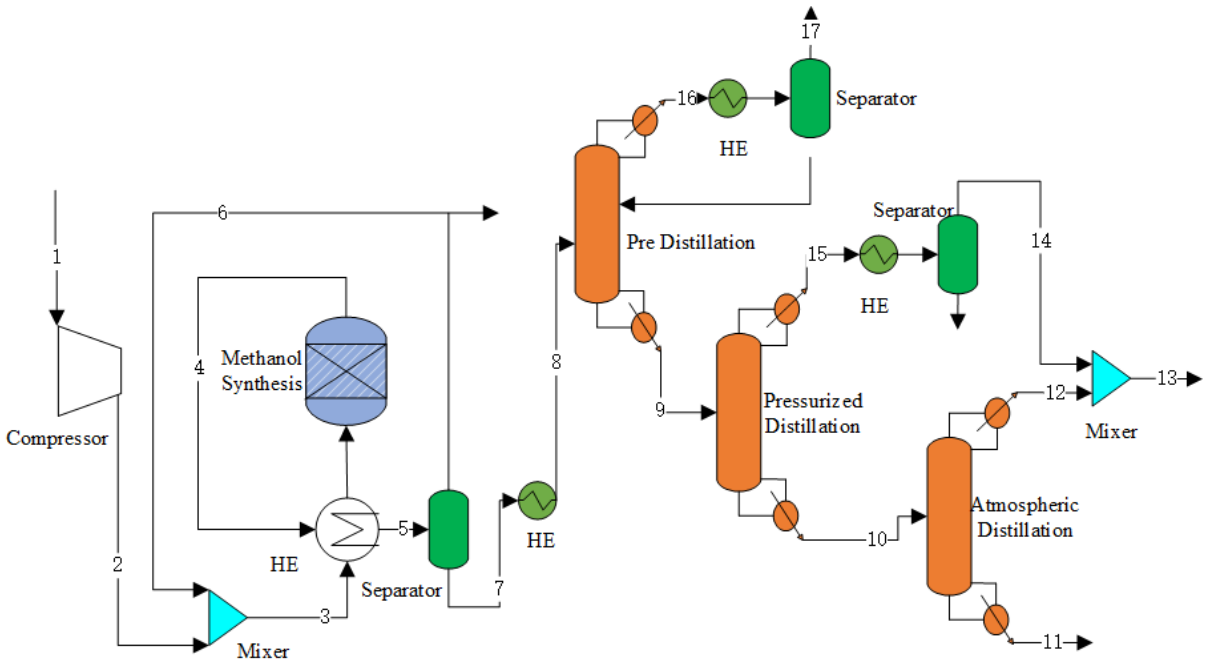

Figure 3. Process flowsheet of the methanol synthesis and distillation unit

Table 2. The composition between reference and simulated results in methanol synthesis and distillation unit

Components	Stream 1		Stream 13	
	Ref. [22]	Sim.	Ref. [22]	Sim.
CO (%)	28.88	28.88	0	22.49
CO_2 (%)	1.34	1.34	0	31.01
H_2 (%)	67.21	67.15	0	46.23
H_2O (%)	0	0	0	0.26
CH_3OH (%)	0	0	99.98	99.99

3. Results and discussion

For the WGS unit, the number of moles of water vapor divided by the number of moles of carbon dioxide and carbon monoxide is known as the steam-to-carbon (S/C) ratio ($M(H_2O)/M(CO+CO_2)$). The increase in water to gas ratio is beneficial for improving the conversion rate of CO reaction and accelerating the conversion rate. On the other hand, it is beneficial for controlling the bed temperature. But if the value is increased, it will reduce the reaction time of CO, increase the load of waste heat recovery equipment, and thus affect the production capacity of the device. The effect of the S/C ratio on the molar flow rate of methanol is shown in Fig. 4. As is displayed the mole flow rate of methanol decreases with the S/C ratio increasing.

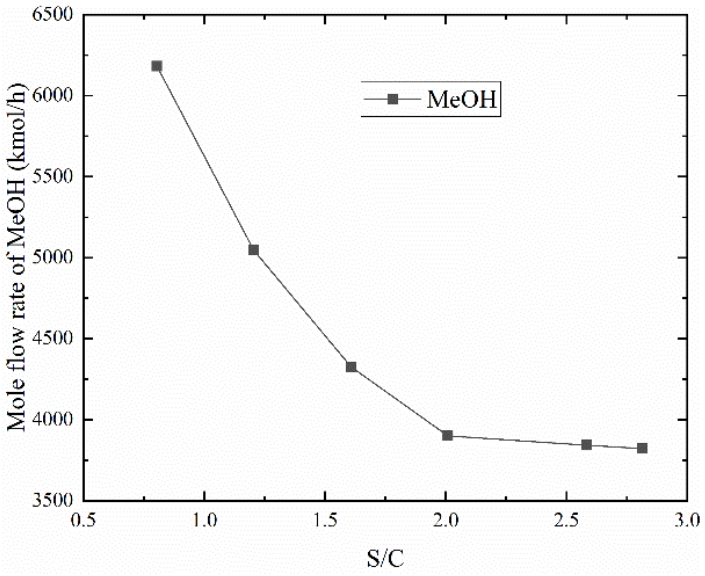

Figure 4. Effect of S/C ratio on the molar flow rate of methanol

The effect of the S/C ratio on product gas molar flow rate is shown in Fig. 5. It can be seen that the molar flow rate of CO, CO_2, and H_2 drops with the S/C increasing. Additionally, the effects of the S/C ratio on $\dot{m}_{product}$ and $\dot{Q}_{product}$ are shown in Fig. 6. It can be seen that the mass flow rate of product gas (

$\dot{m}_{product}$) and calorific value of product gas ($\dot{Q}_{product}$) decrease with the S/C increasing. For the reason that, the ratio of S/C increases will increase energy consumption, as well as increase bed resistance and burden on waste heat recovery equipment. Therefore, the amount of steam should be reasonably adjusted based on factors such as gas composition, conversion rate requirements, reaction temperature, and catalyst activity.

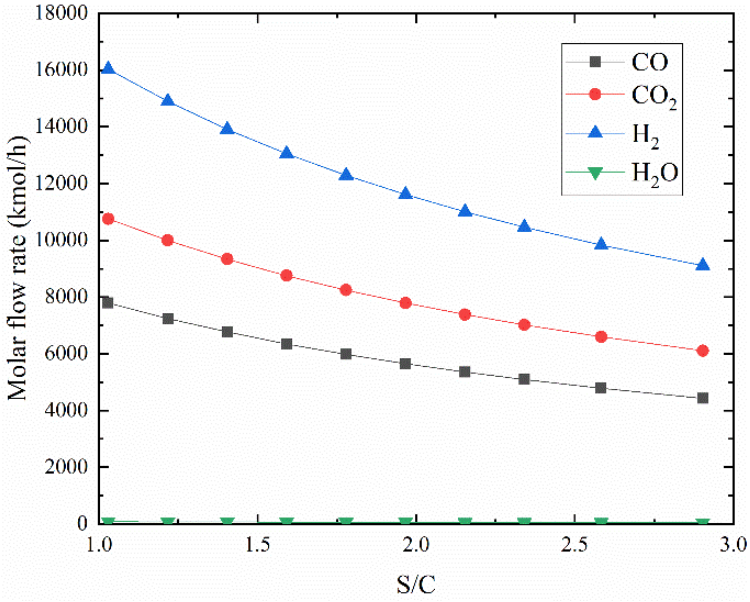

Figure 5. Effect of S/C ratio on the molar flow rate of product gas

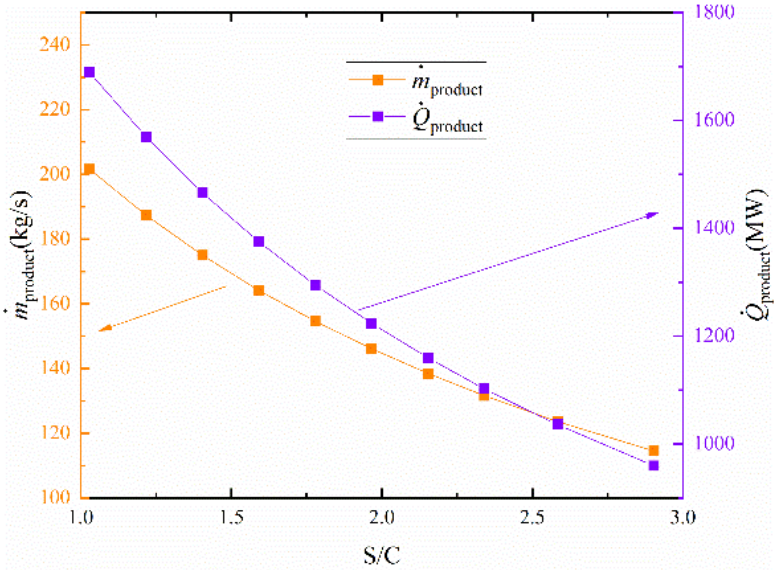

Figure 6. Effect of S/C ratio on the $\dot{m}_{product}$ and $\dot{Q}_{product}$

The H_2/CO ratio has a significant influence on the methanol yield. The variation of H_2/CO ratios in the range of 0.4~3.0 is investigated at 240 ℃ and 83 bar. The effect of the H_2/CO ratio on the molar fraction and mole flow rate of MeOH are shown in Fig. 7 and Fig. 8. As can be seen, the enhancement

of the H_2/CO ratio can increase the mole fraction of H_2, thereby increasing the methanol yield. The mole flow rate of methanol dramatically increases from 925.0 kmol/h to 3725.2 kmol/h. Herein, it should be noticed that the mole fraction of product gas is adjusted in the WGS unit, and the mass flow rate of the product gas increases with the H_2/CO ratio increasing. Therefore, the mole flow rate of methanol shows a gradual upward trend.

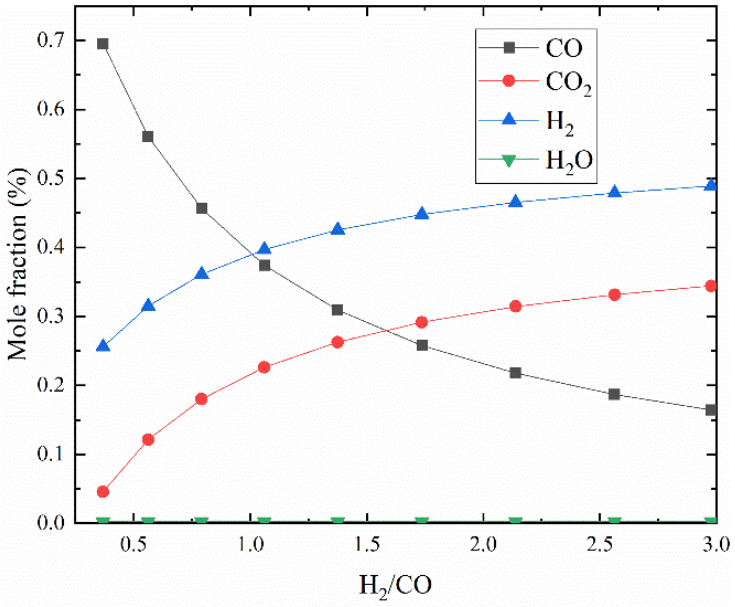

Figure 7. Effect of H_2/CO ratio on molar fraction

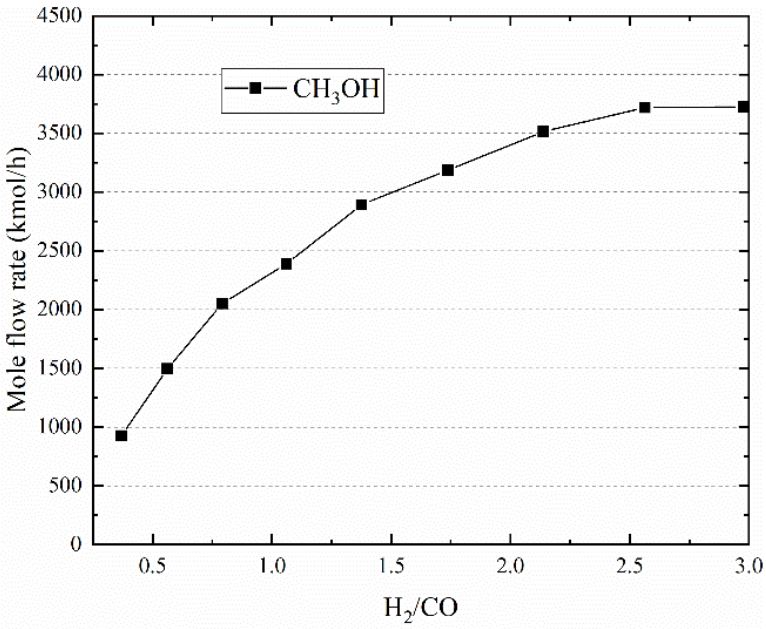

Figure 8. Effect of H_2/CO ratio on the molar flow rate of MeOH

In this proposed biomass gasification and water electrolysis-based green methanol production system, the H_2 generated from the water electrolysis unit is considered to be mixed with the product

gas, and the WGS unit can be ignored. Accordingly, in this present system, the carbon element in biomass can be maximally converted into CO, and increase the yield of methanol. As can be seen from Fig. 9, the mole flow rate of methanol in the proposed system is 10776.0 kmol/h, larger than the traditional system of 3603.4 kmol/h at the H_2/CO ratio of 2. Herein, the traditional system refers to the methanol synthesis systems without additional hydrogenation. Where, the proposed system refers to the methanol synthesis systems without additional hydrogenation. Moreover, the carbon element conversion rate of the proposed system is 94.6 %, higher than the 31.5 % of the traditional system.

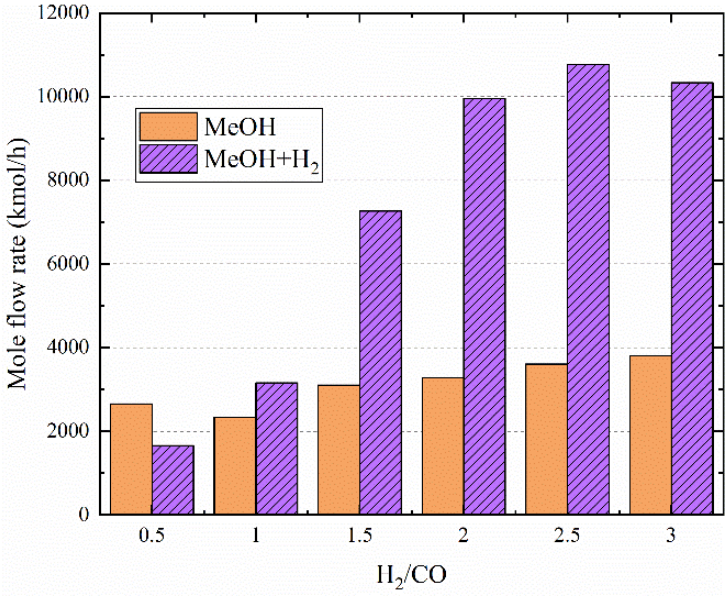

Figure 9. Comparisons of H_2/CO ratio on the molar flow rate of MeOH

4. Conclusion

In this study, a green methanol production system based on the integration of biomass gasification and water electrolysis is proposed. The models of WGS, methanol synthesis, and distillation are validated with the reference data. The parameter analysis and thermodynamic analysis are conducted. The conclusion can be shown as follows:

(1) For parameter analysis, the molar flow rate of CO, CO_2, and H_2 drops with the increase of S/C. Moreover, the mass flow rate of product gas ($\dot{m}_{product}$), the calorific value of product gas ($\dot{Q}_{product}$), and the mole flow rate of methanol decrease with the increase of S/C; The enhancement of the H_2/CO ratio can increase the mole fraction of H_2, thereby increasing the methanol yield. The mole flow rate of methanol dramatically increases from 925.0 kmol/h to 3725.2 kmol/h.

(2) For thermodynamic analysis, considering the carbon element in biomass can be maximally converted into CO for the proposed system, the mole flow rate of methanol is 10776.0 kmol/h, larger than the traditional system of 3603.4 kmol/h. Moreover, the carbon element conversion rate of the proposed system is 94.6 %, higher than the 31.5 % of the traditional system.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgment

This work was supported by the Jilin Provincial Department of Education (JJKH20230115KJ).

Appendix

The specific parameters for each stream of the WGS unit are listed in Appendix A.

***Appendix A*. Calculated thermodynamic parameters of the WGS unit.**

No.	*T* (℃)	*P* (MPa)	*H* (kJ/kg)	m_{mol} (kmol/s)	m_{mass} (kg/s)
1	188.0	3.7	3389.2	11.9	231.9
2	195.6	3.7	3244.4	11.9	231.9
3	477.4	3.5	1474.2	8.34	162.4
4	383.1	3.5	1414.3	8.34	162.4
5	195.6	3.7	1527.3	3.58	69.6
6	159.0	0.4	1205.2	11.9	231.9
7	159.0	0.4	7679.9	11.9	231.9
8	159.0	0.4	7559.5	8.34	162.4
9	159.0	0.4	7440.9	3.58	69.6
10	64.0	3.2	7338.2	4.27	89.04
11	47.3	3.1	14543.1	3.58	69.6
12	50.0	3.2	7477.0	5.92	123.8
13	40.0	3.1	7477.0	5.91	123.6
14	50.0	3.2	7477.0	1.93	34.8

The specific parameters for each stream of methanol synthesis and distillation unit are listed in Appendix B.

***Appendix B*. Calculated thermodynamic parameters of the methanol synthesis and distillation unit.**

No.	*T* (℃)	*P* (MPa)	*H* (kJ/kg)	m_{mol} (kmol/s)	m_{mass} (kg/s)
1	32.0	5.50	3389.2	2.36	25.8
2	85.1	8.30	3244.4	2.36	25.8
3	47.4	0.40	1474.2	14.3	195.3
4	240.0	8.30	1414.3	12.8	195.3
5	188.7	8.30	1527.3	12.8	195.3
6	40.0	0.40	1205.2	11.9	169.5
7	40.0	0.40	7679.9	0.73	22.8
8	73.0	0.24	7559.5	0.73	22.8

9	105.2	0.42	7440.9	0.72	22.8
10	130.0	8.50	7338.2	0.72	22.8
11	165.5	0.88	14543.1	0.03	0.62
12	40.0	1.00	7477.0	0.30	9.55
13	40.0	1.00	7477.0	0.69	22.2
14	40.0	1.00	7477.0	0.39	12.6
15	129.0	0.85	6147.7	0.39	12.6
16	74.1	0.15	6202.2	0.28	9.03
17	63.6	0.15	4313.3	0.00	0.04

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Submitted: 26.02.2024
Revised: 17.05.2024
Accepted: 28.05.2024