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Co-Production of Hydrogen and Methanol Using Fuel Mix Systems: Technical and Economic Assessment

Usama Ahmed ^{1,2,*} , Umer Zahid ^{1,3}, Sagheer A. Onaizi ^{1,2}, Abdul Gani Abdul Jameel ^{1,4} , Nauman Ahmad ^{5,6}, Nabeel Ahmad ⁷ and Hamad AlMohamadi ⁸

- ¹ Chemical Engineering Department, King Fahd University of Petroleum and Minerals, Dhahran 31261, Saudi Arabia; uzahid@kfupm.edu.sa (U.Z.); onaizi@kfupm.edu.sa (S.A.O.); a.abduljameel@kfupm.edu.sa (A.G.A.J.)
- ² Interdisciplinary Research Center for Hydrogen and Energy Storage, King Fahd University of Petroleum & Minerals, Dhahran 31261, Saudi Arabia
- ³ Interdisciplinary Research Center for Membranes & Water Security, King Fahd University of Petroleum & Minerals, Dhahran 31261, Saudi Arabia
- ⁴ Center for Refining and Advanced Chemicals, King Fahd University of Petroleum & Minerals, Dhahran 31261, Saudi Arabia
- ⁵ Department of Chemical Engineering, Monash University, Clayton 3800, Australia; nauman_1@live.com
- ⁶ Institute of Chemical Engineering and Technology, University of the Punjab, Lahore 53700, Pakistan
- ⁷ Department of Chemical Engineering, COMSATS University Islamabad, Lahore 54000, Pakistan; nabeelahmad@cuilahore.edu.pk
- ⁸ Department of Chemical Engineering, Faculty of Engineering, Islamic University of Madinah, Madinah 42351, Saudi Arabia; hha@iu.edu.sa
- * Correspondence: usama.ahmed@kfupm.edu.sa



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Abstract: With the increase in global energy requirements, the utilization of fossil fuels has also increased, which has caused global warming. In this study, a process integration framework based on an energy mix system is proposed to simultaneously produce two cleaner fuels (methanol and H₂). Aspen Plus is used to develop process models followed by their techno-economic assessment. Case 1 is considered the base case process, where the coal–biomass gasification process is used to produce the synthesis gas, which is further converted into H₂ and methanol. Conversely, the case 2 design represents the novel process configuration framework, where the coal–biomass gasification technology in case 1 is sequentially integrated with the methane reforming technology to minimize the energy penalties while increasing the net fuel production. To perform the technical analysis, the fuel production rates, carbon conversion efficiencies and specific energy requirements are compared for both models. It is analyzed from the results that the case 2 design offers higher methanol and H₂ production rates with lower energy requirements. Additionally, the specific energy requirement for case 2 is 29% lower compared to the case 1 design, leading to an increase in the process efficiency of case 2 by 3.5%.

Keywords: gasification; reforming; methanol; hydrogen; carbon capture; process integration



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1. Introduction

Global warming and greenhouse gas emissions have become one of the biggest debated issues around the globe. With recent industrialization and deforestation, the carbon footprint around the world has increased to dangerous levels during the last few decades. The intergovernmental panel on climate change (IPCC) clearly indicates this in its reports about global warming and the depletion of energy resource [1,2]. A reduction in greenhouse gas emissions can be achieved by either minimizing fuel consumption, by improving the process performance, by reducing the greenhouse gas emissions or by using renewables forms of energy. The current global capacity of renewables is not enough to meet the world energy and supply demand. Therefore, the dependence on

conventional fuel resources will remain dominant in the future too. Energy in any form is very valuable around the world to meet societal and industrial needs, and a lot of research has been dedicated to improving the process efficiency or to finding alternative energy resources. Hydrocarbon-based fossil fuels have remained the major source of energy supply during the last few decades, owing to their high efficiency and availability. However, the utilization of fossil fuel based-processes also increased the global CO₂ emissions to the highest level ever.

Gasoline, diesel and natural gas are the main fuels that are used in the automotive, chemical, fertilizer and process industries, the use of which steadily increased at a rate of nearly 2–2.5% annually, paralleling the global economic growth, which will be further increased up to 50% by the year 2030 [3]. Compared to conventional fuels used for power generation, methanol and hydrogen are considered not only cleaner fuel sources but also raw materials for many process industries [4]. Oxygenates such as methanol and ethanol are also blended with petroleum fuels to improve various combustion properties such as octane number [5] and emission characteristics such as sooting [6] propensity. Large-scale methanol and hydrogen production processes are based on natural gas feedstocks using intermediate reforming technologies for synthesis gas production. As hydrogen is a key component of methanol production, the increase in methanol production also increases the need for hydrogen production. Yao et al. [7] also mentioned that the methanol demand has increased two times during the last 5–7 years, which also increases the methanol production requirement by 1.7 times. Methanol can be produced from conventional fuels that can be converted into synthesis gas, mainly composed of CO and H₂. For instance, commercially, natural-gas-to-methanol (NGTM) processes are used where the natural gas is first reformed to generate synthesis gases with higher hydrogen-to-carbon (HCR) ratios followed by methanol synthesis. Coal-to-methanol (CTM) production processes have also received a lot of attention due to the lower cost of coal compared to natural gas and the larger reserves around the world. The CTM production process is slightly more complex compared to the natural-gas-to-methanol processes as it contains more unit processes. Khalafalla et al. [8] demonstrated pathways to produce methanol from coal by developing alternative designs and compared the results in terms of carbon conversion, specific energy consumption and methanol production cost.

Reaching a fully sustainable clean energy source for power, electricity and cleaner fuel production is a long ways off, and a lot of research is being focused on finding alternative pathways to increase the efficiency of already existing systems. Castellani et al. [9,10] studied the carbon-neutral process of producing methane, methanol and ammonia using a solar-based renewable hydrogen supply process. Poly-generation processes are under the spotlight for being one of the most promising solutions for this problem. This is because poly-generation processes are flexible in combining multiple energy sources that can enable the sustainable supply of energy demand while meeting environmental regulations in terms of reduced greenhouse gas (GHG) emissions. For a sustainable energy system, available resources should be utilized at the maximum possible efficiency followed by a reduction in GHG emissions. Ng et al. [11] and Jana et al. [12] provided reviews on poly-generation processes using multiple feedstocks with multiple outputs. The poly-generation process can be utilized for co-currently producing multiple chemicals and products including electricity, fuel, chemicals, etc.

In this study, poly-generation process models are discussed, in which coal, natural gas and biomass feedstocks are used for the production of two useful and cleaner fuels (H₂ and methanol). Some of the state-of-the-art methanol and H₂ production processes involve syngas production from fossil fuels, which is treated in the cleaning and water–gas shift units before the final production and purification of methanol and H₂. For instance, the natural-gas-to-methanol (NGTM) process has been extensively used in various industries for the production of both H₂ and methanol. The hydrogen-to-carbon (HCR) ratio of 2–2.2 is usually required for methanol production, which can be achieved by steam methane reforming (SMR) processes, which generates a syngas of the HCR ratio of approximately

3:1. Coal to methanol (CTM) has also emerged as an attractive technology to produce H₂ due to higher coal reserves globally, but the syngas produced from coal gasification has a drawback of offering a lower amount of HCR in the synthesis gas. This issue is resolved by using additional WGS (water–gas shift) units to enhance H₂ production by transforming the CO over the nickel-based catalyst using high-pressure steam. The research studies also showed that retrofitting already developed processes does not need major process modifications to enhance the process sustainability by reducing both the fuel consumption and GHG emissions. Wang et al. [13] and Huang et al. [14] demonstrated the pathways for producing methanol and electricity in the poly-generation process integration framework and employed WGS reactors to achieve the desired HCR in the synthesis gas. Ahmed et al. [15,16] developed the process models based on coal and natural gas feedstocks for generating hydrogen and methanol in a co-production manner. Recently, Rehfeldt et al. [17] discussed the 2030 goal for global energy resources and mentioned the use of biomasses up to 10% with the primary energy resources. Similarly, Bazzanella and Ausfelder [18] developed the process model on using biomasses as a feedstock to explore fuel switch technologies. AlNouss et al. [19] used various biomass feedstocks in the research and highlighted that the dependencies on the conventional fuels can be reduced by energy mix and fuel switch systems. Recently, Hamid et al. [20] and Ahmed et al. [21] also demonstrated a process integration framework using a combination of both gasification and reforming technologies to enhance the power and H₂ production capacities. The goal of this work is to investigate the development of standalone models for coal and biomass gasification processes followed by their integration with natural gas reforming units to enhance overall syngas production potentials. Furthermore, the heat exchanger network is developed to utilize the heat from the hot and cold streams to minimize the external energy requirements. This process integration resulted in the utilization of three different feedstocks, which reduced the reliance on a single fuel and improves the H₂ and methanol production rates. Finally, a techno-economic analysis is performed for all designs to analyze the project feasibility of the processes.

2. Development of Simulation Model

In this work, Aspen Plus V11 was used as the process simulator to develop the models for the simultaneous methanol and H₂ production. The Peng Robinson with Boston Mathias (PR–BM) equation of state was employed as the thermodynamic package for the calculation of physical properties. As the composition of fossil fuels is not consistent globally, the coal and biomass used in this study were modelled as unconventional components in Aspen Plus. Proximate, ultimate and sulfanal analyses were taken from the experimental results to specify the composition and heating values of the feedstock. Table 1 represents the modelling units of all processes developed in Aspen Plus along with their operation conditions.

Table 1. Design parameter and modelling approach.

Process Section	Simulation Unit/Conditions
Gasification	Model: RGIBBS Temperature: 1350–1370 °C Pressure: 56 bar Feed flow rate = 62.01 kg/s (coal) + 6.2 kg/s (biomass)
Reformer	Model: RGIBBS Temperature: 900 °C Pressure: 32 bar Feed flow rate: 5.5 kg/s (natural gas) H ₂ O:CH ₄ = 3:1 Ni-based catalyst

Table 1. Cont.

Process Section	Simulation Unit/Conditions
Shift Conversion (WGS)	Model: REquil Reactor 2 Adiabatic reactors Co-Mo based catalyst for sour shift Steam/CO: ~2.2 CO conversion ~99%
Air Separation Unit (ASU)	Model: HeatX, Compr O ₂ purity: 95% (vol) Energy consumption: 0.25 kW _h /kg
AGR Unit	Model: Flash, RadFrac H ₂ S Removal = 100 ppbv CO ₂ Removal = 90% Rectisol Process) Temp/Pressure = -33 °C/5.5 MPa
Methanol Reactor	RGibbs (Reactor) Cu/ZnO/Al ₂ O ₃ based catalyst Temp/Pressure = 200 °C/5.5 MPa
Heat Exchangers	Heater, HeatX, MHeatX $\Delta T_{\min} = 10\text{ °C}$

2.1. Case 1: Coal and Biomass-Based Model for the Simultaneous Methanol and Hydrogen Production (CBMH Process)

Case 1 is considered the base case model for producing methanol and hydrogen from coal and biomass feedstocks, as represented in Figure 1. The process starts with coal and biomass blending at a weight ratio of 90:10%. The blend feed for the gasifier, which is a mixture of coal and biomass, enters the entrained flow gasifier at 56 bar pressure. A controlled amount of high-purity oxygen (95 vol. %) also enters at the top of the gasifier, which partially oxidizes the feed stream to produce raw syngas. The syngas is then passed through a series of radiant and convective heat exchangers to reduce the temperature to 20 °C, which is further reduced to -35 °C in the AGR unit to remove H₂S and CO₂. The syngas is then pre-heated to 200 °C and fed to the methanol synthesis unit to produce methanol. As the conversion of syngas to methanol is not complete, the syngas is fed to the WGS unit, where it reacts with steam to convert the CO in the syngas to H₂. The mixture of H₂ and CO₂ is then fed into the CO₂ removal unit to capture CO₂, and pure H₂ is recovered and sent to the storage section.

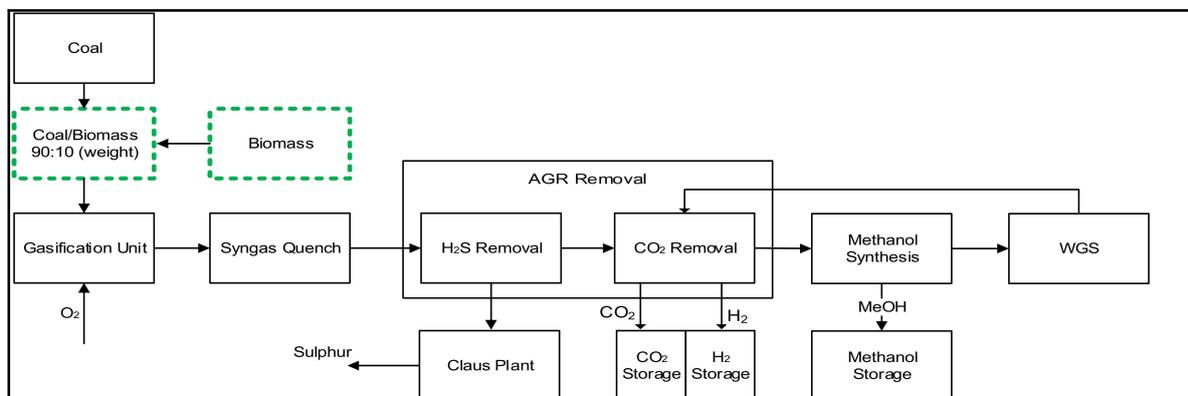


Figure 1. Process model for converting coal and biomass to methanol and hydrogen.

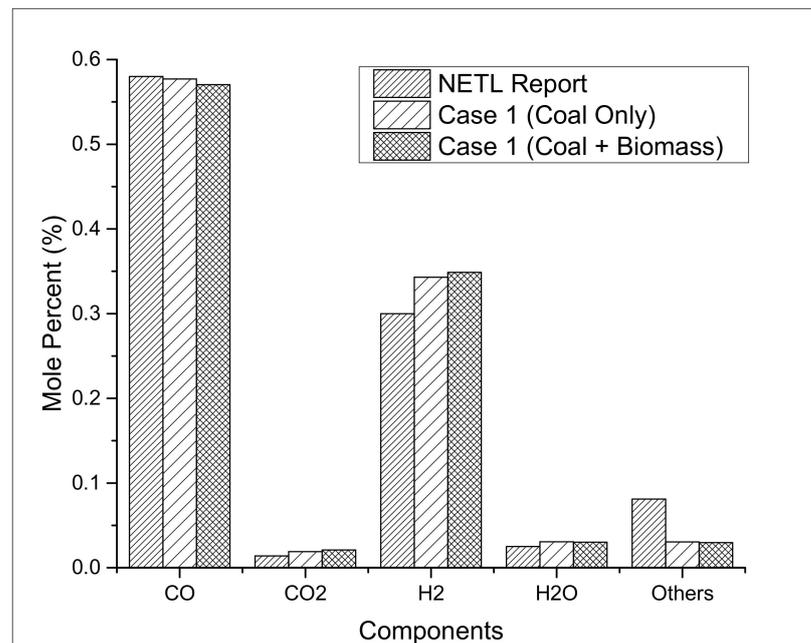


Figure 3. Validation of process models.

3.1. Equations Used for Comparative Analysis

3.1.1. Process Efficiency

The overall process efficiency for the poly-generation process is an important process performance indicator, which represents the fuel and electricity production per unit of energy consumption, as shown in Equation (1).

$$\text{Process Efficiency } (\eta_{net}) = \frac{\text{Methanol thermal energy [MWth]} + \text{H}_2\text{ thermal energy [MWth]} + \text{Electricity [MWe]}}{\text{Feed stock thermal energy [MWth]} + \text{Energy consumed [MWth]}} \quad (1)$$

3.1.2. CO₂-Specific Emission

CO₂-specific emissions represent the un-captured CO₂ from the process, as represented in Equation (2). In this study, methanol and H₂ are produced in the process. Therefore, CO₂-specific emissions give an indication of un-captured CO₂ during the production of the aforementioned fuels.

$$\text{CO}_2 \text{ specific emissions} = \frac{\text{CO}_2 \text{ Emissions } \left(\frac{\text{kmol}}{\text{hr}} \right)}{\text{Methanol Production} + \text{H}_2 \text{ Production } \left(\frac{\text{kmol}}{\text{hr}} \right)} \quad (2)$$

3.1.3. Carbon Conversion Efficiency

Carbon conversion efficiency is an important environmental metric that reflects the amount of carbon conversion from the feedstocks to the produced fuel, as represented in Equation (3). In this study, methanol is produced in the process. Therefore, carbon conversion efficiency highlights the carbon flow rates as a part of the methanol per unit feedstock flow rates containing carbon, where C_{MeOH} and $C_{\text{feedstock}}$ represents the carbon in the methanol and feedstocks, respectively.

$$\eta_C = \frac{C_{\text{MeOH}}}{C_{\text{feed stock}}} \times 100\% \quad (3)$$

3.2. Process Performance Analysis

3.2.1. Methanol and Hydrogen Production Rates

Two case studies are developed in this study for the dual production of methanol and H₂. In this study, the fuel production represents the simultaneous production of H₂ and methanol. The comparative analysis is performed between two cases to determine the fuel (H₂ and methanol) production rates, overall process performance, CO₂ emissions and carbon conversion efficiencies. The methanol production rate is calculated as 29.97 kg/s and 37.91 kg/s from the case 1 and case 2 designs, respectively. On the other hand, the hydrogen generation capacity is estimated as 4.22 kg/s and 3.87 kg/s for case 1 and case 2, respectively. The comparative analysis for both cases in terms of methanol and hydrogen flow rates are also represented in Figure 4, where, the case 2 design takes a lead in the methanol production rate between the two cases.

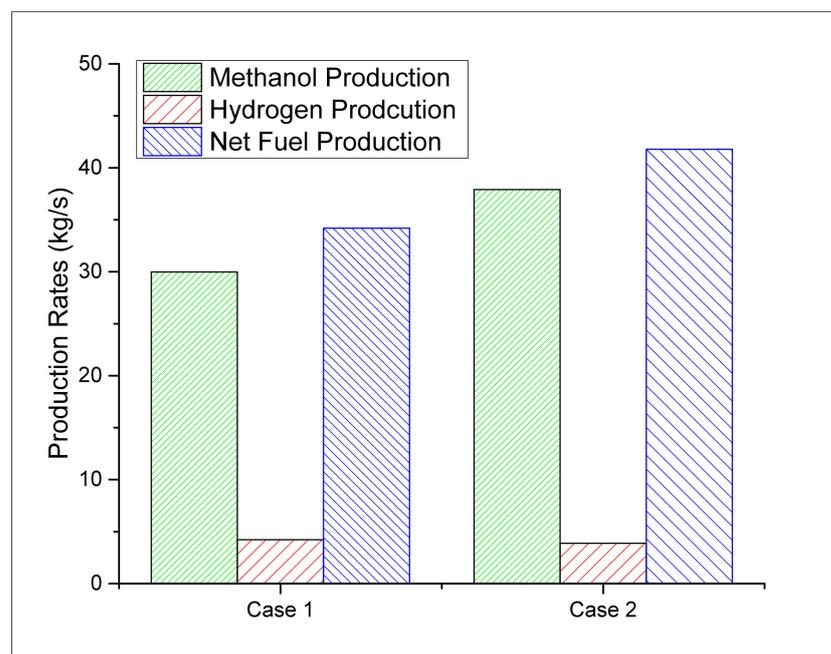


Figure 4. Hydrogen and methanol production rates.

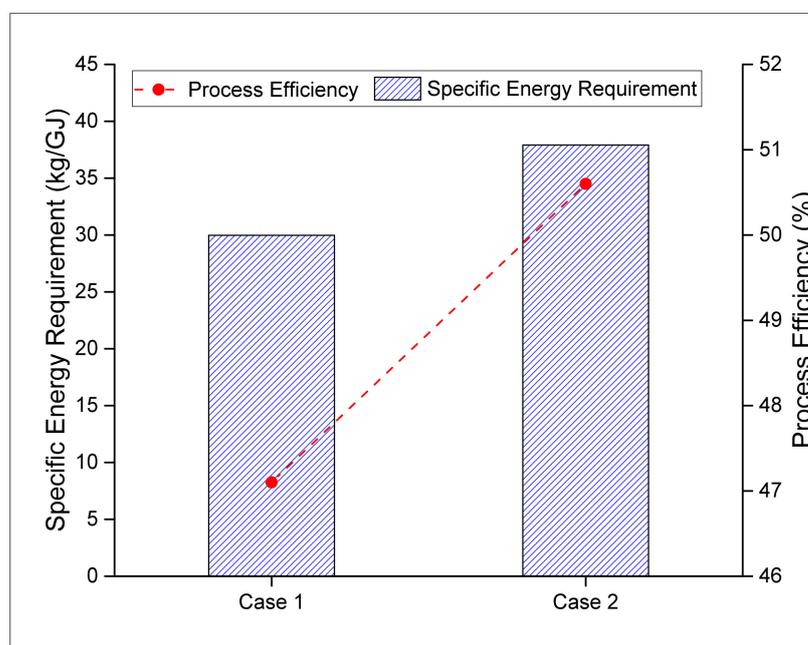
However, case 1 takes a lead in terms of H₂ production rate compared to the case 2 design. The comparative analysis between the two cases showed that the case 2 design offers 22.2% higher overall fuel production rates compared to the case 1 design. The results showed that case 2 offers higher production of the fuel (methanol and H₂) and, therefore, requires higher heating and cooling duties compared to case 1. The energy required in the syngas processing is also higher in case 2 due to larger volumes of the syngas. Table 2 provides a summary for both cases in terms of fuel production rates and other process performance indicators for comparative analysis. CO₂-specific emission is used for estimating the environmentally friendly nature of the process. The processes showing higher carbon emissions may increase the overall carbon footprint and is therefore less preferred. The comparative analysis between the two cases for the CO₂-specific emissions reveals that case 2 has 38% fewer emissions than the case 1 design.

Table 2. Comparison of case 1 and case 2 in terms of process performance.

	Units	Case 1	Case 2
CH ₃ OH Production	kg/s	29.97	37.91
H ₂ Production	kg/s	4.22	3.87
Total Fuel Produced (CH ₃ OH+H ₂)	kg/s	34.19	41.78
Energy of the Produced Fuel (H ₂ and CH ₃ OH)	kg/GJ	8.893	11.501
Electricity Produced	MWe	709	620
Heating Duty Required	MWt	1666.07	1680.49
Cooling Duty Required	MWt	280.17	303.00
Energy Integration	MWt	956.22	1142.96
Auxillary	MW	413.42	543.60
Efficiency (CH ₃ OH+H ₂ +Electricity)		47.1%	50.6%

3.2.2. Specific Energy Consumption and Process Efficiency

Specific energy consumption and process efficiency indicators were used in this study to evaluate the reliability and sustainability of the process. It can be seen from the results that case 1 and case 2 produce 8.9 kg and 11.5 kg of fuel (including both H₂ and methanol) per unit (GJ) energy consumed. By incorporating the heating values of the feedstocks and the fuel produced, the process efficiencies are calculated as 47.1% and 50.6% for case 1 and case 2, respectively. The comparison between the process efficiencies of two cases reveals that case 2 offers 3.5% higher process efficiency and 29% higher fuel production capacity than the base case (case 1) design (Figure 5).

**Figure 5.** Process efficiency and energy requirements.

3.2.3. Carbon Conversion and Emissions

The carbon conversion from feedstock to fuel (methanol in this study) and net CO₂ emissions are some of the performance indicators that are used to analyse the conversion of carbon from feedstock to product. The comparative analysis between two cases on carbon efficiency (η_c) in case 2 takes a lead of almost 4% over the case 1 design, where the actual η_c of case 1 and case 2 is calculated as 29% and 33%, respectively. Moreover, the CO₂-specific

emissions for case 1 and case 2 during the production of methanol and hydrogen on a mass basis are estimated to be 2.12 and 1.62, respectively. Figure 6 represents the comparison between CO₂-specific emissions and the carbon conversion efficiency of both cases.

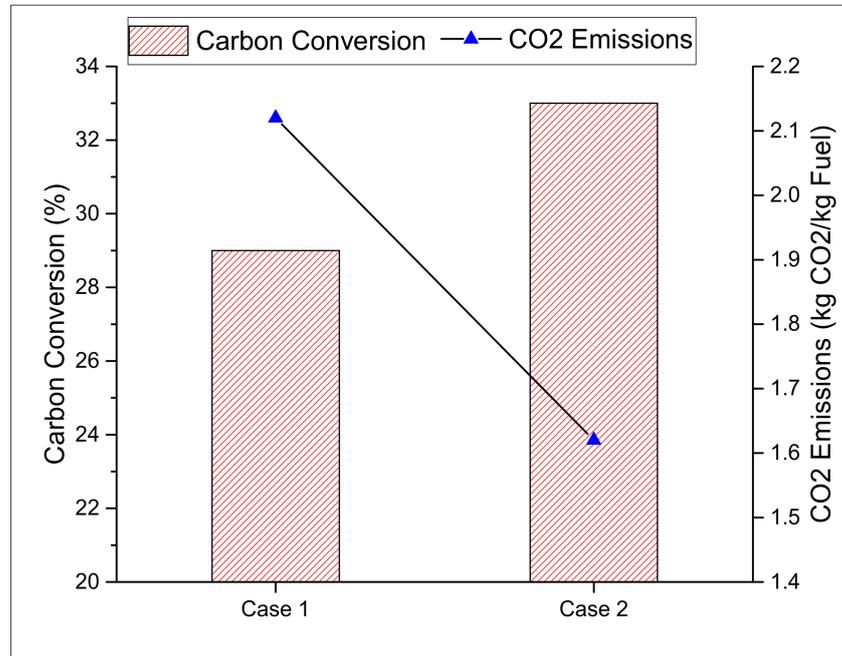


Figure 6. Comparison of carbon conversion and emissions.

3.3. Economic Analysis

To evaluate the overall sustainability of the process, the capital (CAPEX) and Operational cost (OPEX) have been estimated for both cases. All of the equipment has been sized in Aspen Plus to find the flow rate and size of the equipment. The CAPEX of all equipment is calculated using the capacity, cost of equipment and cost index information, as represented in Equation (4), where the value of x is considered 0.6.

$$\text{Cost}_{\text{New}} = \text{Cost}_{\text{Old}} \times \left(\frac{\text{Capacity}_{\text{New}}}{\text{Capacity}_{\text{Old}}} \right)^x \times \frac{\text{CEPCI}_{\text{New}}}{\text{CEPCI}_{\text{Old}}} \quad (4)$$

The results show that the CAPEX for case 1 and case 2 is estimated as 2337 M€ and 2578 M€, respectively. Due to the installation of an additional reforming unit, case 2 incurs a higher CAPEX. Moreover, the flow rates of syngas are also higher in case 2, which increases the sizes of all equipment, which also affects the overall CAPEX. Similarly, the OPEX of both cases is calculated on the basis of several assumptions including the feedstock price, catalyst cost and other utility costs. OPEX/yr represents the annual cost of running the plant and is estimated as 270 M€ and 308 M€ for case 1 and case 2, respectively. As natural gas has been used in case 2 as a third fuel, the OPEX/yr for case 2 tends to be higher, as shown in Figure 7. For a reliable analysis, the combination of CAPEX/MT and OPEX/MT during the lifetime of the plant is estimated in terms of fuel production (€/MT) for both cases. The comparative analysis showed that the CAPEX/MT and OPEX/MT required for fuel production are 403.4 M€ and 373.9 M€ for case 1 and case 2, respectively.

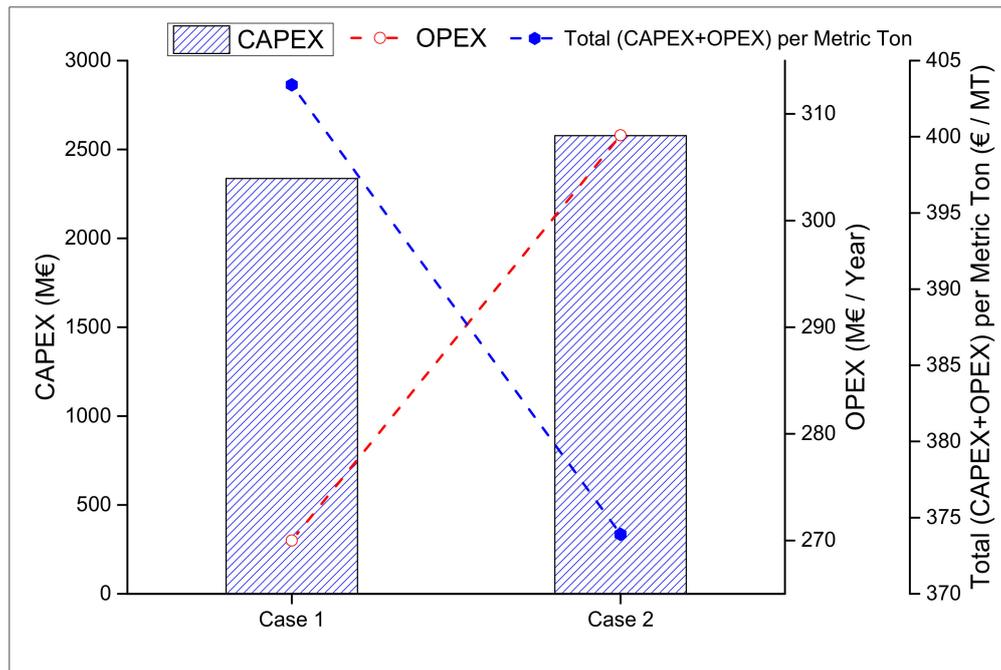


Figure 7. Comparison of capital and operational cost.

While evaluating the process economics, it has been seen that the production cost and selling price of fuels are highly influenced by the CAPEX and OPEX. Table 3 highlights the detailed results of the economic analysis for both cases. Keeping the discount rate at 10%, the selling price of fuel is calculated as 443.74 and 411.30 €/MT of fuel. The cost breakdown reveals that the methanol selling price is significantly lower for the case 2 design. On the other hand, the H₂ selling price is lower for the case 1 design. While comparing the results obtained from economic analysis, case 2 reflected a better process feasibility and offers lower fuel production prices.

Table 3. Economic analysis results for case 1 and case 2.

	Units	Case 1	Case2
Selling Fuel Price (H ₂ +CH ₃ OH)	€/MT	443.74	411.30
Selling Price of Methanol	€/MT	412.26	371.06
Selling Price of H ₂	€/MT	31.49	40.24

4. Conclusions

This study involves the development of two process models for the simultaneous production of methanol and H₂. Case 1 used the coal and biomass-based gasification process for the production of two clean fuels, namely methanol and hydrogen. On the other hand, the case 2 design integrates both the gasification and reforming techniques while using three different feedstocks, namely coal, biomass and natural gas, for the co-production of fuels, i.e., H₂ and methanol. By conducting the technical and economic analysis, it has been found that case 2 outperforms the case 1 design in terms of process efficiencies and economics. The methanol and hydrogen production rates from case 1 and case 2 are calculated as 29.97 kg/s and 4.22 kg/s, and 37.91 kg/s and 3.87 kg/s, respectively. The comparative analysis between the two cases reveals that case 2 offers 3.5% higher process efficiency. Moreover, the analysis on the process economics showed that the fuel production/selling prices in case 2 are 27.8% less compared to the case 1 design. It has been analysed from the results that the proposed integration of reforming technology with the gasification process has a potential to improve the overall process performance. Moreover,

retrofitting existing processes is also possible to incorporate the proposed design along with an efficient heat exchanger network that can improve the overall process economics of existing processes.

Author Contributions: Conceptualization, U.A.; methodology, U.A. and U.Z.; Software, U.A.; Formal analysis, H.A.; Investigation, N.A. (Nauman Ahmad); Writing—original draft preparation, U.A.; writing—review and editing, U.Z., A.G.A.J., N.A. (Nauman Ahmad), S.A.O., N.A. (Nabeel Ahmad) and H.A.; visualization, A.G.A.J. All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest: The authors declare no conflict of interest.

Nomenclature

AGR	Acid Gas Removal
CAPEX	Capital Expenditure
CBMH	Coal and Biomass to Methanol and Hydrogen
CBNMH	Coal Biomass and Natural Gas to Methanol and Hydrogen
CTM	Coal to Methanol
GHG	Global Greenhouse Gas
HCR	Hydrogen-to-Carbon Ratio
IPCC	Intergovernmental Panel on Climate Change
NGTM	Natural Gas to Methanol
OPEX	Operational Expenditure
SMR	Steam Methane Reforming
WGS	Water–Gas Shift

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