

Process Design and Techno-economic Analysis of Dual Hydrogen and Methanol Production from Plastics Using Energy Integrated System

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Abstract

This study has been dedicated towards the conversion of plastics to methanol and hydrogen. The base design (case 1) represents the conventional design for producing syngas via steam gasification of waste plastics followed by CO₂ and H₂S removal. The syngas is then processed in the methanol synthesis reactor to produce methanol, whereas, the remaining unconverted gases are processed in water gas shift reactors to produce hydrogen. On the other hand, an alternative design (case 2) has been also developed with an aim to increase the H₂ and methanol production, which integrates the

plastic gasification and the methane reforming units to utilize the high energy stream from gasification unit to heat up the feed stream of reforming unit. Both the cases have been techno-economically compared to evaluate the process feasibility. The comparative analysis revealed that case 2 outperforms the case 1 in terms of both process efficiency and economics.

Keywords: Gasification; SMR; Waste Plastic; Methanol production; Technical Analysis, Economic Analysis.

1. Introduction

The global demand for energy has considerably increased during the last few decades due to fast industrialization, an increase in population, and transportation. According to Global Energy Outlook, the forecasted increase in the energy demand for the next two decades is 30 % [1]. Nevertheless, conventional processes produce a significant amount of carbon dioxide (CO₂) emissions. Therefore, increasing the utilization of these conventional processes enhances the CO₂ emissions by up to 415 ppm which would drastically affect the human health, causes climate change and global warming. The Paris Climate Change Conference stated that the global rise in temperature should be maintained below 2 °C till 2050 [2]. It could be achieved by enhancing the efficiency of the processes and by developing alternative processes that could limit CO₂ emissions [3]. Consequently, development of alternative processes to produce energy with a control level of CO₂ emissions is crucial. One of the alternative way is to utilize the plastic waste to produce energy and valuable chemical commodities.

Globally, 6.3 billion tons of plastics were produced in the period between 1950 and 2018, however, only 9% was recycled, and 12% was burnt [4]. This huge amount of plastics could be utilized for energy production, especially with the huge increase in energy demand. Additionally, plastic

wastes critically affects the environment, ecosystem, and human life [5]. The annual disposal of plastic waste to ocean was estimated to be 4.8-12.7 million metric tons, which has severe impacts on marine life [6]. Therefore, several countries establish a regulation policy to limit those impacts [7]. Hence, efficient recycling of the plastic waste will ultimately reduce pollution and contribute in meeting the substantial increase in energy demand [8]. One of the promising recycling techniques that produce clean energy from waste plastics is thermochemical recycling.

Thermal recycling of plastics has an advantage over the other mechanical and catalytic techniques because it yields high gaseous products with no char generation thereby avoiding further cleaning up [9–12]. There are several sectors in thermochemical recycling such as pyrolysis, thermal liquefaction, hydrocracking, and gasification [13–16]. Among these thermochemical conversion processes, pyrolysis is a cracking process which occurs in the absence of O₂ to produce liquid hydrocarbons. The syngas composition from pyrolysis should be managed for the corresponding applications. Another sector in thermochemical conversion is hydrocracking which is a catalytic process that produces chemical fractions in the range of light naphtha to heavy diesel. It requires a bi-functional selective catalyst to facilitate the process. The product's properties depend on the type of catalysts and the operational conditions. Additionally, it requires hydrogen as part of the feed. Similarly, the thermal liquefaction process uses a different solvents (water or organic solvents) to produce high yield of liquid hydrocarbons at relatively low temperatures. Among all the thermochemical conversion processes, gasification (steam, oxygen, or air gasification) is one of the main process that generates a high amount of syngas (with traces of other gases depending on the feedstock contents) as a product. The process is usually conducted at very high temperatures (i.e., 800 -1500 °C) [17]. The process is feasible, and it has less complexity in terms of design and laboratory studies. The syngas could be further processed to produce pure hydrogen or alcohols

and other valuable fuels and chemicals via Fischer-Tropsch technology [18]. Hydrogen and methanol are the most essential products because they are considered clean fuels and they have several industrial applications [19].

Hydrogen has several appealing characteristics such as high energy conversion, renewable production, zero emissions, reliability, wide sources, and large storage capacity [20]. It is considered the cleanest fuel in this century. Hydrogen receives great global attention as it is a raw material for wide industrial processes such as ammonia synthesis, water gas shift, and petroleum refinery. It is reported that the 96% of hydrogen main production pathway is from traditional fossil sources (48 % from natural gas, 30% from naphtha reforming, and 18% from coal gasification) [21]. Therefore, investigating the other alternatives (such as waste utilization) to produce hydrogen is essential and need of time. Similarly, methanol is also an important chemical with a wide range of applications in manufacturing industries. It is used as a raw material to produce dimethyl ether, formaldehyde, acetic acid, and MTBE [22]. Additionally, there is a growing trend for the utilization of methanol to be blended with gasoline to enhance the fuel octane number and other properties. It is also used as a direct fuel in methanol fuel cells [23]. Moreover, it is utilized in the synthesis of olefins and polymers via MTO, and then polymerization processes [24]. Thus, hydrogen and methanol are very vital chemicals, and they need to have to be abundant to meet industrial growth via alternative technologies.

There are limited studies on the thermal conversion of plastic wastes to hydrogen and methanol. Acomb et al. [25] studied the production of hydrogen from plastics taken from electronic waste equipment via pyrolysis-gasification in two different stages. The plastics were first pyrolyzed in the first stage at 500 °C, and then they were fed to the second stage (i.e., steam gasification reactor) at 800 °C. It was reported that increasing the steam-to-plastic ratio enhances hydrogen production.

Additionally, using two stages (pyrolysis and then gasification) enhances the hydrogen production from plastic electronic wastes. Another study on the production of hydrogen from several plastics (polypropylene, polystyrene, and high-density polyethylene) as feedstocks was conducted by Chunfei and Williams using the two thermo-chemical processes stage wise [26]. Higher gaseous yields were perceived for HDPE and PP compared to PS (53.5 %, 59.6 %, and 11.2 %, respectively). Namioka et al. [27] studied the production of hydrogen-rich syngas from waste plastic (polystyrene) via pyrolysis, then steam reforming. The results showed the optimal gas production (60 g/h) with minimum coke fractions achieved at a pyrolyzer temperature of 400 °C along with a steam reforming temperature of 630 °C. Takafumi et al. [28], conducted a study on producing methanol via partial oxidation of waste plastic (polyvinyl chloride). Demonstration plant feeding 5 tons of plastic per day was developed; producing syngas via entrained single-stage reactor. Then, methanol was synthesized via liquid-phase catalytic process. The successful production of methanol(98% purity) with conversion and gas cold efficiency of 96% and 63% respectively was achieved. In general, there is a lack of detailed information on the conversion of plastics to hydrogen and methanol. Only few studies are available on the dual production of hydrogen and methanol from coal and biomass. A study conducted by Usama et al. [29] using Aspen Plus, in the coproduction of hydrogen and methanol using coal-biomass mixture. A novel design was developed which incorporate the steam methane reforming process with the coal-biomass gasification process. The incorporated design reduced the specific energy requirement by 29% and enhanced the process efficiency by 3.5%. The techno economic evaluation was performed for the two designs and perceived a 7.3% reduction in the total fuel cost.

Another dual study was performed by Usama Ahmed [30] on the production of hydrogen and methanol from coal in two cases. The base case was the normal gasification process to produce

syngas, then the syngas was catalytically converted to methanol and the remaining gases were processed via water gas shift to produce hydrogen. On the other hand, the alternative case was an integrating design with the steam methane reforming process (SMR) to utilize the high energy outlet stream from the gasifier. The results revealed that the alternative case had higher process efficiency by 4.28% with an 18.3 % reduction in the specific energy requirement. Additionally, the alternative case offered a reduction in fuel production cost by 13%. Moreover, the specific CO₂ emissions were lowered in the alternative case by 34.3%. These studies reported the feasibility and efficiency of the co-production of hydrogen and methanol from a carbon-based feedstock. However, the dual production system from plastics has not been investigated.

Therefore, the main target of this study is to develop two dual process designs for producing hydrogen and methanol from plastics. The first design is plastic gasification and the second one is the integrated design with reforming technology. The two designs were evaluated and analyzed in terms of energy consumption and economic feasibility. The main innovative part of this study is the design of an alternative case that produces hydrogen and methanol utilizing the energy from the gasifier to heat the inlet streams to the SMR process. The syngas from the two processes were mixed which contributed in the enhancement of stoichiometric ratio (SN), and the H₂/CO ratio of the syngas subsequently it also boosted the methanol yield.

2. Modelling and simulation approach

In this study, Aspen Plus ® V12 was used as a simulation tool, selecting Peng Roberson (PR) as a suitable property package. It is commonly indorsed for gas systems, and it has the capability to deal with the wide-range of pressure and temperature [31]. Additionally, it also has been used by several gasifications and reforming studies [32–34]. The feedstock composition of the waste

plastics; Polyethylene (PE) and Polypropylene (PP) is given in Table 1. PE and PP have the highest global consumption among the different plastic types (33.5% and 19.5% respectively) [35]. Their heating value is high when compared with other types. Therefore, PE and PP have been selected in this study. The HCOALGEN models in the Aspen Plus were specified as the plastics heating value. The plastics were first decomposed in two separate decomposition reactors at a temperature of 650°C. Then, through the steam gasification reactor, syngas was produced containing CO, H₂, CO₂, H₂O, CH₄, and traces of H₂S. RYield reactor was used to represent the decomposition step (the yield was the ultimate analysis). Then, RGibbs reactor was selected, representing the steam gasification of the plastics. The gasifier temperature and pressure were selected based on sensitivity analysis seeking the optimal hydrogen and methanol yields. The H₂S drastically affects the methanol catalyst. Therefore, those traces were removed via a methanol absorption unit. The outlet gases were fed to the methanol synthesis unit controlling the inlet operational conditions. Three reactions are involved in methanol synthesis which are CO hydrogenation, CO₂ hydrogenation, and water gas shift. Using a recycle loop, methanol production was maximized. The unconverted gases were separated and introduced to WGS to convert the remaining CO to hydrogen and CO₂ using two REquil reactors connected in series. After that, the CO₂ was removed in AGR (acid gas removal), producing pure hydrogen with some methane. The alternative case was developed in such a way that the high energy available at the outlet of the gasifier was utilized to facilitate the SMR process. In this model, the production of syngas from the reforming process and gasification process was simultaneous. The chemical reactions for methanol synthesis, WGS, and SMR are given in Table S1. The plastic feedstock, natural gas and the steam for gasification and reforming were specified on the basis of selectivity analysis to ensure the syngas high heating value with proper utilization of energy.

2.1 Design assumptions

Certain assumptions had been made to pursue the plastics conversion to hydrogen and methanol. Table 2 illustrates the major assumptions which were taken for the processes. The gasification model was constructed based on Saebea et al.[36]. The blend of PE and PP were designated via sensitivity analysis for the plastic gasification. Additionally, the inlet flow rate of the plastic feedstock was assumed to be 100 kg/h. The steam to plastic ratio for the gasification process was set to be 1.25:1 (mass basis), which was based on the sensitivity analysis performed in previously published work [37]. The natural gas to the plastic ratio (in mass basis) was set to 1.5 to achieve maximum heat utilization. The ratio of steam to natural gas was set to be 1.6:1 (mass basis) to maximize the H₂/CO from the SMR process based on a sensitivity analysis conducted in this study. The temperature and pressure of the gasifier were selected such that the heating value of produced syngas is maximal, and the H₂/CO ratio is maximized along with the SN ratio [37]. Nickel-based catalyst was selected for the reformer unit because it ensures the high activity and low cost [38]. γ -Al₂O₃- α -Ga₂O₃ was selected as a catalyst for the WGS process tested by Leon et al. [30], this catalyst can be activated at atmospheric pressure. Rectisol process has been used for the AGR unit. The operational conditions were validated with the literature [40].

2.2 Governing equations for technical analysis

To evaluate the developed models, some parameters were evaluated for technical and economic analysis. Table S2 represents some of the formulas used in evaluating the developed designs. HPF is the mass flow rate of produced hydrogen per total feedstock. The hydrogen and methanol thermal energy was calculated from the lower heating value multiplied by the corresponding mass flowrate. The process efficiency and the CO₂ specific emission were evaluated according to Usama Ahmed [30]. The total investment cost (TIC) demonstrates the capital cost divided by the fuel

production rate. To calculate the capital investment cost, we used historical related data modified by Chemical Engineering Plant Cost Index (CEPCI). The exponent x is between 0 and 1, and its value was selected to be 0.6. The total fuel cost of €/kg represents the production cost of the fuel (hydrogen and methanol) in the dual production system. The SN ratio gives an indication of the H_2/CO_2 content in the mixture [41]. SN should be around 2 as a condition for methanol synthesis, and as it is larger, the methanol yield is expected to be higher as well [42].

2.3 Kinetics of Methanol synthesis

The methanol synthesis operational conditions and kinetics were based on Graaf et.al (1989) [43]. The catalyst is Cu/Zn/Al with temperature and pressure ranges of 210-290 °C and 15-50 bars respectively [44]. The temperature and pressure of methanol reactor (PFR) was 270 °C and 25 bars correspondingly. This process consists of three main parts (i.e., reactions); hydrogenation of CO, hydrogenation CO_2 , and reverse gas shift. The reactions and their kinetics were provided in Table S3. The reaction constants were provided in Table S4. The reaction equilibrium constants were introduced and given by Graaf et.al (1986) [45]. K_{P2} is the multiplication of K_{P1} and K_{P3} , because the CO_2 hydrogenation reaction is the sum of CO hydrogenation reaction and RWGS reactions.

The kinetics were introduced to the Aspen Plus matching the software interphase. The reaction was selected to be LHHW (Langmuir-Hinshelwood-Hougen-Watson). Three existing kinetic sections in the software were filled (i.e., kinetic factor, driving force and adsorption expression).

The state of the process was selected to be vapor with a packed bed catalyst. The software approach in keying the kinetic constants is based on the equation 1.

$$\ln (K) = A + \frac{B}{T} + C \ln (T) + D T \quad (1)$$

Therefore, the reaction rate constants, adsorption equilibrium constants, and reaction equilibrium constants were presented using equation (2), equation (3), and equation (4) respectively.

$$\ln (K) = \ln(A) + \frac{B/R}{T} \quad (2)$$

$$\ln (K_P) = \frac{B}{\text{Log}_{10}(e)} - \frac{A}{\text{Log}_{10}(e) T} \quad (3)$$

$$K_{P2} = K_{P1} \times K_{P3} \quad (4)$$

The kinetic factor (k) is just A , and $E = -B$. The other constants were evaluated based on the above equations and were summarized in Tables S5 and S6.

3 Development and validation of case Studies

3.1 Case 1 (Base case)

The hydrogen and methanol were successfully produced from plastics wastes (i.e., PE and PP) in five main processes. The first process is the decomposing of solid plastics at high temperature (i.e., 650°C). Then, on the gasifier at a higher temperature (1500°C) [46] and moderate pressure 25 bars, syngas and other gases were produced with H₂/ CO molar ratio of 1.87. The outlet gaseous mixture was introduced to H₂S removal unit after cooling using lean as an absorbent. The operational conditions of the absorber were 50°C and 25 bars. The amount of H₂S was reduced to 1 ppm not to affect the methanol synthesis catalyst [47]. The inlet temperature and pressure of the methanol synthesis reactor was specified to be 270 °C and 25 bars respectively, meeting the experimental methanol synthesis operational conditions [48]. The methanol catalyst characteristics and the inlet specifications were based on a study conducted by Portha et al. [49], which extensively studied Graaf. et. al kinetics to synthesize methanol. Although, the methanol reactions are exothermic,

however, increasing the temperature enhanced the kinetic constants which was superior to the reduction in the equilibrium constants. Additionally, the process was performed at a relatively moderate pressure based on sensitivity analysis conducted on the gasifier conditions, methanol and hydrogen production. The unrecovered gases in the methanol synthesis process were fed to the WGS unit maximizing the hydrogen production, and then via CO₂ removal unit, pure hydrogen was obtained. Figure 1 shows the process flow diagram for base case 1. The process flow diagram is also provided in the supplementary file Figure S1.

The design was validated based on syngas composition evaluation methanol yield. Saebea et al. [36] studied the conversion of PP and PE to syngas. They successfully produced syngas from the plastic blend (i.e., PE and PP) using Aspen plus software. The syngas composition from PE and PP were validated based on two different experimental studies [50][51] at a temperature of 900°C and steam to plastic ratio of 1.5. Due to design constrains, the gasifier temperature was raised to 1500 °C based on a simultaneous investigation of the effects of temperature and pressure of syngas composition. Figure 3 represents the syngas composition from plastic gasification for PE and pure PP compared with experimental references. The design results are highly consistent with experimental based results. Therefore, the model was further developed toward hydrogen and methanol production. The methanol synthesis results were in the correspondence to the results recorded by Graaf et al. [43].

3.2 Case 2 (Alternative Case)

The alternative model (Case 2) is basically, the integration of SMR and plastic gasification to boost the syngas heating value and exchange the heat of the outlet gasification stream with the SMR feed stream. The process block flow diagram is given in Figure 2, where the process flow diagram is given as Figure S2 in the supplementary file. The SMR feed flowrate was defined such that the

exchanged heat will be sufficient. The process scheme for SMR was taken from Hamid et al. [52]. The steam was mixed with the natural gas containing mainly methane. The steam to methane mass ratio was set to be 1.60. The RStoic and RGibbs reactors were used as a reforming reactor with temperature of 894 °C and 1200°C respectively. The RStoic reactor was simulated at atmospheric pressure, where the RGibbs reactor pressure was 25 bars. These operational conditions are in match with the literature [53], and they were selected such that the SN ratio is maximized.

The hydrogen to carbon monoxide produced from SMR was in good agreement with the literature [54]. The H₂S was removed from the plastic gasification and then the outlet gases were mixed with the syngas produced by SMR. Then, the syngas was fed to the methanol synthesis unit with the appropriate conditions. The methanol was successfully produced, and the unconverted gases were mainly hydrogen (94% molar purity) with traces of methane. Since the CO and CO₂ were converted completely with the aid of the recycling loop, no further processing of the gas was made. This result was achieved by performing extensive sensitivity analysis to control the feed specifications enhancing the SN to 2.51. The validation of the methanol synthesis unit was performed comparing the two designed cases with two previous studies [44,55]. The key parameter in the comparison was the methanol yield which was defined as the mass production rate of methanol per syngas molar flow rate. The comparison is given in Figure 4.

4 Result and discussion

Two cases were modeled for dual hydrogen and methanol production. Sensitivity analysis was performed for the parameters to test in case 1 and case 2 to properly select the optimal design conditions. To perceive the expected enhancement in the process efficiency, technical and

economic analysis were conducted. The most reprehensive parameters are the syngas heating value, overall process efficiency, CO₂'s specific emissions, and fuel production cost.

4.1 Sensitivity analysis

The main paramours in the two cases that ultimately impact the fuel production and energy consumption are the gasifier operational conditions and the reforming feed flow rate. Those parameters were tested and evaluated seeking the optimal values.

4.1.1 Impact of gasifier temperature and pressure on syngas composition

The impacts of plastic gasifier pressure and the temperature were investigated on the syngas heating value represented by H₂/CO and SN ratio. Figure 5 shows the effect of gasifier temperature and pressure on the SN and H₂/CO ratio. Increasing the gasifier temperature enhances the SN and H₂/CO. However, increasing the gasifier pressure decreases the SN and H₂/CO ratio. The high temperature promotes higher hydrogen content. The results can be explained by Le Chatelier's principle. Increasing the temperature will lead to promoting the endothermic relations in the gasifier towards the product. However, increasing the pressure will promote methane production over hydrogen and carbon monoxide production. Harris et al. [56], performed a study on the effect of gasifier temperature on the produced gaseous concentrations, and they found that, as the gasifier temperature raised, CO₂ concentration reduced. Moreover, the concentration of CO and hydrogen increased. According to Marek et al. [57], increasing the pressure reduces the hydrogen concentration. Our findings are in good agreement with the two experimental studies.

4.1.2 Impact of total SMR flowrate on hydrogen and methanol production

Case 2 contains two main processes that produce syngas. Therefore, the ratio of reforming feed flowrate to the plastic flowrate is a very significant factor affecting the whole design in terms of

energy utilization and syngas heating value. The plastic mass flow rate was constant with a value of 100 kg/h. The SMR flowrate was varied testing the impact on syngas heating value and methanol production. The steam to natural gas ratio was maintained to be 1.6:1. The syngas flow rate at the inlet to methanol process was recorded at each reforming feed flowrate. Apparently, increasing the SMR flowrate would increase the syngas production, however, the syngas heating value will be different. Figure 6 represents the effect of SMR flowrate on hydrogen and methanol production, and on the purity of hydrogen. Increasing the reforming flow rate enhances the hydrogen and methanol production. The reforming flowrate produces higher syngas heating value, therefore, higher hydrogen and methanol from the syngas were produced. The model is restricted by the heat integration between plastic gasification and reforming; therefore, 390 kg/h was selected as the total reforming flowrate.

4.2 Stream composition and energy evaluation

The energy analysis for the designs is essential to grasp the effect of using the novel integrated design on the overall process performance. Table 3 represents the operational conditions related to the units for case 1 and case 2. The steam to plastic ratio was set to 1.25 and it was confirmed via sensitivity analysis. The operating temperature was set to 300 °C with atmospheric pressure. Steam and plastics were mixed in the gasifier at high temperature and pressure. The outlet composition from the gasifier had a H₂/CO ratio of 1.87 with SN of 1.86. The CO₂ production was extremely lower than the carbon monoxide. The SMR operated at 25 bars and produced 389 kg/h of syngas which was 63.4% of the total produced syngas. It produced higher H₂/CO and SN ratios when compared to the plastic gasification by 69.9% and 57.6% respectively. Then the two syngas streams were mixed for case 2 which resulted in the enhancement of syngas heating value. After that, the syngas was cleaned from H₂S via H₂S removal unit. The composition was slightly changed

because H_2S was removed with some traces of CO_2 . In methanol synthesis unit, methanol was produced successfully with very high purity in both cases. Case 2 had higher methanol purity than case 1 by 0.83%. The case 2 directly produces hydrogen, therefore, the WGS and CO_2 removal units were not required. The WGS unit increased the hydrogen mole composition for case 1 to 39% with a CO_2 content of 39%. Therefore, AGR unit was required to remove the CO_2 . The produced hydrogen in case 1 was at the outlet of CO_2 removal unit with a rate of 6.15 kg/h. The purity of hydrogen was low because of the remaining methane from the plastic gasification unit. The alternative case produces higher hydrogen rate than case 1 with a higher purity than the base case by 49%. Therefore, the results clearly stated the superior performance of case 2 over case 1. Table 4 shows the essential evaluated parameters representing the performance of the two cases. The results revealed a dominance of case 2 over case 1 in terms of energy evaluation. The heating value of the total syngas represented by H_2/CO and SN ratios, for case 2 is greater than case 1 by 2.65 MJ/kg. The production rate of hydrogen per total feedstock in case 2 is greater than case 1 by 4.04 (i.e., 66%) with a higher H_2 purity of 30%. The methanol production rate for the two cases were also evaluated, and the result revealed an enhancement in case 2 by 2.8%. The purity of the methanol is slightly higher in case 2 as compared to case 1. The syngas conversion to methanol in the alternative case was higher than the base case by 1.84%. A terminology that represents the energy performance is the process efficiency which was improved by 3% for case 2. Hence, the alternative case showed a significant improvement in the fuel production rate and energy performance.

4.3 Overall process performance

A more illustration of the process performance is given in this section, which includes the overall process efficiency, and the normalized methanol production rate. The alternative case has higher

process efficiency than case 1 with a 3% enhancement. This improvement was attributed to the energy integration between plastic gasification and SMR, which results in strong reduction in energy consumption. The process efficiency was calculated after performing heat integration for each model. Additionally, with the alternative model, the production rate and fuel heating value was significantly increased. Figure 7 demonstrates the overall process efficiency with the methanol final production rate for the two developed cases. The normalized methanol production rate was higher in case 2 than the case 1 by 2.81%. The SN for the inlet syngas in case 2 was 2.51 which improved the methanol production [58]. In terms of fuel production, fuel heating value, and energy reduction, case 2 was superior to case 1. Therefore, this new integrated design made the thermal recycling of plastic waste more improved, unique, more feasible and convenient. The economic analysis is crucial to study the applicability of the two cases in the industry.

4.4 CO₂ specific emissions

Owning to GHG emissions for the environmental obligations, the control of CO₂ emissions are very essential in any chemical process. Therefore, extensive sensitivity analysis was performed to ensure the lowest CO₂ emissions in the both cases. The alternative case was found to be very efficient in the reducing the CO₂ emissions. Figure 8 shows the CO₂ specific emissions and the hydrogen production rate per total feedstock (i.e., plastics and natural gas). The hydrogen production rate for the alternative case was higher by 66.7%. Therefore, with this new alternative design, a very limited level of CO₂ emissions with significant enhancement in fuel production rate was achieved.

5 Economic Analysis

To perform the economic analysis for the both cases, the information available in literature was the source for the initial plant cost estimation. The fixed capital investment was evaluated based on the individual cost estimation of the processes based on the process capacity and CEPCI. The power law was the main equation for estimating the fixed capital expenditure (CAPEX). Certain assumptions were taken in account for estimating the operating expenditures. The maintenance, working capital, land and salvage were estimated based on the fixed expenditures. The labor cost was calculated based on the number of nonparticulate processing steps. Then, the administration cost was evaluated based on labor cost. Table S7 represents the major assumptions taken for the economic analysis. The exponent x for the power law was estimated to be 0.6. The utility cost was calculated for each equipment based on the corresponding duty. The reference for that calculation was Donald E. Garrett book [59]. The taxation rate was considered and selected to be 15%. To construct the plant cash flow diagram, plant life of 30 years was selected considering three daily shifts with a stream factor of 0.95. The assumptions were in good match with previous studies in the literature [57] [81].

5.1. Estimation of CAPEX and OPEX

The two main expenditures are the fixed capital expenditure (CAPEX) and the operational expenditure (OPEX). There are several factors such as process efficiency, operational time, the raw materials, and the plant capacity which affect these two expenditures. All of these factors were selected for study and illustrated in the previous sections. Equipment and installation costs were deduced from previous studies. The gasification equipment and installation costs were based on a study performed on entrained gasifier [62]. The methanol synthesis cost estimation was based on Graaf et al. kinetics in estimating the methanol synthesis price [63]. The estimation of the other

units was according to Usama et al. [52]. The order of magnitude was the approach in calculating the CAPEX based on previous cost estimations for each unit. Table S8 represents the details of the CAPEX calculation. The economic results of case 1 and case 2 were presented in Table 5, which were calculated by applying the assumptions and formulas from Table S2. The results consist of the main parameters for the CAPEX and OPEX. The two cases have different costs due to the difference in the capacity and feedstock compositions. Case 1 has no SMR, however, the case 2 has no WGS. The total investment cost for case 2 is higher than case 1 by 13.4%. It was attributed due to the high amount of feedstock utilization in case 2 as compared to case 1. Nevertheless, the TIC for case 2 is lower than case 1 by 72.5%. Hence, case 2 is more efficient than case 1 in terms of capital expenditures.

The operating expenditures are divided into fixed and variable OPEX. The fixed OPEX includes labor, maintenance, and administrative costs. The variable OPEX involves the catalysts expenses, the feedstock price, the solvent price, and waste disposal cost [59]. The total fixed manufacturing cost for case 1 and case 2 were 0.504 and 0.526 M€/Year respectively. The total OPEX in M€/Year for case 1, and case 2 were 0.839 and 1.0322, respectively. Case 2 had higher OPEX than case 1 by 23%. The alternative case requires higher operating expenses due to high plant capacity. A proper representation parameter for OPEX is to normalize the total operating cost by the production rate of the fuel. The alternative case cost in M€ per ton of fuel was lower than case 1 by 70%. It proves the superiority of case 2 over case 1 in terms of reducing the operating expenditures.

5.2. Comparison of fuel costs with the literature

This section demonstrates the fuel production cost (hydrogen and methanol) and total investment cost per fuel production rate. Additionally, the cash flow diagram was constructed for base and

alternative cases. Then, the fuel production cost was validated and compared with the literature. Figure 9 represents the TIC in M€/year for the two cases, and the fuel production cost in €/kg. The dual production cost of methanol and hydrogen was lowered in case 2 as compared to case 1 by 0.277 €/kg.

To evaluate and construct the cash flow diagram, the initial investment cost was equally divided for three years. The revenue was calculated based on the production rate of hydrogen and methanol multiplied by the corresponding average price of hydrogen and methanol. The depreciation was considered and assumed to be 10% of the total investment cost. Figure 10 shows the cash flow diagram for case 1 and case 2. Clearly, the cash flow for case 2 is much more than case 1. The revenue was greater in case 2 than case 1 by 3.526 M€/year. The net present value (NPV) for case 1 and case 2 was evaluated to be 2.734 and 26.201 M€ respectively. The present value ratio (PVR) was also calculated for the two cases which were 1.614 and 5.301 respectively. Those calculations clearly indicate the significant improvement in the economic efficiency of the alternative case than case 1.

To have a good understanding of the cost analysis of the dual hydrogen and methanol production models, Table 6 is designed to illustrate several levelized fuel costs from the literature [37,64–66]. The results from Table 6 showed that, the hydrogen production cost is significantly higher than the methanol cost. However, when producing hydrogen and methanol simultaneously, the fuel production cost is relatively low. Additionally, case 2 has the lowest fuel production cost when compared to the other studies that produce standalone methanol or dual hydrogen and methanol. Plastics have relatively high hydrogen content and when plastic gasification process was integrated with reforming process, the fuel production cost was significantly reduced. The two cases were also compared with a study in the literature producing hydrogen and methanol from coal [67]. The

study utilizing the coal produced hydrogen and methanol from the conventional and integrated design with the costs of 0.28 €/kg and 0.23 €/kg respectively. Figure 11 shows the comparison between this study and our study in terms of levelized hydrogen production cost. To conclude, integrating plastic gasification with SMR results in a significant enhancement in the process efficiency and appreciable reduction in fuel production cost. The study has a great potential for commercialization in the industry, based on the technical and economic analysis. However, further investigation should be performed to perfectly settle down the process conditions. The cost estimation could be improved by following a preliminary estimate approach, which will give a more appropriate cost estimation. The study supports the environment via plastic recycling, and it offers an alternative feasible energy source.

Conclusion

The simultaneous production of hydrogen and methanol from plastics has been accomplished. Two cases were developed: the classical design (case 1) and the integrated design (case 2). Sensitivity analysis was performed to ensure the optimal design conditions. Gasifier pressure and temperature were tested seeking their effects on the syngas heating value and stoichiometric number. The impact of SMR total flowrate on the methanol, and hydrogen production rate was also investigated. Additionally, the two cases were evaluated in terms of syngas heating value, production rate, fuel purity, energy consumption, cost analysis, CO₂ specific emissions and the results are summarized as follows:

5.1 The syngas's H₂/CO for case 1 and case 2 was calculated as 1.87 and 2.6, respectively.

- 5.2 The stoichiometric number (SN) for the base and alternative case is found to be 1.87, and 2.51 respectively. Thus, the net syngas heating value is calculated for case 1 and case 2 as 23.46, and 26.11 MJ/kg, respectively.
- 5.3 The produced hydrogen in case 2 was found to be higher than case 1 by 66.7 %. The production of methanol per feedstocks for case 1 and case 2 are 2.12 and 2.18, respectively.
- 5.4 The process efficiency of case 1 and case 2 was calculated as 73% and 76% respectively.
- 5.5 The total investment cost per fuel ton for case 1 and case 2 was found to be 0.433 and 0.119 million Euro/ton respectively.
- 5.6 The total operating cost per fuel ton for case 1 and case 2 was calculated as 16.4 and 4.9 thousand Euro/ton correspondingly.
- 5.7 The fuel production cost per fuel production rate was estimated to be 0.53 and 0.26 for case 1 and case 2, correspondingly.

The whole analysis ensured the effectiveness and promptness of the integrated case over the classical one. The economic analysis ensures the feasibility of plastic conversion to hydrogen and methanol. The integrated novel design should be implemented because it saves the environment from the negative impacts of plastic waste and offers alternative energy sources. The process promotes the energy consumption and enhances the fuel quality (hydrogen and methanol). The two fuels are the cleanest fuels among others which also will attribute in saving the environment. It is suggested that further investigation should be conducted for this process by testing the blends of different plastics and other carbonaceous solid waste.

Nomenclature

PE:	Polyethylene
PP:	Polypropylene

SMR:	Steam Methane Reforming
GHG:	Greenhouse gases
LHV:	Lower Heating Value
WGS:	Water gas shift
CAPEX:	Capital Expenditures
OPEX:	Operating Expenditures
SN:	Stoichiometric Number
PR:	Peng Roberson
ER:	Equivalence ratio
CSE:	Specific CO ₂ Emissions
HPF:	Hydrogen Production Rate per Feedstock Rate
HHV:	Higher Heating Value
MeOH:	Methanol
AGR:	Acid Gas Removal
CEPCI:	Chemical Engineering Plant Cost Index
TIC:	Total Investment Cost per Ton
SP:	Steam to Plastic Ratio

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