

# Profitability Analysis of Selected Steam Methane Reforming Methods for Hydrogen Production

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## Abstract

One of the matured methods for producing hydrogen in bulk is steam methane reforming (SMR). The two commercial aspects of producing hydrogen from SMR are SMR with shift reactor (SR) and SMR with dry methane reforming (DRM). Although SMR with SR produces high hydrogen yield, it emits a large quantity of carbon dioxide (CO<sub>2</sub>). On the contrary, SMR and DRM produce low hydrogen yield but favorably emit a low quantity of CO<sub>2</sub>. However, it is not obvious which of these methods is more favourable economically. Consequently, using UNISIM Software Package, this study investigates three SMR methods namely SMR with SR, SMR with DRM and SMR with the combination of DRM and SR for the purpose of determining the most favourable route for producing hydrogen. This was done on the basis of feedstock rate of 100 kmol/hr of methane which reacted with 250 kmol/hr of steam for 8000 hrs annually using the rate of CO<sub>2</sub> and CO emissions (CO<sub>x</sub>) and the plant net profit percentage as performance indices. The profitability analysis shows that SMR/SR process is the most profitable process with a net profit percentage of 41.3%. Moreover, SMR/SR process has the highest yield and interestingly lowest CO<sub>x</sub> emission rate. It is therefore concluded that the most favourable process route, technically and economically, is SMR/SR for the production of hydrogen using methane as feedstock.

## Keywords

Hydrogen, Steam Methane Reforming, Dry methane Reforming, Carbon Oxides

## 1. Introduction

Hydrogen is used in diverse industries such as chemical, petrochemical and pe-

troleum refining and metallurgy. Hydrogen also finds specialty application as a rocket fuel due to its high energy release during combustion [1]. Hydrogen can be produced from a variety of feedstock which includes natural gas and coal. There are several methods through which hydrogen can be produced from fossil fuel sources; these include steam methane reforming (SMR), autothermal reforming, partial oxidation and the use of coal using coal gasification. Steam reforming is currently one of the most widespread and is also one of the least expensive processes of hydrogen production through which more than 90% of the hydrogen is being produced [2]. The most frequently used raw materials are natural gas and lighter hydrocarbons, around 50% of global hydrogen demand is met by natural gas and steam reforming, 30% from oil reforming, 18% from coal gasification, 3.9% from water electrolysis and 0.1% from other sources [3]. The first step of the steam methane reforming process involves methane reacting with steam to produce a synthesis gas (syngas), a mixture primarily made up of hydrogen ( $H_2$ ) and carbon monoxide (CO). In the second step, known as water gas shift (WGS) reaction, the carbon monoxide produced in the first reaction is sent to the water gas shift reactor to produce hydrogen and carbon dioxide ( $CO_2$ ) before it is sent for purification at the pressure swing adsorption (PSA) unit.

Anthropogenic activities which cause the emission of carbon dioxide ( $CO_2$ ) include the combustion of fossil fuels and other carbon-containing materials, the fermentation of organic compounds such as sugar and the breathing of humans.  $CO_2$  gas has a slightly irritating odour, is colourless and is denser than air. Green house gas (GHG) emissions are often measured in  $CO_2$  equivalent according to United States Environmental Agency (USEPA) [4], while transportation is the second largest source of  $CO_2$  emissions accounting about 31% of total U.S  $CO_2$  emissions and 26% of total US GHG emissions in 2011 [5]. Carbon dioxide accounts for over 84% of the greenhouse gas released into the atmosphere and originate almost exclusively from the utilization of fossil fuels [6]. The Energy Information Administration (EIA) estimates that if the current trends continue, worldwide carbon dioxide emissions will increase from 1559 million metric tons to 2237 million metric tons equivalent (1.5% annual change) by the year 2025 [7]. There is strong scientific evidence of a rapid, persistent and uncontrolled increase in atmospheric carbon dioxide ( $CO_2$ ) due to humans' activities, largely resulting from the burning of fossil fuels. The breathing in of  $CO_2$  is toxic to humans when levels are high with numerous deaths reported based on occupational exposure [8]. The  $CO_2$  exposure limit for an 8-hour working day has been set at 5000 ppm [9]. The issue of climate change is one of the main reasons for introducing hydrogen technology. One of the matured methods for producing hydrogen in bulk is steam methane reforming (SMR) and the two commercial methods of producing hydrogen from SMR are SMR with shift reactor (SR) and SMR with dry methane reforming (DRM). Although SMR and SR method produces high hydrogen yield, unfortunately, it produces a high quantity of carbon dioxide ( $CO_2$ ). On the contrary, SMR and DRM method produces low hydrogen

yield but favorably, low quantity of CO<sub>2</sub>. As the quantity of CO<sub>2</sub> emitted from SMR and SR is high, the cost of capturing it will increase the total cost of production. Hitherto, it is not obvious which of these methods is most favourable economically. Consequently, a comprehensive study of selected steam methane reforming methods becomes necessary.

## 2. Equipment Design and Specification

The shape of the unit operators used in this study is cylindrical with hemispherical ends while the equations for system specifications were obtained from Coulson and Richardson's Chemical Engineering Series, Volume 6 [10] while the prices of materials used were gotten from Alibaba website [11].

### Methods

To determine the volume of the unit operators, the first step is to determine the amount of the stream in molar flow rate in (kmol/hr), from mass flow rate in (kg/hr) and then dividing the mass flow rates (kg/hr) of the components with their respective molar mass (kg/kmol) as shown in Equation (1).

$$\text{Molar flow rate (kmol/hr)} = \sum \frac{\text{mass flow rate of component } i}{\text{molar mass of component } i} \quad (1)$$

In converting from molar flow rate (kmol/hr) to volumetric flow rate (m<sup>3</sup>/hr), note that 0.5 m<sup>3</sup>/kmol was the standard conversion factor used in multiplying molar flow rates of each component to obtain the volumetric flow rate as shown in Equation (2), as A is used to designate the feeds used in this study.

This implies that:

$$Q = \sum \frac{\text{Molar flow rate of A in kmol}}{\text{hr}} \times \frac{0.5 \text{ m}^3}{\text{kmol}} \quad (2)$$

The volumetric flow rate is the volume of fluid which flows per unit time. Equation (2) is the volumetric flow rate designated as (*Q*) and Equation (3) was used in calculating the volume (*V*) of the shape of the unit operator given as:

$$V = \frac{2\pi D^3}{3} \quad (3)$$

where (*V*) is the volume of the unit operation vessel, (*D*) is the diameter of the unit operation vessel. It is to note that according to design heuristics, 90% of the volume of the vessel should be filled with stream and as such relating the volume of the column (*V*) to the volumetric flow rate of the stream (*Q*) gives:

$$V = Q \times 1.1$$

The volume of the stream entering the unit operator is known and the diameter of the vessel is given as:

$$D = \sqrt[3]{\frac{3V}{2\pi}} \quad (4)$$

Equation (4) was obtained from Equation (3) by making *D* the subject formula.

The height of the column,  $H$ , is given as:

$$H = 3 \times D \quad (5)$$

The length of the column becomes:

$$\text{Length of the column } (L) = H - D \quad (6)$$

### 3. Process Route for Hydrogen Production and Purification

In this study, three processes namely steam methane reforming (SMR) and shift reactor (SR), SMR and dry reforming of methane (DRM) and SMR with the combination of SR and DRM were simulated. The feedstock for the production of hydrogen for each of the processes was methane which reacted with steam with a molar flow rate of 100 kmol/hr for methane and 250 kmol/hr for steam, respectively. The flow rate of methane which is 100 kmol/hr was the value chosen as the basis for the simulation performed on the three processes which was based on the ratio 2.5:1 to get the inflow of steam which is 250 kmol/hr and the annual operating time used for each of the three processes was 8000 hrs per year which was gotten by multiplying 24 hrs by 365 which gives a total of 8765 hrs per year. However, 765 hrs was set aside for maintenance and shutdown of plants while 8000 hrs was used for production.

To obtain a high yield of hydrogen using SMR process in this study, different ratios of steam to methane molar flow ratios like 2:1, 2.5:1, and 3:1 were used for the simulations but the ratio of 2.5:1 was the ratio that gave highest yield of hydrogen among the other three ratios. The operating conditions used in the simulations for the inlet temperature and pressure of the natural gas and steam for this process were 20°C, 520 kPa (for methane) and 180°C, 965 kPa (for steam) respectively, as these were the standard operating conditions for the SMR process [12]. The procedures that were followed in this study and the stream specifications are shown in **Figure 1**.

**Figure 2** shows the reaction that took place in SMR/SR process. After the production of the syngas by the reformer, the product was sent to shift reactor where carbon monoxide reacted with steam to produce CO<sub>2</sub> and more hydrogen and then the product was sent to the PSA for purification.

**Figure 3** shows the reaction that took place in SMR/SR/DRM process. After the production of the syngas by the reformer, the product was sent to shift reactor where carbon monoxide reacted with steam to produce CO<sub>2</sub> and more hydrogen, the feed leaving the shift reactor was sent to DRM where CH<sub>4</sub> reacted with CO<sub>2</sub> to produce more hydrogen and then the product was sent to the PSA for purification.

**Figure 4** shows the reaction that took place in SMR/DRM process. After the reaction of methane and steam in the reformer to produce syngas, the product was sent to the DRM where methane reacted with CO<sub>2</sub> to produce hydrogen and carbon monoxide; the product was then sent to PSA for purification as shown in **Figure 4**.

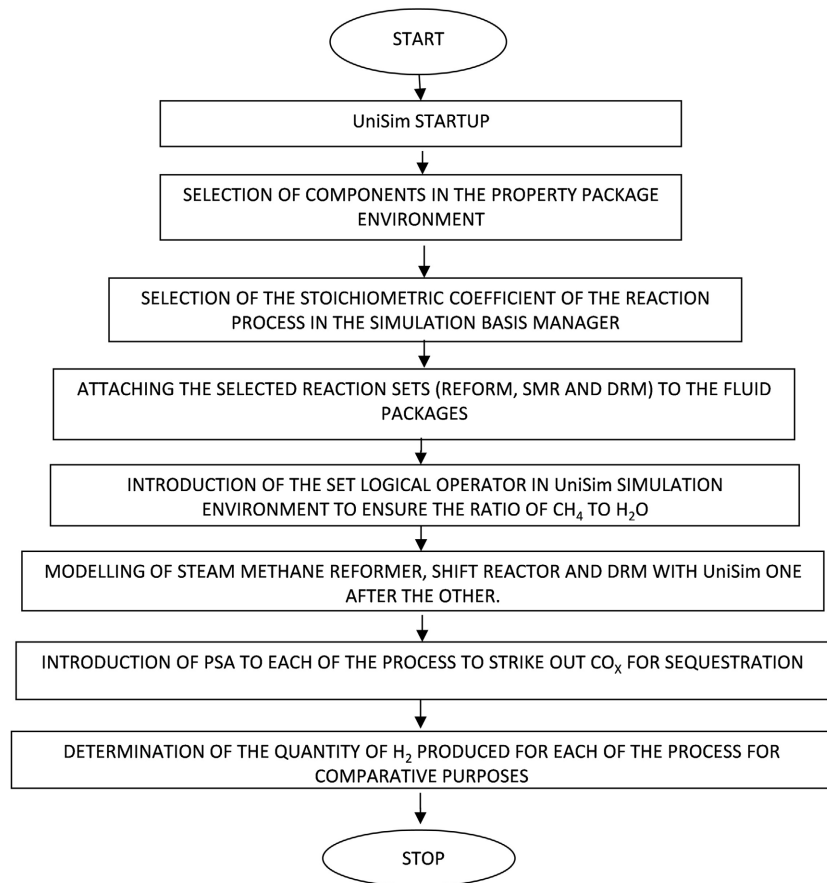


Figure 1. Modeling and simulation of steam methane reforming process with UNISIM.

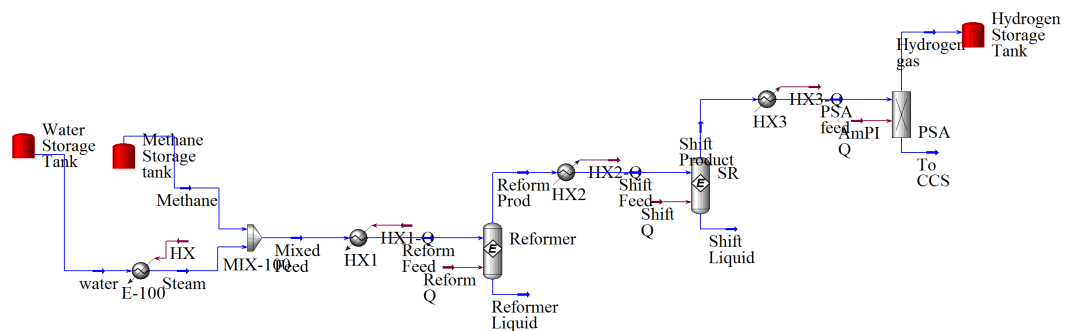


Figure 2. Process flow diagram for SMR and SR process.

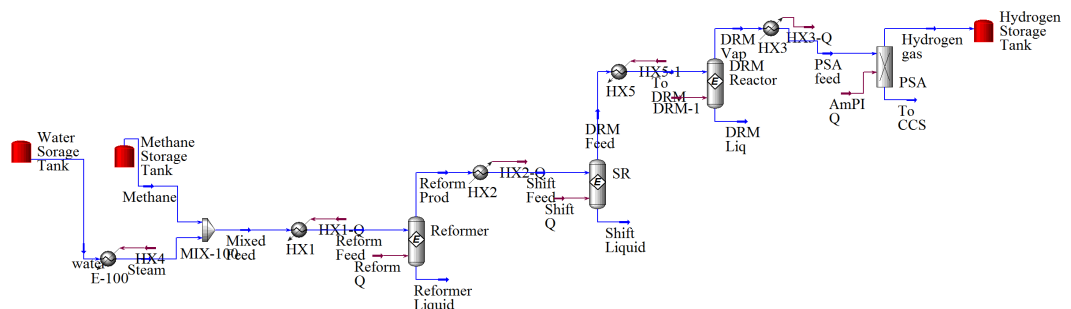
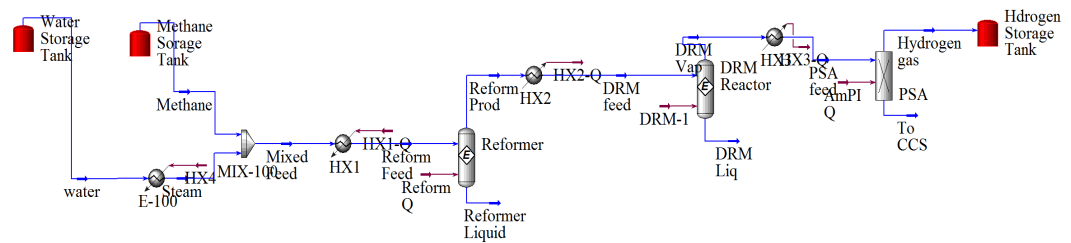


Figure 3. Combination of both shift reactor and DRM in the SMR process.



**Figure 4.** Process flow diagram for SMR and DRM process.

## 4. Results and Discussion

The simulation and economic analysis results for the three selected SMR process are as presented in **Tables 1-7**.

The reaction balance of steam methane reforming where methane reacted with steam to produce a syngas with inflows of methane and steam at 100 kmol/hr and 250 kmol/hr, respectively, is as shown in **Table 1**.

**Table 2** is the reaction balance that occurred in SMR/SR process. After the production of the syngas by the reformer, where there was an increase of  $\text{CO}_2$  but a decrease in CO while the hydrogen was also increased after the feed left the reformer.

**Table 3** is the reaction balance that occurred in SMR/SR/DRM process which shows that there was an increase in hydrogen, CO was decreased but an increase in  $\text{CO}_2$ .

**Table 4** shows reaction balance of SMR/DRM process, as there was an increase in  $\text{CO}_2$  but the CO was decreased.

**Table 5** shows the yield percentage of the three processes while **Table 6** shows the quantity of  $\text{CO}_x$  produced by each of the processes.

## 5. Discussion

For steam methane reforming (SMR) process, it is observed from **Table 1** that the quantity of methane and steam that reacted were 87.89 kmol/hr and 118.5 kmol/hr, respectively to produce 294.3 kmol/hr of hydrogen, 57.25 kmol/hr of CO and 30.63 kmol/hr of  $\text{CO}_2$  at 87.89% conversion. It is observed from **Table 2** that the addition of shift reactor (SR) increases the quantity of hydrogen produced from SMR from 294.3 kmol/hr to 327.4 kmol/hr and the level of CO is reduced from 57.25 kmol/hr to 24.17 kmol/hr; this is in agreement with the work of Younus [13]. Unfortunately, the level of  $\text{CO}_2$  increases from 30.63 kmol/hr to 63.72 kmol/hr with a conversion rate of 57.78%. **Table 3** shows that the quantity of  $\text{CO}_2$  produced by SMR and SR process is reduced from 63.72 kmol/hr to 51.60 kmol/hr, while the Hydrogen produced in this process is increased from 327.4 kmol/hr to 351.6 kmol/hr after incorporating DRM and SR into the SMR process. The shift reactor (SR) process has higher hydrogen yield than the SMR produced in the first reactor but the introduction of DRM into the process increase the quantity of hydrogen produced because methane had to react again with carbon dioxide to produce additional hydrogen, which means that the

**Table 1.** Reaction balance for SMR.

Component	Inflow (kmol/hr)	Outflow (kmol/hr)
Methane	100	12.11
H <sub>2</sub> O	250	131.5
Hydrogen	0	294.3
CO	0	57.25
CO <sub>2</sub>	0	30.63
Total	350	525.79

**Table 2.** Reaction balance for SMR and shift reactor.

Component	Inflow (kmol/hr)	Outflow (kmol/hr)
Methane	12.11	12.11
H <sub>2</sub> O	131.5	98.40
Hydrogen	294.3	327.40
CO	57.25	24.17
CO <sub>2</sub>	30.63	63.72
Total	525.79	525.8

**Table 3.** Reaction balance for the SMR/SR/DRM process.

Component	Inflow (kmol/hr)	Outflow (kmol/hr)
Methane	12.11	0
H <sub>2</sub> O	98.4	98.40
Hydrogen	372.40	351.6
CO	63.72	48.40
CO <sub>2</sub>	24.17	51.60
Total	525.8	550

**Table 4.** Reaction balance for the SMR/DRM process.

Component	Inflow (kmol/hr)	Outflow (kmol/hr)
Methane	12.11	0
H <sub>2</sub> O	131.5	131.5
Hydrogen	294.3	318.5
CO	57.25	81.48
CO <sub>2</sub>	30.63	18.52
Total	525.79	550

**Table 5.** Hydrogen produced in kmol/hr for the three processes.

Process Type	Yield Percentage (%)
SMR and Shift Reactor	91
SMR, Shift Reactor and DRM	83
SMR and DRM	72

**Table 6.** The quantity of CO<sub>x</sub> (CO and CO<sub>2</sub> produced by each of the process).

Process Type	Yield Percentage (%)
SMR and Shift Reactor	87.89
SMR, Shift Reactor and DRM	100
SMR and DRM	100

**Table 7.** Results of techno—economic analyses of the three processes.

Process Type	Cox Emission (kmol/hr)	ROI (%)	Payback Time (Months)
SMR and Shift Reactor	87.89	52	28 Months
SMR, Shift reactor and DRM	100	44.2	32 Months
SMR and DRM	100	30.8	48 Months

combination of SR/DRM into SMR process produces more hydrogen than SMR and SR process, which consequently reduces the quantity of CO<sub>2</sub> but unfortunately leads to an increase in CO.

**Table 4** shows that SMR and DRM alone produces the least hydrogen compared to the other processes; however, it has the least CO<sub>2</sub> present but has highest CO which means that this process can minimize the quantity CO<sub>2</sub> production but has the capacity to increase the rate of CO generation. **Table 5** shows that the SMR and SR has the highest yield percentage of 91% while SMR and DRM has the least yield percentage of 72%; this is in agreement with the report by Nikolaidis and Poullikkas [12]. **Table 6** shows that the combination of SMR and shift reactor has the least CO<sub>x</sub> of 87.89 kmol/hr compared to the other two processes which produces CO<sub>x</sub> of 100 kmol/hr each. Finally, **Table 7** shows the results of the economic analysis of the three processes with SMR and SR having the highest return on investment of 52% and the shortest payback time of 28 months. From all the results presented, it is obvious that SMR and SR is the most favourable process for the production of hydrogen from methane as feedstock because from an economic point of view, it has the highest profit and also has the least CO<sub>x</sub> produced as byproduct.

## 6. Conclusions

The most popular method of producing hydrogen is the SMR process. However, it is not obvious which of its two commercial variants, SMR and SR, and SMR



and DMR is more favorable considering their profitability and CO<sub>x</sub> emission levels.

This study, therefore, investigates the profitability and environmental implications of these two plants individually and compared them with the combined plants (SMR/SR/DRM) using the same performance indices. The result shows that SMR/SR process is the most profitable process with a net profit percentage of 41.3%. Furthermore, SMR/SR process has the highest yield and interestingly lowest CO<sub>x</sub> emission rate of 87.9 kmol/hr. It is therefore concluded that the most favourable process route, technically and economically is SMR/SR process for the production of hydrogen using methane as feedstock.

### Conflicts of Interest

There is no conflict of interest associated with this work.

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