International Journal of Engineering Research & Technology (IJERT)
ISSN: 2278-0181
Vol. 2 Issue 7, July - 2013

Simulation, Sensitivity Analysis and Optimization of Hydrogen Production by Steam Reforming of Methane Using Aspen Plus

Abdulwahab GIWA[1] and Saidat Olanipekun GIWA[2]

[1]Department of Chemical Engineering, Faculty of Engineering, Middle East Technical University, 06800, Ankara, TURKEY

[2]Department of Chemical Engineering, Faculty of Engineering, Ankara University, 06100, Ankara, TURKEY

ABSTRACT

Methane steam reforming has been simulated, analyzed and optimized with the aid of Aspen Plus, in this work. The model of the steam reforming process used was developed using equilibrium reactor type. Both steam reforming process model with and without mixer were developed and simulated in Aspen Plus environment. The sensitivity analyses and the optimization of the process were accomplished using the Sensitivity and the Optimization sections of Model Analysis Tool of Aspen Plus. The results obtained revealed that the simulations with and without mixer gave the same mole fractions of the process components and that the simulation with liquid methane feed was able to produce highmole fraction of the desired product(hydrogen) at approximately atmospheric pressure. Furthermore, it was revealed from the sensitivity analyses of the process that optimization was necessary to obtain the operating variables that would yield hydrogen in very high and highest purity among the components involved in the process and the optimization was able to give a satisfactory value of 0.7432 as the mole fraction of hydrogen present in the top product of the reactor when the optimum reactor temperature, reactor pressure, volumetric flow rate of feed water and volumetric flow rate of feed methane were approximately 964.89 ^{o}C, 1 bar, 0.0172 L/min and 0.0353 L/min, respectively.

Keywords: Steam reforming, Aspen Plus, sensitivity analysis, optimization, hydrogen.

INTRODUCTION

Steam reforming is a process that consists of catalytically reacting a mixture of steam and hydrocarbons at an elevated temperature to form a mixture of H_2 and oxides of carbon (Austin, 1984).

Steam reforming of methane or natural gas is still the predominant method for producing the hydrogen rich synthesis gas (hydrogen and carbon monoxide) (Elnashaie et al., 1993; Abashar et al., 2003; Abashar, 2003; Basile et al., 2003; Gallucci et al., 2004; Abashar, 2004; Yu et al., 2008; Ye et al., 2009). In fact, about 50% of hydrogen demand is satisfied by means of methane-steam reforming (Barbieri et al., 1997). The recent increases in the demand for hydrogen by many processes such as oil refining, methanol, metallurgy, ammonia, aniline, space transportation, etc., have imposed a strong economic incentive to improve hydrogen production technology (Aboosadi et al., 2011). This improvement also includes obtaining valid optimum conditions for hydrogen production.

In steam reforming, basically, the following reactions shown in Equations (1) and (2) below occur:

$$C_nH_m + nH_2O \leftrightarrow nCO + \left(\frac{m}{2} + n\right)H_2 \quad (1)$$

$$CO + H_2O \leftrightarrow CO_2 + H_2 \quad (2)$$

Although the equations are shown for the general case of any hydrocarbon feed, only light hydrocarbons have been successfully used in commercial practice. Natural gas is the most common, and propane and butane (LPG) are also frequently used. With the use of a specially prepared catalyst, naphtha is also a suitable feedstock (Austin, 1984).

The first reaction (Equation (1)) is the reforming reaction. It is highly endothermic, and the moles of product exceed the moles of reactant so the reaction goes to completion at high temperature and low pressure. Excess steam is normally used. Although the basic purpose is to prevent carbon formation, it also helps to force the reaction to completion (Austin, 1984).

The second reaction (Equation (2))is the water-gas-shift reaction. It is mildly exothermic and is favored by low temperature but unaffected by pressure. Excess steam also forces this reaction to completion and is so used. A catalyst is usually employed. Both these reactions occur together in the steam-reforming furnace at temperatures of 760 to 980 ^{o}C. The composition of the product steam depends upon the process conditions, including temperature, pressure, and excess steam, which determine equilibrium, and the velocity through the catalyst bed, which determines the approach to equilibrium. A typical product contains approximately 75% H_2, 8% CO, and 15% CO_2; the remainder consists of nitrogen and unconverted methane (Austin, 1984).

For the production of additional hydrogen, the reformer is followed by a separate stage of water-gas-shift conversion. Additional steam is added, and the temperature is reduced to 315 to 370 ^{o}C to obtain more favorable equilibrium conditions. A single stage converts 80 to 95% of the residual CO to CO_2 and H_2. Because the reaction is exothermic, the reactor temperature rises; this enhances the reaction rate but has an adverse effect on the equilibrium. When high concentrations of CO exist in the feed, the shift conversion is usually conducted in two or more stages, with interstage cooling to prevent an excessive temperature rise. The first stage may operate at higher temperatures to obtain high reaction rates, and the second at lower temperatures to obtain good conversion (Austin, 1984).

According to the information gathered from the literature, it was discovered that some researches have been carried out on modeling and simulation of hydrogen production. For instance, Jin et al. (2010) simulated hydrogen production via oxidative steam reforming of ethanol with a dense tubular membrane reactor (DMR) sequentially with ASPEN PLUS. They discovered from their simulation results that there was an optimal length of tubular membrane reactor at the operating temperature and steam-to-ethanol (H_2O/EtOH) ratio, under which hydrogen and carbon monoxide formation reach their maxima. They found that at the fixed ratio H_2O/EtOH = 0.5, the maximum hydrogen yield occurred at a temperature of 1123 K with the length of tubular DMR of 4 cm, while for the temperature of 1073 K, the maximum hydrogen yield appears at H_2O/EtOH = 5 with the length of tubular DMR of 0 cm. Pérez-Moreno et al. (2013) studied the effects of the main operating variables (temperature, steam/oxygen ratio, steam/methane ratio and relative velocity with respect to the minimum fluidization velocity) on the oxidative steam reforming of methane in a two-zone fluidized-bed reactor over a Ni/Al_2O_3 catalyst. They compared the hydrogen yield obtained with those of the literature and found out that, despite the very low steam/methane ratios that were worked with, high values of hydrogen yield at both high methane conversion and at steady state were obtained from the two-zone fluidized-bed reactor. Ye et al. (2009) simulated hydrogen production via steam methane reforming with in situ hydrogen separation in fluidized bed membrane reactors using Aspen Plus by dividing the reactor into several successive steam methane sub-reformers and membrane sub-separators. They investigated the influences of reactor pressure, temperature, steam-to-carbon ratio, and permeate side hydrogen partial pressure on reactor performances. They discovered from their studies that extracting hydrogen in situ was able to shift the equilibrium of steam methane reactions forward, removing the thermodynamic bottleneck, and improving hydrogen yield while neutralizing, or even reversing, the adverse effect of pressure. Xie et al. (2010) developed an equilibrium model of steam methane reforming coupled with in-situ membrane separation for hydrogen production. Their model employed Sievert's Law for membrane separation and minimum Gibbs energy model for reactions. The reforming and separation processes were coupled by the mass balance. They assumed a continuously stirred tank reactor for the fluidized bed hydrodynamics in the model. The influences of reactor pressure, temperature, steam to carbon ratio, and permeate side hydrogen partial pressure on solid carbon, NH_x and NO_x formation were studied in their work using the model they developed. Dehkordi et al. (2011) developed a compartment model to describe the flow pattern of gas within the dense zone of a tapered membrane-assisted fluidized-bed reactor in the bubbling mode of operation for steam reforming of methane under wall heat flux. The parameters of their developed model were determined using experimental data and good agreements were obtained between the model predictions and the corresponding experimental data. The developed model was then utilized to predict the behavior of the tapered membrane-assisted fluidized-bed reactor under various operating and design conditions. The influences of tapered angle, bed operating temperature and pressure, and feed temperature on the methane conversion and the total yield of hydrogen were carefully investigated. They discovered that there was a tapered angle that could maximize the performance capability of the tapered membrane-assisted fluidized-bed reactor. Chen et al. (2003) investigated the performance of a novel circulating fast fluidized bed membrane reformer (CFFBMR) using a reliable mathematical model. They discovered that the removal of product hydrogen using hydrogen permselective membranes was able to break the thermodynamic equilibrium in the reversible system and made it possible to operate the process at lower temperatures. It was also pointed out in their work that the oxidative reforming of a part of the feed methane by oxygen input into the reformer using direct feed or through oxygen-permeable membranes supplied the heat needed for the highly endothermic steam reforming of methane. So, they showed that the combination of the exothermic oxidative reforming and endothermic steam reforming not only produced high yield hydrogen but also made it possible to operate the CFFBMR under autothermal conditions. They, therefore, concluded that the novel configuration was a highly efficient hydrogen producer with minimum energy consumption. Gallucci et al. (2004) used modelling viewpoint to investigate methane steam reforming reaction by considering the effects of different parameters on methane conversion. They considered the influence of lumen pressure on methane conversion at constant temperature, and it was found that increasing this parameter made the equilibrium methane conversion to increase for membrane reactor, whereas it decreased for the traditional one. Patel and Sunol (2007) developed a distributed mathematical model for thermally coupled membrane reactor that was composed of three channels for methane steam reforming. Taking the mass and energy balance equations for the thermally coupled membrane reactor, their developed model formed a set of 22 coupled ordinary differential equations. Using appropriate boundary conditions, they were able to solve the distributed reactor model for steady-state operation as a boundary value problem and investigated the performance of the reactor numerically for various key operating variables such as inlet fuel concentration, inlet steam/methane ratio, inlet reformer gas temperature and inlet reformer gas velocity. The results they obtained showed that the introduction of membrane in the thermally coupled reactor resulted in a significant improvement in conversion and hydrogen recovery yield. Sadooghi and Rauch (2013) also developed a mathematical model to simulate synthesis gas production by methane steam reforming process in a fixed bed reactor filled with catalyst particles. In their work, due to the endothermic nature of the reforming reactions, heat was supplied into the reactor by means of electrical heating, therefore, the reactor and catalyst particles were exposed to significant axial and radial temperature gradients. They used a pseudo heterogeneous model in order to exactly represent the diffusion phenomena inside the reactor tube. Heat and mass transfer equations were coupled with detailed reaction mechanisms and solved for both the flow phase and within the catalyst pellets. The reaction was investigated from a modeling view point considering the effect of different temperatures ranging from 873 to 1073 K on methane conversion and hydrogen yield. Their results provided temperature and concentration distribution along the reactor axial and radial coordinates and found strong radial temperature gradients particularly close to the entrance of the reactor.

Looking at the past researches carried out so far on the reforming of methane to produce hydrogen, it was noticed that no study has been done on this research using equilibrium reactor for the accomplishment of the process. Therefore, it is aimed in this work to use an equilibrium reactor to simulate the production of hydrogen from methane using steam reforming process.

PROCEDURES

The Aspen Plus (Aspen, 2012) model used for the simulation of the steam reforming of methane for the production of hydrogen is as shown in Figures 1 and 2 below.

The two models had two feed streams – water and methane. In the first model (Figure 1), the two feed streams were passed directly into the reactor while in the second model (Figure 2), they (the feed streams) were fed into a mixer before being passed into the reactor. The two models were run to find out whether there was any significance effect of the mixing of the reactants (water and methane) before being fed into the reactor. In comparing the two models (one without mixer and the other with mixer), the temperature and the pressure of the water feed were 25 ^{o}C and 1 bar, respectively. Also, methane feed was fed into the reactor at a temperature of -163 ^{o}C and a pressure of 1 bar. In other words, for the comparison of the developed Aspen Plus models, the two reactants were fed in liquid form because the feed conditions were below their boiling conditions.

Thereafter, the effects of the phase of methane feed (liquid or vapor) were investigated by simulating one of the developed Aspen Plus steam reforming models using a reactor pressure of 1 bar as well as using methane feed, first, in vapor form, and, then, in liquid form. In this case, both water and methane were passed into the reactor at a pressure of 1 bar, but the temperatures of methane and water passed into the reactor were -100 and 25 ^{o}C, respectively. This simulation was carried out to know the phase of methane feed suitable for steam reforming being carried out at a reactor pressure of 1 bar.

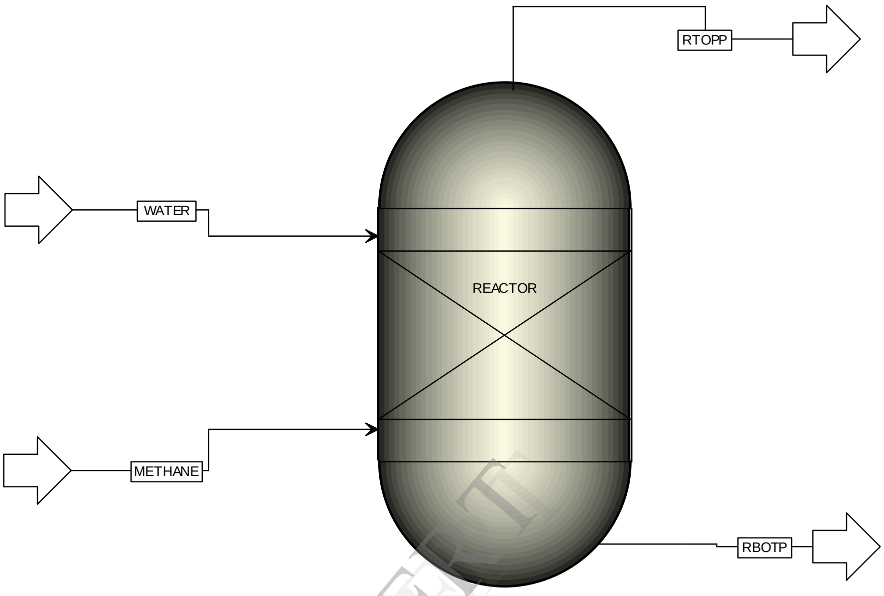

Figure 1. Aspen Plus model of methane steam reforming process without mixer

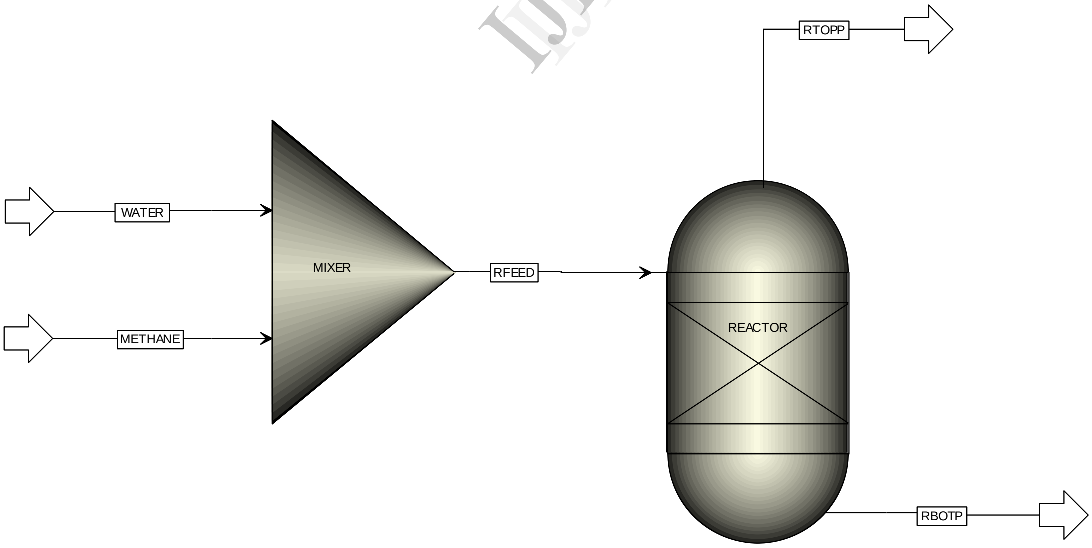

Figure 2. Aspen Plus of model methane steam reforming process with mixer

Furthermore, the sensitivity analyses of the mole fractions of the components obtained from the top of the reactor towards each of the reactor temperature, the reactor pressure, feed water volumetric flow rate and feed methane volumetric flow rate were performed with the aid of Sensitivity Analysis section of Model Analysis Tool of Aspen Plus to find out how each of the variables influence the mole fractions of the process components obtained from the top product of the reactor. The ranges of the operating variables used for the sensitivity analyses are given in Table 1 below.

Table 1. Ranges of operating variables used for sensitivity analyses

Parameter	Lower range	Upper range	Increment
Temperature (^{o}C)	760.0000	980.0000	5
Pressure (bar)	1.0000	11.0000	0.25
Feed water flow rate (L/min)	0.0100	0.0500	0.001
Feed methane flow rate (L/min)	0.0100	0.0500	0.001

Finally, since it was desired to have high purity of hydrogen in the top product of the reactor, the optimization of the reactor was carried out using the Optimization section of Model Analysis Tool of Aspen Plus by making the objective function of the optimization to be the maximization of the mole fraction of hydrogen in the top product of the reactor. The ranges specified for the optimization of the variables were the same as the ones used for the sensitivity analyses (see Table 1).

The chemical reactions involved in the steam reforming of methane considered in this work are given as shown in Equations (3) and (4).

$$CH_4 + H_2O \leftrightarrow CO + 3H_2 \tag{3}$$

$$CO + H_2O \leftrightarrow CO_2 + H_2 \tag{4}$$

Equation (3) is the reforming reaction while Equation (4) is the water-gas-shift reaction of the process. Both reactions were simulated simultaneously in Aspen Plus.

RESULTS AND DISCUSSIONS

The results obtained from the steady-state simulations of the steam reforming process models, with and without mixer, developed using Aspen Plus are as given in Table 2 below. These simulations were carried out using a reactor temperature of 900 ^{o}C and a pressure of 1 bar.

Table 2. Component mole fraction obtained from reforming process with and without mixer

Component	Mole fraction obtained from reforming process	
	without mixer	with mixer
CH_4	0.0002	0.0002
H_2O	0.1777	0.1777
CO	0.1606	0.1606
H_2	0.6256	0.6256
CO_2	0.0359	0.0359

As can be seen from the results, it was discovered that the mole fractions of the process components (methane – CH_4, water – H_2O, carbon monoxide – CO, hydrogen – H_2, and carbon dioxide – CO_2) obtained from the simulations of the Aspen Plus steam reforming models with and without mixer were exactly the same. Looking at Table 2, it can be seen that the two models gave the mole fraction of hydrogen of 0.6256 as the highest value among the mole fractions of the components contained in the top product of the reactor. Apart from that, also seen in Table 2, considering the results of the two process models, methane was found to have the least mole fraction in the top product of the reactor. It was thus discovered that the use of mixer was not necessary for this process. That is, it was seen that the feed streams could be fed into the reactor directly without being mixed inside the mixer that will have a negative effect on the economic advantage of the process. Based on this, the Aspen Plus model without any mixer developed for the steam reforming of methane was chosen as the one used for further investigations of the process.

Furthermore, another steady-state simulation was carried with the developed Aspen Plus steam reforming model without mixer at a temperature and a pressure of 850 ^{o}C and 1 bar respectively using vaporized and liquid methane feeds. As mentioned before, the vaporized methane feed was achieved by passing methane feed into the reactor at a temperature of -100 ^{o}C and a pressure of 1 bar that was higher than its boiling point conditions while the temperature and the pressure of the liquid one (methane feed) was -163 °C (a temperature less than its boiling point) and 1 bar.

Table 3. Mole fraction obtained from the reforming process with vaporized and liquid methane feed

Component	Mole fraction obtained from reforming process	
	with vaporized methane feed	with liquid methane feed
CH_4	0.0000	0.0004
H_2O	0.9937	0.1743
CO	0.0000	0.1566
H_2	0.0051	0.6289
CO_2	0.0013	0.0398

The results of the simulations with the vaporized and the liquid methane, shown in Table 3, revealed that very high mole fraction of hydrogen was obtained at the operating conditions considered using liquid methane because the mole fraction of hydrogen obtained when liquid methane feed was used was 0.6289 while that obtained when vaporized methane was fed into the reactor was just 0.0051. It has thus been established that the Aspen Plus model without any mixer and with water and liquid methane as the feeds was suitable enough to give relatively high mole fraction of reactor top hydrogen at the considered operating conditions, especially when the reactor pressure was 1 bar.

Moreover, the results obtained from the sensitivity studies carried out by finding out how the mole fractions of the components obtained from the top product of the reactor responded to some input variables of the process are given in Figures 3 – 6. The input variables considered in the sensitivity analyses were the reactor temperature, the reactor pressure, the feed water volumetric flow rate and the feed methane volumetric flow rate.

Shown in Figure 3 are the responses of the mole fractions of the components to the reactor temperature. As can be noticed from the figure, the mole fractions of methane, carbon dioxide and hydrogen present in the top product of the reactor decreased with increase in the reactor temperature while those of carbon monoxide and water increased with increase in the reactor temperature. It can be observed from the results (Figure 3) that, the more the temperature of the reactor was being increased, the more carbon monoxide was being formed in the reaction. This was discovered to mean that increase in reactor temperature favored the reforming reaction than the water-gas-shift reaction. This observation was found to be in support of the information obtained from the literature that says, water-gas-shift reaction is favored by low temperature.

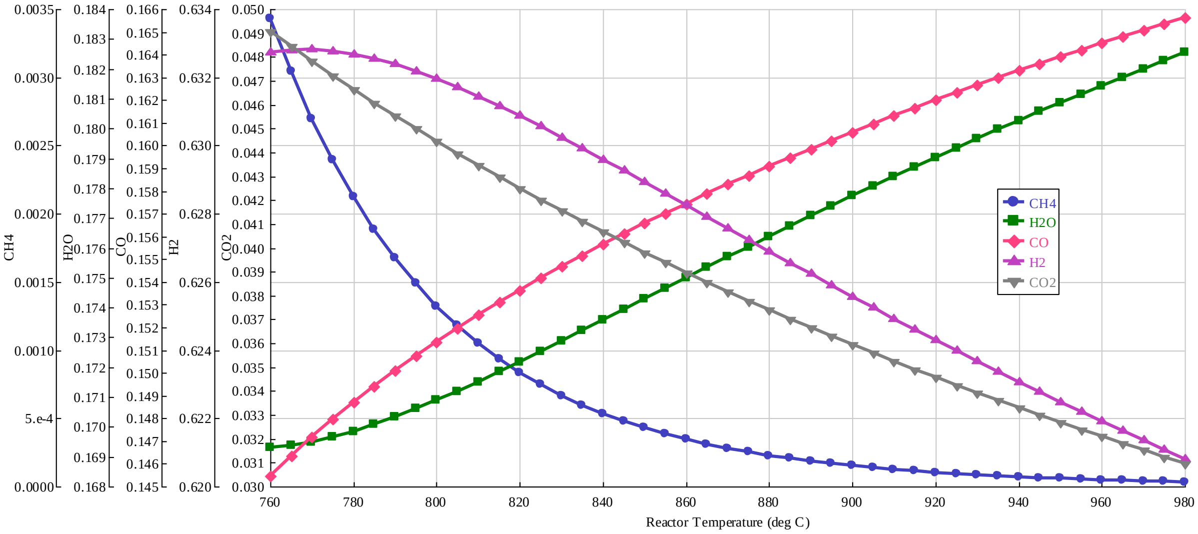

Figure 3. Responses of the mole fractions of process components to reactor temperature

The results obtained from the sensitivity analysis of the mole fractions of the components present in the top product of the reactor towards the reactor pressure are as shown in Figure 4. It was observed from the results that the mole fractions of carbon monoxide and hydrogen decreased with increase in the reactor pressure while those of methane, water and carbon dioxide increased as the reactor pressure was increased. The suppression of the production of carbon monoxide with increase in the reactor pressure was actually favorable, but the response of the mole fraction of hydrogen (which was the desired product) towards the increase in the reactor pressure was found to be unfavorable because hydrogen was desired to have very high mole fraction in the top product stream of the reactor.

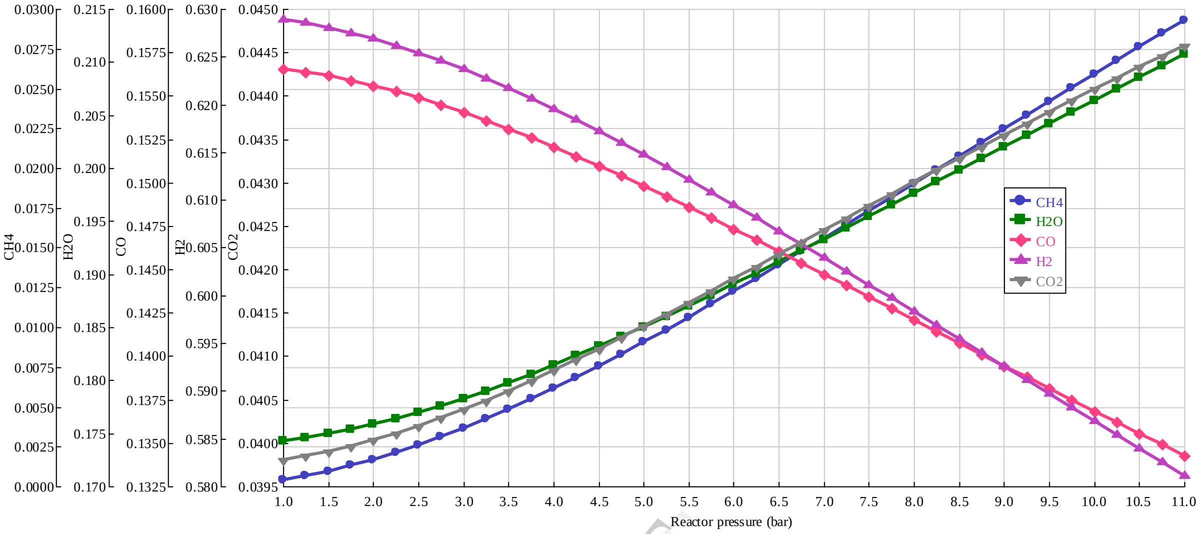

Figure 4. Responses of the mole fractions of process components to reactor pressure

Similarly, in Figure 5, the changes that occurred in the mole fractions of the components as the volumetric flow rate of feed water was varied (increased) are shown. From the figure, it was discovered that the mole fractions of water and carbon dioxide increased with increase in the feed water flow rate while that of methane was found to decrease. As can be seen from the results shown in the figure, the mole fractions of carbon monoxide and hydrogen first increased and later decreased with increase in the volumetric flow rate of feed water. Itwas observed from the results shown in Figure 5 that the component with the highest mole fraction was not hydrogen (the desired product of this work) when the feed water volumetric flow rate was varied. As such, there was the need to look for another input variable to be manipulated in order to have hydrogen as the component with the highest mole fraction in the top product of the reactor. Based on this, it was decided to also manipulate the volumetric flow rate of feed methane and obtain the corresponding responses of the process components.

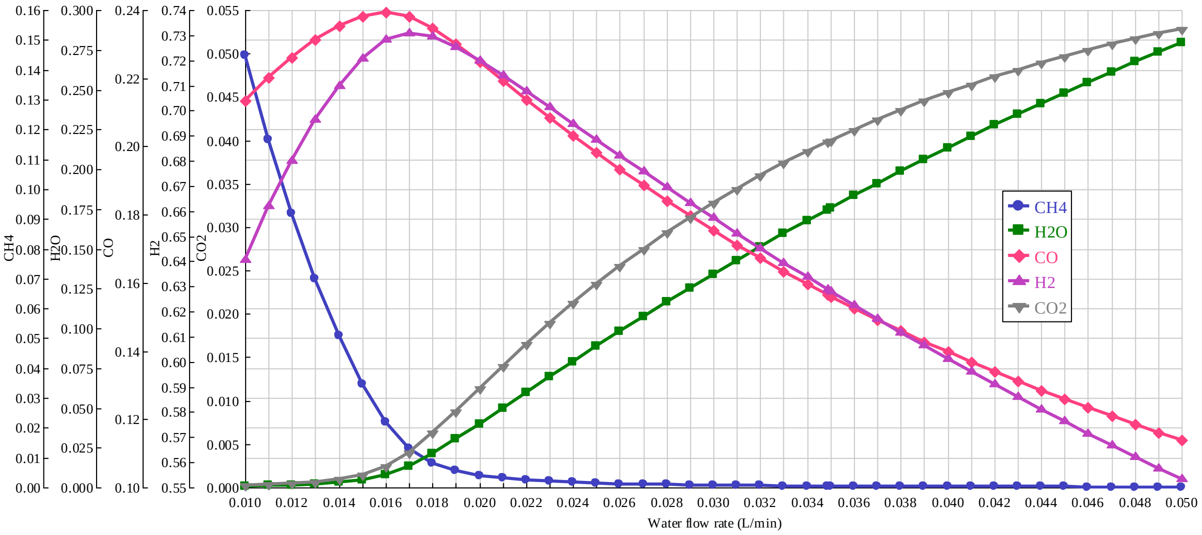

Figure 5. Responses of the mole fractions of process components to water volumetric flow rate

Investigating the responses of the mole fractions of the components further by varying the volumetric flow rate of methanol feed, the results obtained are as shown in Figure 6. From the results, it was seen that, as the volumetric flow rate of methane feed was increased, the mole fraction of carbon dioxide first increased and later decreased, that of water decreased while those of hydrogen, carbon monoxide and methane increased. As can be seen from the figure, within the range of the input variables investigated, the mole fraction of hydrogen was discovered not to be the highest one. Besides, the value of the mole fraction of hydrogen obtained in this case was found to be less than the maximum mole fraction value obtained when the volumetric flow rate of water was varied (see Figure 5).

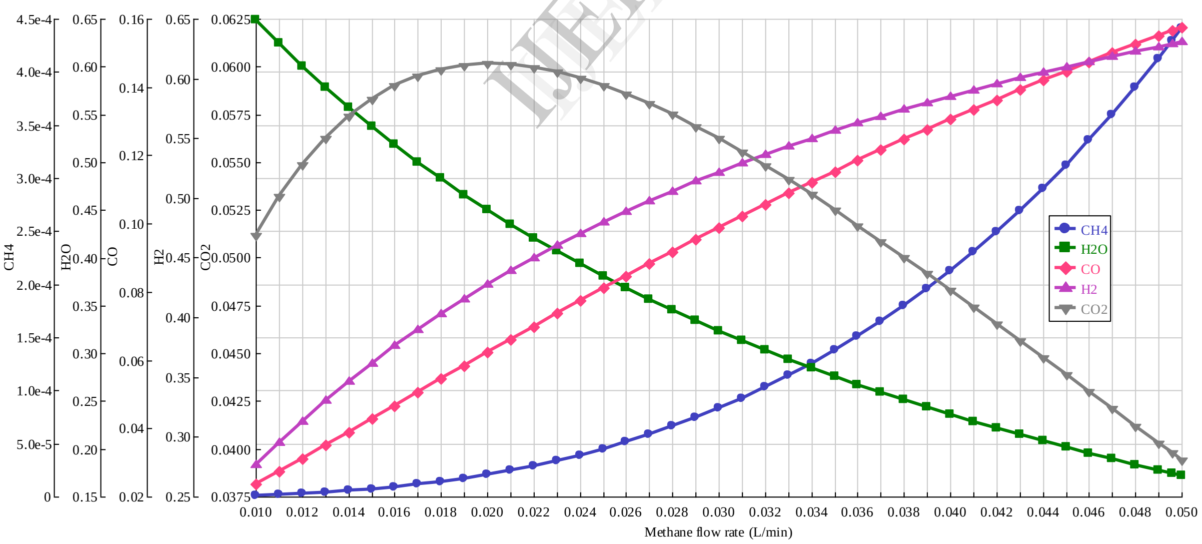

Figure 6. Responses of the mole fractions of process components to methane volumetric flow rate

As can be observed from the sensitivity analyses of the process carried out considering the four input variables (the reactor temperature, the reactor pressure, the feed water volumetric flow rate and the feed methane volumetric flow rate), the tangible input value(s) to give the mole fraction of hydrogen as the highest one among the mole fractions of the components involved has/have not been obtained. This was what called for the optimization of the process using the same Aspen Plus.

The optimization of this process was carried out to obtain the optimum input variables that would give the mole fraction of hydrogen contained in the top product of the reactor as the highest one among the mole fractions of the components involved in the process. The optimum input variables obtained from the optimization carried out and those of the steady-state simulation carried out prior to it (the optimization) are as given in Table 4. From the table, it was discovered that the input values obtained from the optimization were different from those of the steady-state simulation carried out prior to the optimization except that of the reactor pressure. Also noticed from the values of the optimum values given in Table 4 was that the values given by the optimization were within the ranges specified for the input variables during the optimization. This was an indication that the function of the optimization tool of Aspen Plus used to obtain the optimum conditions of this process was good.

Table 4. Steady-state and optimum parameters obtained from the process with liquid methane feed

Parameter	Values obtained from	
	Steady state	Optimization
Reactor temperature (^{o}C)	870.0000	964.8890
Reactor pressure (bar)	1.0000	1.0000
Water flow rate (L/min)	0.0350	0.0172
Methane flow rate (L/min)	0.0350	0.0353

Also recorded and given in Table 5 are the steady-state mole fractions of the components obtained from the simulations carried prior to the optimization at the reactor temperature and pressure of 870 ^{o}C and 1 bar respectively and the ones obtained from the optimization.

Table 5. Steady-state and optimum mole fraction obtained from the process with liquid methane feed

Component	Mole fraction obtained from reforming process from	
	Steady state	Optimization
CH_4	0.0003	0.0033
H_2O	0.1757	0.0062
CO	0.1583	0.2459
H_2	0.6276	0.7432
CO_2	0.0382	0.0014

From the table, it was noticed that the optimized mole fractions of the components were different from their steady-state values, just as it was discovered in the case of the steady-state and the optimum input variables. Specifically, the value of the mole fraction of hydrogen present in the top product of the reactor when the steady-state simulation was carried out was actually obtained to be 0.6276. After the optimization, that is, with the simulation of the model carried out with the optimum input values, as can be seen from the results shown in Table 5, hydrogen had the highest mole fraction of 0.7432 among the components involved in the steam reforming process investigated.

CONCLUSIONS

The results obtained from the simulation of the developed model of methane steam reforming with the aid of Aspen Plus have revealed that the simulations with and without mixer gave the same mole fractions of the process components and that liquid methane feed was able to produce good results at approximately atmospheric pressure. Furthermore, the sensitivity analyses of the process carried out pointed out that optimization was necessary to obtain the operating variables that would yield hydrogen in highest purity among the components of the process and the result of the optimization gave a value of 0.7432 as the mole fraction of hydrogen present in the top product of the reactor when the reactor temperature, the reactor pressure, the volumetric flow rate of feed water and the volumetric flow rate of feed methane were approximately 964.89 ^{o}C, 1 bar, 0.0172 L/min and 0.0353 L/min, respectively. The optimized mole fraction (0.7432)of hydrogen obtained was found to compare very well with the literature fraction value (0.75), which implied that the developed Aspen Plus steam reforming model was able to represent the process very well.

ACKNOWLEDGEMENTS

The authors acknowledge and appreciate the supports received from the Prime Ministry of The Republic of Turkey, Presidency for Turks Abroad and Related Communities for their programmes.

NOMENCLATURES

CFFBMR	Circulating fast fluidized bed membrane reformer
CH4	Methane
CO	Carbon monoxide
CO2	Carbon dioxide
DMR	Dense tubular membrane reactor
EtOH	Ethanol
H2	Hydrogen
H2O	Water
LPG	Liquefied petroleum gas
RBOTP	Reactor bottom product
RFEED	Reactor feed
RTOPP	Reactor top product

REFERENCES

1) Abashar, M.E.E. 2003a. Stagewise fluidized bed membrane reactors for methane-steam reforming. Afinidad, 59(504), 184-191.
2) Abashar, M.E.E. 2004. Coupling of steam and dry reforming of methane in catalytic fluidized bed membrane reactors. International Journal of Hydrogen Energy, 29(8), 799-808.
3) Abashar, M.E.E., Alhumaizi, K.I. and Adris A.M. 2003b. Investigation of methane-steam reforming in fluidized bed membrane reactors. Chemical Engineering Research and Design, 81(2), 251-258.
4) Aboosadi, Z. A., Rahimpour, M.R., Jahanmiri, A. 2011. A novel integrated thermally coupled configuration for methane-steam reforming and hydrogenation of nitrobenzene to aniline. International Journal of Hydrogen Energy, 36(4), 2960-2968.
5) Aspen. 2012. Aspen Plus. Aspen Technology, USA.
6) Austin, G.T. 1984. Shreve's Chemical Process Industries. 5th Edition, McGraw-Hill, Singapore.
7) Barbieri, G., Violante, V., Maio, D., F.P., Criscuoli, A. and Drioli, E. 1997. Methane steam reforming analysis in a palladium-based catalytic membrane reactor. Industrial and Engineering Chemistry Research, 36(8), 3369-3374.
8) Basile, A., Paturzo, L. and Vazzana, A. 2003. Membrane reactor for the production of hydrogen and higher hydrocarbons from methane over Ru/Al_2O_3 catalyst. Chemical Engineering Journal, 93(1), 31-39.
9) Chen, Z., Prasad, P., Yan, Y. and Elnashaie, S. 2003. Simulation for steam reforming of natural gas with oxygen input in a novel membrane reformer. Fuel Processing Technology 83(1-3), 235-252.
10) Dehkordi, A.M., Savari, C. and Ghasemi, M. 2011. Steam reforming of methane in a tapered membrane - assisted fluidized - bed reactor: Modeling and simulation. International Journal of Hydrogen Energy, 36(1), 490-504.
11) Elnashaie, S.S.E.H. and Abashar, M.E.E. 1993. Steam reforming and methanation effectiveness factors using the dusty gas model under industrial conditions. Chemical Engineering and Processing: Process Intensification, 32(3), 177-189.
12) Gallucci, F., Paturzo, L. and Basile, A. 2004. A simulation study of the steam reforming of methane in a dense tubular membrane reactor. International Journal of Hydrogen Energy, 29(6), 611-617.
13) Jin, Y., Rui, Z., Tian, Y., Lin, Y. and Li, Y. 2010. Sequential simulation of dense oxygen permeation membrane reactor for hydrogen production from oxidative steam reforming of ethanol with ASPEN PLUS. International Journal of Hydrogen Energy, 35(13), 6691-6698.

14) Patel, K.S. and Sunol, A.K. 2007. Modeling and simulation of methane steam reforming in a thermally coupled membrane reactor. International Journal of Hydrogen Energy 32(13), 2344-2358.
15) Pérez -Moreno, L., Soler, J., Herguido, J. and Menéndez, M. 2013. Stable hydrogen production by methane steam reforming in a two-zone fluidized-bed reactor: Effect of the operating variables. International Journal of Hydrogen Energy, 38(19), 7830-7838.
16) Sadooghi, P. and Rauch, R. 2013. Pseudo heterogeneous modeling of catalytic methane steam reforming process in a fixed bed reactor. Journal of Natural Gas Science and Engineering 11, 46-51.
17) Xie, D., Qiao, W., Wang, Z., Wang, W., Yu, H. and Peng, F. 2010. Reaction/separation coupled equilibrium modeling of steam methane reforming in fluidized bed membrane reactors. International Journal of Hydrogen Energy, 35(21), 11798-11809.
18) Ye, G., Xie, D., Qiao, W., Grace, J.R. and Lim, C.J. 2009. Modeling of fluidized bed membrane reactors for hydrogen production from steam methane reforming with Aspen Plus. International Journal of Hydrogen Energy, 34(11), 4755-4762.
19) Yu, W., Ohmori, T., Kataoka, S., Yamamoto, T., Endo, A. and Nakaiwa, M. 2008. A comparative simulation study of methane steam reforming in a porous ceramic membrane reactor using nitrogen and steam as sweep gases. International Journal of Hydrogen Energy, 33(2), 685-692.