



ISSN: 0974 - 0376

The Ecoscan : Special issue, Vol. VIII: 189-192: 2015
AN INTERNATIONAL QUARTERLY JOURNAL OF ENVIRONMENTAL SCIENCES
www.theecoscan.in

STUDY OF HYDROGEN PRODUCTION FROM BIO-METHANE REFORMING PROCESS

M. M. C. Rajivgandhi

KEYWORDS

Hydrogen
Methane
Steam reforming
Biogas
Synthesis gas

**Proceedings of National Conference on
Harmony with Nature in Context of
Bioresources and Environmental Health
(HARMONY - 2015)**
November 23 - 25, 2015, Aurangabad,
organized by
Department of Zoology,
Dr. Babasaheb Ambedkar Marathwada University
Aurangabad (Maharashtra) 431 004
in association with
NATIONAL ENVIRONMENTALISTS ASSOCIATION, INDIA
www.neaindia.org



M. M. C. RAJIVGANDHI

Department of Agriculture Engineering,
RVS Group of Institutions, Sulur, Coimbatore- 641 402, Tamil Nadu, INDIA
e-mail: rajivgandhi.mmc@gmail.com

ABSTRACT

A simulation model of bio-methane reforming has been developed, analysed and optimized using Aspen Plus to determine suitable conditions for the hydrogen production. Simulations were performed as a function of process temperature *viz.* 500 to 900 °C, pressure 1, 2 and 3 bar and steam to carbon ratio 2, 3 to 4. The highest hydrogen content of 71 per cent is obtained in the reformed gas, when the optimum reactor temperature, reactor pressure, steam to carbon ratio are 775 °C, 1 bar and 2:1 respectively. The highest H₂ in the reformed gas results the bio-methane reforming process is a technically and economically feasible technology for hydrogen production.

INTRODUCTION

Hydrogen (H₂) is an attractive energy carrier, because it can be combusted similar to gasoline and natural gas, or converted to electricity in a fuel cell without any carbon emissions at the point of use (Simpson and Lutz, 2007). Currently 80–85 % of the world's total hydrogen production is derived via steam methane reforming (SMR) of natural gas, which is a source of significant CO₂ emissions into atmosphere. The global warming potential of hydrogen production via the SMR process is 13.7 kg CO₂ (equiv.) per kg of net hydrogen produced (Lau *et al.*, 2011). The production of H₂ from renewable sources such as biogas helps to largely reduce green-house gas emissions.

Biogas is a high-potential versatile raw material for reforming processes, which can be used as an alternative to the natural gas. The biogas contains 50 – 55 % of methane, 40–45 % of CO₂ and H₂S in traces. One of the major obstacles for biogas reforming is presence of CO₂ and H₂S. Therefore, instead of reforming biogas directly, after scrubbing of CO₂ and H₂S, hydrogen can be produced. Up to 97% of bio-methane can be obtained using biogas-upgrading system (Rajivgandhi and Singaravelu, 2014).

In most of the studies found in the literature on biogas reforming to produce H₂, the high purity CH₄ (> 99 %) is commonly used to simulate laboratory scale reforming processes (Zhu *et al.*, 2012), with few studies found that use biogas from the direct digestion process. Roy *et al.*, 2015 reported that the steam biogas reforming process was studied using the Aspen Plus process simulation tool. Model biogas (60% CH₄ + 40% CO₂) mixed with steam at S/C (steam to methane) ratios of 1.25, 1.50 and 1.75 was used as the feedstock. Steam biogas reforming equilibrium data were obtained over a temperature range of 649 to 849 °C at 1 bar pressure through Gibbs free energy minimization. Aspen Plus simulation results will help to enable the Steam biogas reforming process optimization with a focus on improving the net thermal efficiency and product syngas yield. However, steam biogas reforming technology currently used encounter numerous technical/scientific challenges, which depend on the quality of raw materials, the conversion efficiency and security needs for the integration of H₂ production, purification and use, among others.

Alves *et al.*, 2013 reported about the most common methane reforming processes for hydrogen production that are, Steam Methane Reforming (SMR), Partial Oxidation Reforming (POR), Auto-Thermal Reforming (ATR), Dry Reforming (DR) and Dry Oxidation Reforming (DOR). Among the reforming technologies to produce hydrogen, it was noticed that no study has been done for the biomethane reforming process. Therefore, it is aimed in this work to use biomethane as a raw material for the production of hydrogen.

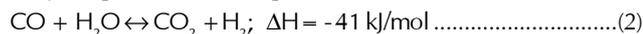
Nowadays, it is considered that to find a suitable pathway to produce hydrogen from biogas through biomethane could represent a new step on the progress. The present work deals the study of H₂ production from bio-methane and optimization of various parameters of the bio-methane reforming process.

*Corresponding author

MATERIALS AND METHODS

Aspen Plus (V 7.3) software was used to predict the operating parameters for the H₂ production such as reactor temperature, S/C ratio and pressure. Fig. 1 shows the model of the bio-methane reforming process.

The chemical reactions involved in the steam reforming of methane considered in this work are given as shown in Equations (1) and (2).



Equation (1) is the reforming reaction while Equation (2) is the water-gas-shift reaction of the process. Both reactions were simulated simultaneously in Aspen Plus.

Simulations were performed using R-Gibbs fixed bed reactor to determine the composition reformed gas as a function of process temperature, steam to carbon ratio and pressure. To determine product gas composition Peng Robinson equation of state was adopted (Galvagno *et al.*, 2013).

The ranges of the operating variables used for the simulation analyses are given in Table 1.

RESULTS AND DISCUSSION

The abstracted results of simulated Bio- methane reforming process, showing the components of the reformed gas in mole fraction percentage are given in Table 2.

In the bio-methane reforming process, H₂ yield of 71 % have been obtained at the temperatures' ranging between 750 and 850 ° C. The results obtained in this study is on par with Boyano *et al.*, 2011 and Zhai *et al.*, 2011, where they obtained up to 60-70 % H₂ yields in the temperatures ranging between 650 and 850 °C.

Effect of reactor temperature on H₂ production

Fig. 2 shows the effect of reactor temperature on the mole fractions of the reformed gas.

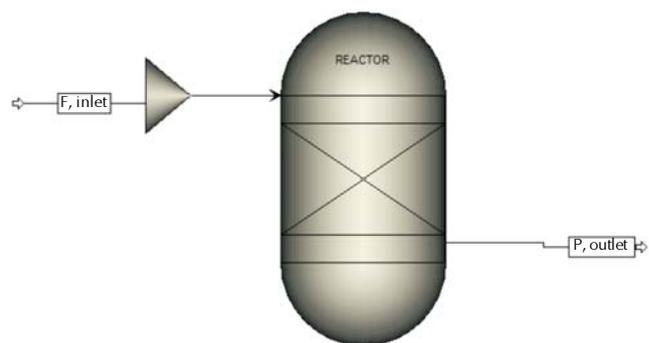
It can be noticed from the figure that the mole fractions of CH₄

decrease with the increase in the reactor temperature, while carbon monoxide and water increase with the increase in the reactor temperature. Moreover, the higher temperature of the reactor increases the CO content in the reformed gas. It indicates the reactor temperature in the range of 800 to 900°C favours the occurrence of reverse water gas shift reaction and due to this reaction H₂ and CO₂ contents in the reformed gas decreases. As the temperature increases, the hydrogen yield in the reformed gas increases and beyond 800°C, it decreases. Further, this observation is in agreement with the finding that water-gas-shift reaction is favoured at the low temperature of 500 to 800°C, which is reported in the Galvagno *et al.*, 2013.

Effect of pressure on H₂ production

The results obtained from the simulation analysis on the effect of pressure on the mole fractions of the components present in the product of the reactor are shown in Fig. 3.

It is observed from the Fig. 3 that the mole fractions of CO and H₂ decrease with the increase in the reactor pressure, while those of CH₄, H₂O and CO₂ increase, as the reactor pressure increases. Even though the decrease in the production of CO with the increase in the reactor pressure is favourable, the



F, inlet - Feed inlet (Bio-Methane + Steam); P, outlet - Reformed gas outlet

Figure 1: Model of Bio-Methane reforming process

Table 1: Ranges of operating variables used in the simulation analyses

S.No	Parameters	Lower range	Upper range	Increment
1.	Temperature, °C	300	900	25
2.	Pressure, bar	1	5	0.25
3.	Mole flow rate of H ₂ O, mol h ⁻¹	32	64	16
4.	Mole flow rate of CH ₄ , mol h ⁻¹	16		

Table 2: Abstracted results of simulated Bio- methane reforming process

S.No.	Temp.°C	Pre. (bar)	Mole flow of CH ₄ , (mol/h)	Mole flow of H ₂ O, (mol/h)	CH ₄ , (%)	H ₂ O, (%)	CO, (%)	H ₂ , (%)	CO ₂ , (%)
1.	750	1.00	16	32	0.56	5.97	16.61	71.45	5.40
2.	775	1.00	16	32	0.31	5.98	17.12	71.55	5.05
3.	775	1.25	16	32	0.47	6.05	17.01	71.39	5.09
4.	800	1.25	16	32	0.26	6.06	17.45	71.45	4.77
5.	800	1.50	16	32	0.37	6.11	17.38	71.34	4.80
6.	825	1.00	16	32	0.10	6.08	17.85	71.49	4.49
7.	825	1.50	16	32	0.21	6.13	17.77	71.37	4.51
8.	850	1.00	16	32	0.06	6.15	18.13	71.41	4.25
9.	850	1.25	16	32	0.09	6.16	18.11	71.38	4.26
10.	850	1.50	16	32	0.12	6.18	18.09	71.34	4.27

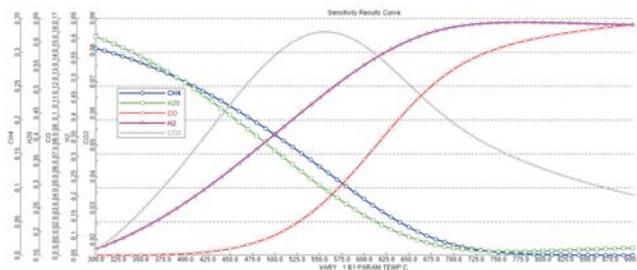


Figure 2: Effect of reactor temperature on the mole fractions of reformed gas

same decreasing trend of hydrogen with respect to the increase in the reactor pressure is unfavourable, because a very high mole fraction of H_2 in the reformed gas is desired. It can be concluded that bio-methane feed produces a high mole fraction of H_2 in 1 to 2 bar pressure. This is in agreement with the report given by Chiodo *et al.*, 2012 and Galvagno *et al.*, 2013.

The highest H_2 content of 71 per cent is observed in the reformed gas, when the optimum reactor temperature, reactor pressure, volumetric flow rate of feed steam and volumetric flow rate of feed methane are 775 °C, 1 bar, 8.3 L min⁻¹ and 16.6 L min⁻¹ (2:1) respectively. Hence, it can be concluded that the temperature of 750 to 850 °C, S/C ratio of 2 and pressure 1 bar can be taken as optimum for the Bio- methane reforming process. This result is comparable with the report given by Izquierdo *et al.*, 2012 that for the S/C ratio of 2, the maximum hydrogen content is 67.2 %.

ACKNOWLEDGEMENT

The author thank the Council of Scientific and Industrial Research (CSIR) and Indian Council of Agricultural Research (ICAR), New Delhi, for providing financial support and Tamil Nadu Agricultural University for providing necessary laboratory facilities and support.

REFERENCES

Alves, H. J., Junior, C. B., Niklevicz, R. R., Frigo, E. P., Frigo, M. S. and Araújo, C. H. C. 2013. Overview of hydrogen production technologies from biogas and the applications in fuel cells. *Int J. Hydrogen Energ.* **38(13)**: 5215 - 5225.

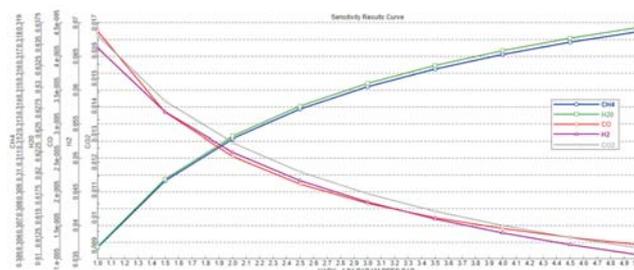


Figure 3: Effect of reactor pressure on the mole fractions of reformed gas

Boyano, A., Blanco-Marigorta, A. M., Morosuk, T. and Tsatsaronis, G. 2011. Exergoenvironmental analysis of a steam methane reforming process for hydrogen production. *Energ.* **36**: 2202-14.

Chiodo, V., Urbani, F., Galvagno, A., Mondello, N. and Freni, S. 2012. Analysis of biogas reforming process for molten carbonate fuel cells. *J. Power Sources.* **206**: 215-21.

Galvagno, A., Chiodo, V., Urbani, F. and Freni, F. 2013. Biogas as hydrogen source for fuel cell applications. *Int J Hydrogen Energ.* **38**: 3913-3920.

Izquierdo, U., Barrio, V.L., Lago, N., Requies, J., Cambra, J.F. and Guemez, M. B. 2012. Biogas steam and oxidative reforming processes for synthesis gas and hydrogen production in conventional and microreactor reaction systems. *Int. J. Hydrogen Energ.* pp.1-14.

Lau, C. S., Tsolankis, A. and Wyszynski, M. L. 2011. Biogas upgrade to syn- gas (H_2 -CO) via dry and oxidative reforming. *Int J. Hydrogen Energ.* **36**: 397-404.

Rajivgandhi, M. M. C. and Singaravelu, M. 2014. Upgrading Biogas to Biomethane by Physical Absorption Process. *International J. Agriculture, Environment and Biotechnology.* **7(3)**: 639-644.

Roy, P. S., Raju, A. S. K., Park, C. S and Kim, K. 2015. Aspen Plus Simulation of Steam-Biogas Reforming and Comparison with Experimental Data Obtained over a Metal-Foam-Coated Pd-Rh Catalyst, AIChE Spring Meeting and 11th Global Congress on Process Safety.

Simpson, A. P. and Lutz, A. E. 2007. Exergy analysis of hydrogen production via steam methane reforming, *Int. J. Hydrogen Energ.* **32**: 4811-4820.

Zhai, X., Ding, S., Liu, Z., Jin, Y. and Cheng, Y. 2011. Catalytic performance of Ni catalysts for steam reforming of methane at high space velocity. *Int J. Hydrogen Energ.* **36**: 482-9.

Zhu, X., Wang, H, Wei, Y, Li, K. and Cheng, X. 2012. Hydrogen and syngas production from two-step steam reforming of methane over CeO_2 - Fe_2O_3 oxygen carrier. *J. Rare Earths.* **28(6)**: 907-12.