

Production of Pure Hydrogen in Membrane Reactor

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Abstract—Hydrogen as a carbon-less energy carrier is playing an important role in meeting energy demands. It has extensive applications in laptops, Hydrogen vehicles, Electricity production and space shuttle fuels production. Hydrogen production is viable through non-renewable resources including Fossil fuels and electrochemical methods and renewable resources. One of the most important hydrogen production processes is steam methane reforming (SMR) by which hydrogen is separated from syngas along with carbon monoxide and carbon dioxide. Membrane reactors are integrated systems of reaction and separation and are used specifically to increase the rate of reaction, selectivity applications and their pure production. Multichannel membrane reactor (MCMR) is developed to utilize the systems with decentralized hydrogen production through steam methane reforming. In these reactors, the channel in which methane catalytic combustion (MCC) would take place is adjacent to the channel in which SMR that is very endothermic is taking place so it would produce the heat which is needed for the reaction. Pd-Ag membrane in reforming channel would change the balance of the reaction and results in pure hydrogen production in a single vessel. In this research, 1D modeling of multichannel membrane reactor to separate Hydrogen from Syngas through SMR is been studied. Results show that the conversion percentage of methane in SMR channel is %81, while it's in %90 in combustion channel. Also, hydrogen with 99.99% purity is accessible through Pd-Ag membrane.

Keywords: Steam methane reforming, Hydrogen, Membrane Reactor, Combustion, Modeling

I. INTRODUCTION

The rapid development of the industrial and transport sectors and improved living standards has led to an increase in global energy consumption. Today the energy systems are mainly based on crude oil, natural gas and coal. All of these sources of energy are nonrenewable while the use of these energy sources increases the concentration of greenhouse gases (such as CO₂) and global temperature rise.

In recent years, hydrogen as an alternative to fossil fuels has been focused. Hydrogen has the highest energy content among the fuels but it has the lowest normal pressure for gas in terms of volume. Hydrogen can save energy and it is a carbon-free energy carrier. Hydrogen is the most abundant element in the

universe and when it is used in a fuel cell or an internal combustion engine, its product is a pollution-free energy and water as a byproduct. The use of fuel cells

is developing in the automobile industry and to produce energy in power plants.

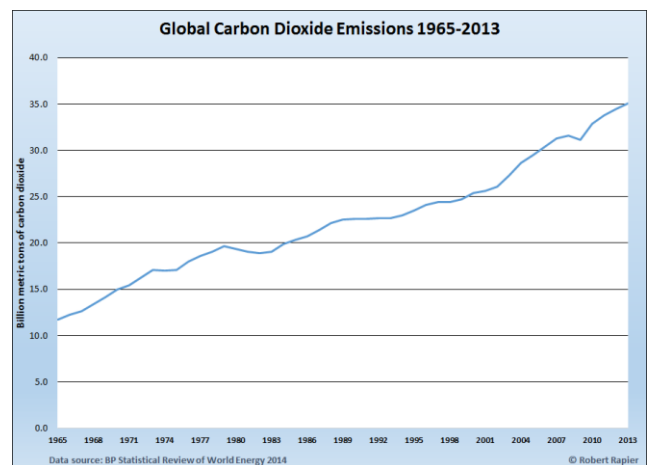


Figure 1. Global carbon dioxide emissions [2].

An advantage of hydrogen as an energy source is its flexibility. While fossil fuels are limited to specific geographic areas, hydrogen production is possible in almost any location because hydrogen can be produced from fossil and non-fossil fuels.

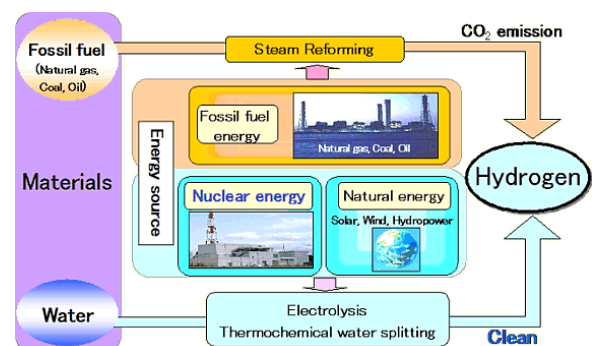


Figure 2. Hydrogen Generation Methods [3].

Today, 95% of produced hydrogen is through 95% the most important part of which is

produced by reforming methane [4]. The most important method of producing syngas is steam methane reforming (SMR). The SMR process can be designed such that the main purpose of which is to produce hydrogen. In this case the produced syngas a large amount of hydrogen [5]. There are various methods for separating hydrogen from syngas the most important of which includes:

- Amine absorption (removing CO₂)
- Pressure swing adsorption (PSA)
- Cryogenic distillation
- Membrane separation

In general, steam methane reforming (SMR) is a reaction which requires high energy consumption and because the reactions are endothermic and the need for proper reforming percentage the gas should be provided with enough energy so that the reaction would move towards equilibrium. The analyses conducted by researchers have shown that if the process is carried out in a membrane reactor, the required operating conditions such as temperature and pressure are adjusted for the reaction and hydrogen can be removed from the system selectively and the production reactions' thermodynamic equilibrium is converted to production which leads to achieve high conversion percentage even at lower operation temperature.

Yu-Ming Lin et al have modeled and studied the impact of removing hydrogen from the palladium membrane based on methane conversion in the SMR process experimentally the results of which suggest that the use of palladium membranes leads to achieving the conversion percentage of higher than 80% [6].

Genyin Ye et al have modeled hydrogen production in a fluidized bed membrane reactor in ASPEN PLUS Software and it is observed that removing hydrogen in the location leads to the equilibrium shift [7].

One-dimensional SMR process modeling is done for hydrogen production in PBR and MR reactors in STEADY STATE and ISOTHERMAL conditions by Jean-François ROUX and compared with each other the results of which indicate that the existence of a membrane in the process modifies the reaction conditions and leads to the achievement of higher conversion rates and finally accessing the methane conversion to 95% at a pressure of 16 bar and the temperature of 600°C [8].

Fausto Gallucci et al have studied the impact of using a sweep gas on methane conversion the results of which indicates that using a sweep gas such as steam in an MR reactor that could be easily separated from hydrogen increases methane conversion at lower temperatures

because this reduces the hydrogen concentration in the membrane and shifts equilibrium to higher methane conversion [9].

The basic concept of multi-channel membrane reactor is used for dehydrogenation. The multi-channel membrane reactor with palladium membrane is proposed for water-gas shift reaction. Multi-channel membrane reactors are usually described as a plate, wall and micro-channels to produce hydrogen with high efficiency. The use of these reactors is followed by reducing the economic costs.

The main idea of these reactors is to follow the condensing heat exchanger's design. The cold channels in which the endothermic reforming reaction is ongoing are located in the vicinity of the warm channels in which exothermic reaction takes place.

The catalyst usually coats the channel walls. The multi-channel reactor increases heat transfer surface and the hydrogen production level compared to the reactor volume.

Increased scale in these reactors is possible by repeating the pattern of the mentioned channels while the reaction physics and hydrodynamics remain as before.

Accordingly A. Vigneault et al have performed the 2D modeling of a multi-channel membrane reactor under steady state in both isothermal and non-isothermal for producing hydrogen by SMR process in MATLAB environment [5].

In the present study the purpose is to combine SMR and MCC processes in STEADY-STATE and non-isothermal conditions to produce hydrogen gas. Also in this modeling steam is used as a sweep gas to modulate reaction conditions and increase the methane conversion. Multi-channel membrane reactors are developed by steam methane reforming (SMR). In this reactor the channel where methane combustion reaction takes place is located in the vicinity of the channel in which endothermic steam methane reforming takes place to provide the heat required for the reaction. These two channels are separated by a wall the main purpose of which is to transfer heat between the two channels. Palladium-silver membrane which has 99% selectivity to hydrogen is located above the reforming channel and shifts equilibrium. Mathematical modeling of SMR to produce hydrogen in a packed bed SMR is done as one-dimensional in the constant pressure and non-isothermal mode.

Flow regime is considered as expanded laminar to facilitate solving the momentum equations and also this current is considered as co-current to reduce the temperature changes.

The catalyst in the process of burning methane is palladium and alumina based nickel in SMR. In both channels the catalyst has coated the channel wall. In this modeling there two channels

that each channel includes a gas phase and is a catalyst layer.

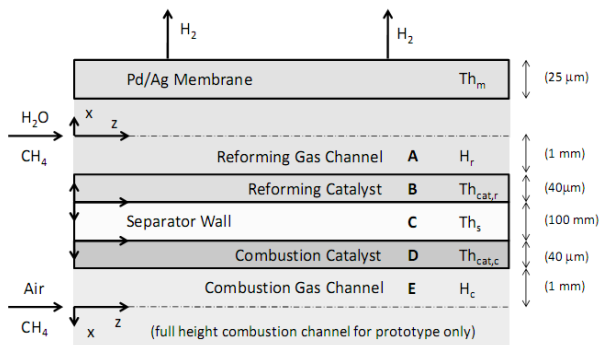


Figure 3. Schematic of 1-D Model, including base case dimensions, not to Scale. (Subscripts: Pd/Ag membrane m ; SMR channel r ; catalyst layer cat ; separator wall s ; MCC channel c .) [5].

Basic assumptions considered for modeling the mentioned process are as follows:

Basic modeling assumptions

- Gas behavior is assumed as ideal
- The process is assumed as adiabatic and there is no heat loss in the environment
- Coke formation is ignored
- Pressure drop is ignored
- Fourier's law is applied for the thermal conductivity
- Fick's law is applied for infiltration

II. MODELING

Accordingly mathematical 2D modeling of SMR process to produce hydrogen in a packed bed multi-channel membrane reactor under the STEADY STATE and ISOTHERMAL conditions are investigated. To solve the modeling, differential equations including mass equilibrium, energy equilibrium for the reactant, reaction kinetics and physical properties are written and obtained by the simultaneous solution of equations the concentration and temperature distribution in the reactor was obtained. Flow regime Flow regime is considered as expanded laminar to facilitate solving the momentum equations. The common differential models by the relevant boundary conditions are solved in MATLAB and the flow and mole fraction of each material is obtained based on the reactant and product components as well as changes in temperature in the reactor. Based on the rate equations for reactions the raw material including methane and steam are used along the reactor and their value is reduced.

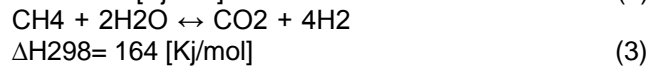
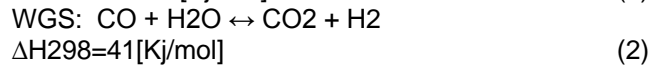
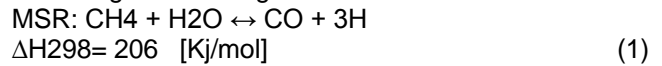
To validate the model and ensure the accuracy of results the results were compared with existing experimental data. The effective operating parameters include the temperature of the input feed to the

reactor, steam ratio and reactor length the effect of which was analyzed on methane conversion and the results were reported. Here the equations, boundary

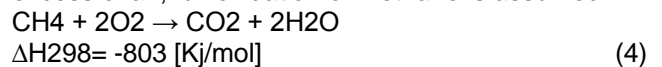
conditions and the materials for MCMR one-dimensional modeling will be presented and the schematic of the process is presented in Figure 3.

A. Kinetics

For the reforming catalyst, methane steam reforming and water-gas-shift reactions are:



For the combustion catalyst, given the stoichiometric excess of air, full oxidation of methane is assumed.



B. Physical Properties

• Diffusivity

For gas mixtures, it is assumed:

$$D_{i,mix} = \phi_{ij} \quad (5)$$

• Heat Capacity

The heat capacity relationship for a single component CP_i as a function of temperature T , neglecting pressure effect, was determined (Sandler, 1999) as:

$$C_{pi} = A_i + B_i T + C_i T^2 + D_i T^3 \quad (6)$$

• Thermal Conductivity

For the thermal conductivity in the gas channels, we neglected the effect of pressure and used the following relation (Bird et al., 2002) for K_{mix} of a gas mixture:

$$k_{mix} \sum_i^n \frac{y_i k_i}{\sum_j y_j \phi_{ij}} \quad (7)$$

• Viscosity

The viscosity of the mixture was estimated (Bird et al., 2002) from:

$$\mu_{mix} \sum_i \frac{y_i \mu_i}{\sum_j y_j \phi_{ij}} \quad (8)$$

$$\mu_i = \mu_i^{ref} \left(\frac{T}{T^{ref}} \right)^\beta \quad (9)$$

C. Component Material and Energy Balance Equations

Based on the assumption of ideal gases, we write:

$$c = \frac{P}{R_g T} \quad (10)$$

$$P_i = y_i P \quad (11)$$

D. Velocity Profiles

Assuming fully developed laminar flow in the gas channel, the velocity profile in the axial direction V_z is assumed to follow the two-dimensional relation (Bird et al., 2002):

$$v_z = 1.5v_{ave,z} \left(1 - \left(\frac{x}{H_k}\right)^2\right) \quad (12)$$

Were integrated:

$$\int_{-H_k}^{H_k} c v_z dx \Big|_{z=0} - \int_{-H_k}^{H_k} c v_z dx \Big|_z + \int_0^z \int_0^{Th_{cat,k}} \sum_i R_i dx dz + \int_0^z J_{H_2,m} dz = 0 \quad (13)$$

Here $Th_{cat,k}$ is the catalyst layer thickness. R_i is the rate of production of component defined as:

$$R_i = \sum_j \sigma_{ij} r_j \quad (14)$$

The rate of reaction R_i is defined in section A. σ_{ij} is the stoichiometric coefficient of component i for reaction j . $J_{H_2,m}$ is the molar flux of hydrogen through the membrane, defined based on Sievert's law:

$$J_{H_2,m} = -\eta_m \frac{A_m}{Th_m} \exp\left(\frac{-E_m}{R_g T_m}\right) (\sqrt{P_{H_2,r}} - \sqrt{P_{H_2,m}}) \quad (15)$$

E. Component Material and Energy Balance Equations

The general equation of continuity at steady state for a component i in a mixture (adapted from Bird et al. (2002)) is:

$$-(\nabla \cdot c_i v^{\rightarrow}) - (\nabla \cdot J_i^{\rightarrow}) + R_i = 0 \quad (16)$$

From Fick's law of diffusion:

$$J_i^{\rightarrow} = -D_{i,mix} \nabla c_i \quad (17)$$

We developed the general energy balance equation elsewhere:

$$\nabla T \cdot \sum_i (C_{pi} c_i v^{\rightarrow}) + \nabla T \cdot \sum_i (C_{pi} J_i^{\rightarrow}) + \nabla \cdot q^{\rightarrow} + \sum_j \Delta H_{ix} r_j = 0 \quad (18)$$

There are four heat transfer/generation terms in Eq. (18), representing in order: (1) transfer by convection, (2) transfer by diffusion, (3) transfer by conduction, and (4) heat generation due to reactions. Assuming Fourier's law of conduction, we write q^{\rightarrow} as:

$$q^{\rightarrow} = -k_{mix} \nabla T \quad (19)$$

To use MATLAB built-in functions, it was necessary to reduce the order of the differential equations to one for both concentration and temperature. To overcome this limitation, we used first order transformations in the transverse direction:

$$c_i = c_{1i} \quad (20)$$

$$\frac{\partial c_i}{\partial x} = c_{2i} \quad (21)$$

$$T = T_1 \quad (22)$$

$$\frac{\partial T_1}{\partial x} = T_2 \quad (23)$$

From this point on, C_i is referred to as C_{1i} , and T becomes T_1 . Backward difference discretization was applied in the axial direction for any dependent parameter u :

$$\frac{\partial u_b}{\partial z} = \frac{1}{\Delta z_{b-1}} (u_b - u_{b-1}) \quad (24)$$

F. Gas Phase Material balance $b \neq 1$

$$\frac{dc_{2i}}{dx} = \frac{1}{D_{i,mix,b}} (v_{z,b} \frac{1}{\Delta z_{b-1}} (c_{1i,b} - c_{1i,b-1}) + c_{1i,b} \frac{1}{\Delta z_{b-1}} (v_{z,b} - v_{z,b-1})) \quad (25)$$

Energy balance $b \neq 1$

$$\frac{dT_{2,b}}{dx} = \frac{1}{k_{mix,b}} \left\{ \frac{1}{\Delta z_{b-1}} (T_{1,b} - T_{1,b-1}) \sum_i (C_{pi,b} c_{1i,b} v_{z,b}) - T_{2,b} \sum_i (C_{pi,b} D_{i,mix,b} c_{2i,b}) \right\} \quad (26)$$

G. Catalyst Layer

Material balance $b \neq 1$

$$\frac{DC_{2i,cat,kb}}{dx} = \frac{1}{D_{i,eff,b}} (-R_{i,b}) \quad (27)$$

Energy balance $b \neq 1$

$$\frac{dT_{2cat,k,b}}{dx} = \frac{1}{k_{cat,k,b}} \left\{ (-T_{2cat,k,b}) \sum_i (C_{pi,cat,k,b} D_{i,eff,b} c_{2i,cat,k,b}) + \sum_j \Delta H_{rx,j} r_j \right\} \quad (28)$$

H. Separator wall

Assuming Neglecting heat conduction in the axial direction will be

$$\frac{dT_{2,s,b}}{dx} = 0 \quad (29)$$

$$\frac{dT_{2,s,b}}{dx} = 0 \quad (30)$$

III. BOUNDARY CONDITIONS

With the same assumptions listed earlier, performing energy and mass balances across interfaces, the continuity principle, and feed conditions at the inlets (subscript o), the boundary conditions are obtained.

A. Gas Phase

Gas channel - inlet conditions ($z = 0$ and $b = 1$) at the entrance of the reactor, the temperature T_1 is assumed constant at the feed condition:

$$T_{1,b=1} = T_{1,0} \quad (31)$$

$$T_{2,b=1} = 0 \quad (32)$$

B. Gas channel - catalyst interface

$$\sum_i J_{i,x,k} H_{i,x} \Big|_{x=-H_k} = \sum_i J_{i,x,catk} * H_{i,cat,k} \Big|_{x=-Th_{cat,k}} \quad (33)$$

$$T_{2,k,b} \Big|_{x=-H_k} = \frac{k_{cat,k,b} T_{2,catk}}{k_{mix,b}} \quad (34)$$

C. Gas channel - membrane interface

$$T_{2,r} \Big|_{x=H_r} = 0 \quad (35)$$

D. Catalyst Layer

$$\frac{dT_{1,catk,b}}{dx} = T_{2cat,k,b} \quad (36)$$

E. Catalyst layer - inlet conditions

$$T_{1cat,k,b=1} = T_{1,k,b=2} \quad (37)$$

F. Catalyst - gas channel interface

$$T_{1cat,k} \Big|_{x=Th_{cat,k}} = T_{1,k} \Big|_{x=-H_k} \quad (38)$$

G. Separator Wall

At the entrance of the reactor, we assume no gradient in the separator wall in the axial direction, therefore:

1. Separator wall - inlet conditions:

$$T_{1,s,b=1} = T_{1,s,b=2} \quad (39)$$

2. Reforming catalyst interface:

$$T_{1,s,b} \Big|_{x=0} = T_{1,cat,r,b} \Big|_{x=0} \quad (40)$$

3. Combustion catalyst interface:

$$T_{1,s,b} \Big|_{x=Th} = T_{1,cat,r,b} \Big|_{x=0} \quad (41)$$

IV. RESULT

In this modeling the changes in temperature and methane conversion percentage, molar flow of the reactants and products to the reactor length and the molar fraction changes of the products and raw materials is assessed along the reactor in the presence of SMR channel. Also changes in heat flux are compared in both MCC and SMR channels along the channel. The hydrogen produced in the reactor is increased slightly at the beginning of the reaction but its flow is reduced during the infiltration in the membrane and transfer to hydrogen collection area and the flow of hydrogen is increased in the infiltration sector. The reason for rapid increase in hydrogen at the beginning of the reactor is the purity of raw materials. With the advance in the reactor, the raw materials are reduced and the amount of carbon dioxide is increased which increases the rate of hydrogen production. On the other hand the amount of carbon monoxide in the reactor remains insignificant

because the produced carbon monoxide always reacts with water due to higher steam flow than the input

methane and is decomposed to hydrogen and carbon dioxide and improves the hydrogen production efficiency.

At the beginning of the reactor the high purity of the raw materials and high temperatures increase the methane conversion percentage and then the severity of the conversion is reduced over time along the reactor. By increasing the length of the reactor the pressure drop increases in the reactor.

By increasing the length of the reactor, the hydrogen produced by the reactions is increased the reason of which is the higher rate of hydrogen

production than its infiltration. By increasing the hydrogen pressure in the reactor, the hydrogen passage through the membrane is increased and by increasing the length the hydrogen collection by sweep gas is improved and the hydrogen flow in the in the reactor tends to constant value and produced hydrogen will be removed from the reactor by passing through the membrane by the sweep gas. For this reason hydrogen recovery increases in the infiltration sector.

There are various effective parameters that affect methane conversion by steam. The increase in the pressure or the sweep gas increases the pressure in the infiltration area and this increase is considered as resistance against hydrogen infiltration through the reactor to the area. Increasing concentration of hydrogen in the reactor reduces the methane conversion percentage. If resistance to hydrogen infiltration through the membrane increases, it leads to the retention of hydrogen in the reactor. As a result by increasing the increasing concentration of hydrogen in the reactor according to Le Chatelier's Principle methane conversion decreases. Thus the sweep gas pressure has an opposite effect on methane conversion level i.e. by increasing the gas pressure in the hydrogen collection area, methane conversion decreases.

Since methane conversion reactions are endothermic reactions, temperature and heat have a major impact on conversion rate. For this reason, heating the reactor input feed through pre-heaters increases the percentage of methane conversion.

As can be seen in Figure 4 at the beginning of the reactor temperature is 873K and it is increased to 890K at 5 cm away from the reactor length because in the MCC channel the concentration of reactants is higher at the beginning of reactor and it is followed by methane burning reaction and thus the release of higher heat. Then since the SMR reaction is endothermic, the heat along the reactor is consumed to convert methane and the temperature at the end of reactor is reduced to 780k.

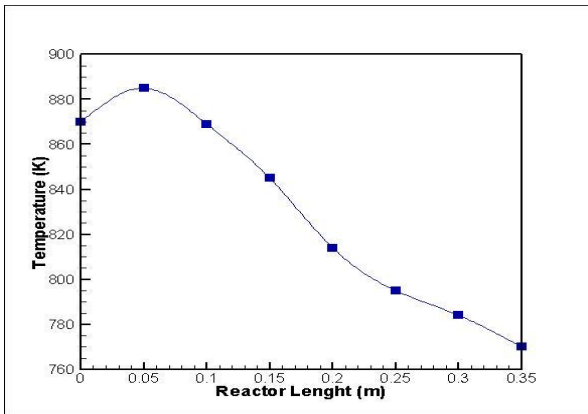


Figure 4: The temperature changes along the reactor in SMR channel

In Figure 5 it can be seen that the conversion of methane along the reactor has an upward trend and methane conversion percentage reaches 81% at the

length of 35cm. This value is increased 7% compared to [5].

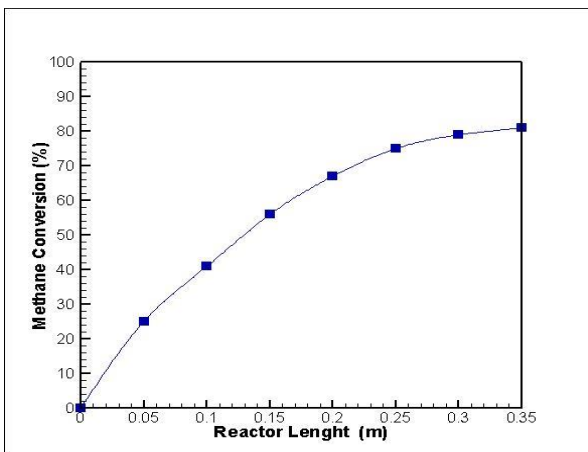


Figure 5: Changes in the methane conversion percent along the reactor in SMR channel

The molar downward trend rate along the reactor can be seen in Figure 6. This value starts at 3.6 moles per second and finally reaches 1.8 moles per second at the end of the reactor which indicates that there is a need for a backflow for methane.

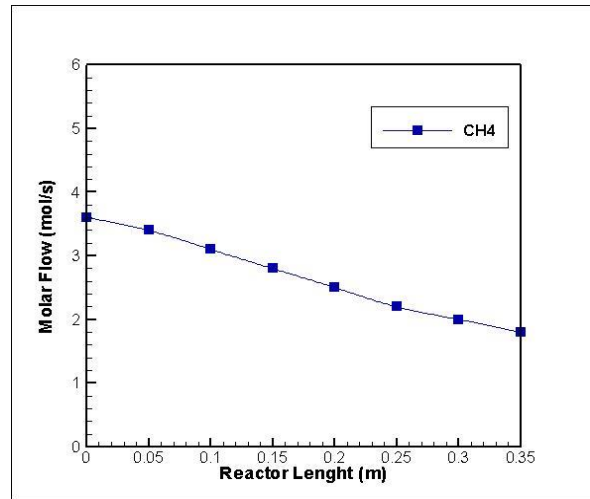


Figure 6: Molar rate changes of methane along the reactor in SMR channel

Based on the reactions (1), (2) and (3) water is constantly being consumed during the reaction and Figure 7 presents the downward trend of molar ratio of water along the reactor which starts at 1.8 moles per second and ends at 0.5 moles per second.

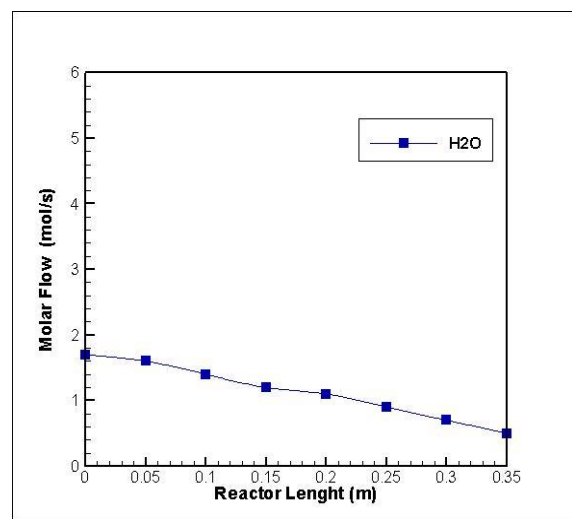


Figure 7: Molar rate changes of water along the reactor in SMR channel

According to equation (1) carbon monoxide is produced and then according to equation (2) carbon monoxide reacts with water and converted to hydrogen and carbon dioxide and only a low amount of it remains in the channel and a low growing trend is observed in Figure 8. This amount is 0.9 moles per second. The amount of carbon monoxide in the reactor remains negligible because due to higher steam flow to the input methane the produced carbon monoxide always reacts with water and is converted to carbon dioxide and hydrogen and improves hydrogen production efficiency.

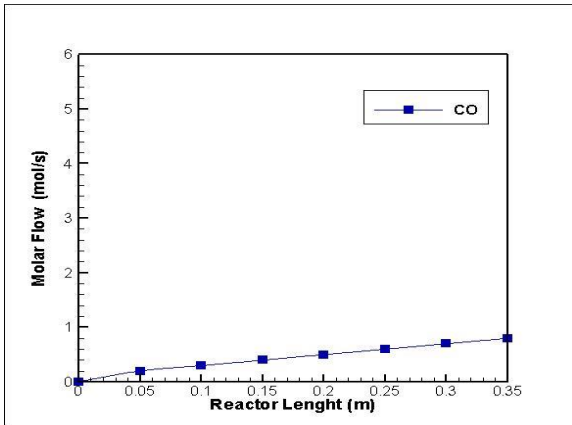


Figure 8: Molar rate changes of carbon monoxide along the reactor in SMR channel

It can be seen in Figure 9 that the amount of carbon dioxide along the reactor has an increasing trend.

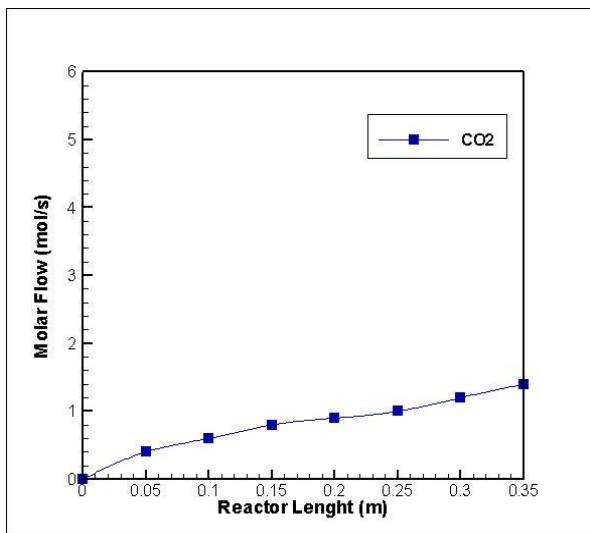


Figure 9: Molar rate changes of carbon dioxide along the reactor in SMR channel

The amount of hydrogen in the beginning of the reaction is zero and it gradually increases along the reactor. Hydrogen is produced by three reactions (1), (2) and (3). At the end of the reactor the molar rate reaches 2.1 moles per second and its concentration increases. Therefore in such conditions there is a need for a sweep gas such as nitrogen in the membrane to reduce the concentration of hydrogen in the membrane and more hydrogen passes through the membrane and based on Le Chatelier's Principle the reaction shifts to higher methane conversion and thus more hydrogen production. Figure 10 presents the increasing molar rate of hydrogen along the reactor.

This value is 0 at the beginning and 2.4 Moles per second at the end of the reactor.

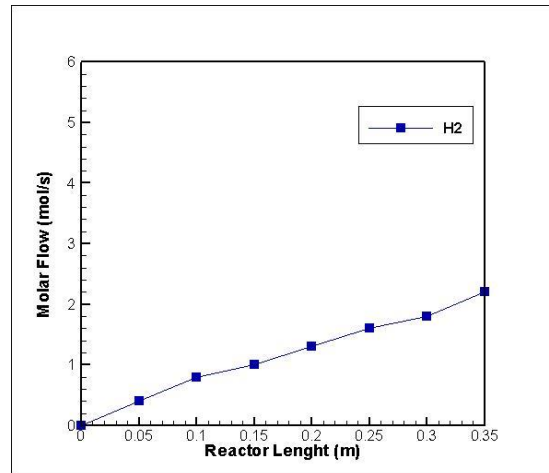


Figure 10: Molar rate changes of hydrogen along the reactor in SMR channel

Conclusion

The performed modeling is to understand the effect of operating and equipment parameters to improve the process. The purpose of the parameters' improvement is the higher production in optimum conditions. Heating the reactor is among the important factors in increasing hydrogen production. Increasing temperature along the reactor is the best condition to increase the conversion rate that is for reducing the purity of raw materials along the reactor, the heat to the reactor must be increased. Temperature is among the factors under consideration. SMR reaction due to being endothermic requires high temperatures, thus increasing the input feed increases the conversion rate.

In the present modeling temperature changes and the conversion of methane are studied in both hot and cold channels in which the temperature of burning methane at a maximum temperature of 890 ° K and according to the temperature required for the SMR reaction which is 873 K at the beginning the reactor with regard to the feed before entering the channel is heated by the pre-heaters and then sent to the reactor, it can be found that the temperature generated by the MCC channel may play a useful role in converting methane in the SMR channel. Temperature rise is limited because it deactivates the catalysts and creation of sintering process and it is better to perform the reaction at lower temperatures.

Another important parameter is the ratios of steam and swept gas to the input methane and the increase in both of them increases the conversion rate. Increasing water steam due to being the raw material to react with methane as the sweep gas reduced the hydrogen concentration in the membrane and cause more infiltration of hydrogen through SMR channel to the palladium membrane and thus based on the Le Chatelier's Principle the reaction moves towards more consumption of methane and more hydrogen production. Methane conversion percentage is

Predicted as 51 and 90% in SMR and MCC channels respectively.

Also considering the palladium membrane the selectivity of which in terms of hydrogen is approximately 99.99%, finally it is possible to achieve this purity.

According to Figure 6 it is found that the amount of converted methane in the reactor output can be retrieved by a backflow so that all methane in the feed could be converted to hydrogen as much as possible.

In this study, the use of membrane reactor leads to combining the reaction and separation in a single vessel which saves energy time and cost.

In addition to the mentioned factors the location of MCC channels in the vicinity of SMR channels to provide heat is important issue in better heat transfer with lower fuel consumption.

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