

Modeling of steam methane reforming (SMR) over a Ni-AL₂O₃ catalyst

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Abstract—In present paper modeling of natural gas reforming by steam over nickel-alumina catalyst is investigated and key parameters such as pressure, temperature and mole ratio have been studied. The results show that the reforming performance is affected by temperature, pressure and ratio of steam to methane. The equations for mass and energy balance equations and catalyst are mentioned.

The combination of carbon monoxide and hydrogen are known as syngas and are used as a feedstock in the production of various products such as ammonia and liquid fuel applications. There are different techniques for the production of synthesis gas. One of these techniques is dry reforming that combination carbon dioxide and methane are converted into synthesis gas.

Keywords—Natural gas, Reforming, Catalyst, Steam

After desulfurization, natural gas and carbon dioxide mixed with water and flue gases and pre-heated to the reaction temperature. Inside the reactor, there are long length and small diameter pipes with nickel-alumina catalyst contained. Reforming conditions with a high temperature furnace are converted all hydrocarbons into products. In this process, the required carbon dioxide usually achieved from returns all the carbon dioxide and also a significant amount of outside unit [2]. Process schematic is as follows:

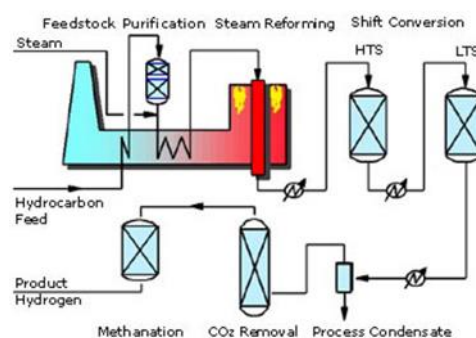
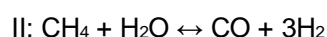
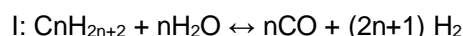


Figure 1- Process schematic [3]

Steam reforming technology was used from 1920 onwards, but from the 1950s onwards was used as a process for synthesis gas production of gas compounds. The key point of these reactions is endothermic that require high temperatures and decomposition reaction of H-C band is the first controller. As shown in the following equations C²⁺reactions (reaction I) are irreversible, but the decomposition of methane (reaction II) equilibrium relationship formed by the reaction of methane is returned to the reaction [4].



$$\Delta H_{II} = +206 \text{ kJ/mol}$$

I. Introduction

The synthesis gas is odorless and colorless poisonous gas that is a mixture of hydrogen and carbon monoxide and use in various industries including petrochemical and metallurgy industries. There are several methods for the production of synthesis gas from steam reforming of natural gas is mostly done with hydrocarbons such as naphtha. Natural gas that primarily containing methane, reacts with water within tubular reactors at high temperature and absorb large amounts of heat, in the presence of nickel-alumina catalyst and reaction final result is the creation of reaction products and the ratio of hydrogen to carbon monoxide is as the most important criteria for reactor design process[1]. Modeling gas production reactor unit in the petrochemical industry is great importance.

II. Process described



$$\Delta H_{\text{III}} = -41 \text{ kJ/mol}$$

To achieve an appropriate level of transformation and product in the output current, the reaction is one temperature of 900-500°C and at pressure 40-30 bar. In this situation, reactions are quick but due to the need for stability and high mechanical resistance of the catalyst, penetrating into the pores of the catalyst is relatively low and a high percentage of responses in the catalyst grains carried out. Because this reaction is endothermic for access to the appropriate conversion, shall be sufficient energy to gas to react to the move towards equilibrium for this purpose, standard reactors can be built for gas reformer with a high number of tubes that catalyst poured into the tubes and the tubes around the burner tube temperature is raised and to supply reaction energy, diameter tubes are small and uniform reaction will be done relatively[5]. Whatever tube diameter is less, the number of tubes mounted higher and will increase the heat transfer surface and although reactions are quick, but the percent conversion of gas at the outlet tubes will not reach final. To achieve complete conversion of methane to hydrogen need to be very long tubes at the end of tubes to reach equilibrium. Order to pressure drop in the reformer tubes primary, design catalysts with different shape and size with adequate resistance against breakage for this reason, the catalyst grains with large holes in its surface have built and used in ammonia units. The loss of yield and catalyst lifetime happens for several reasons: collected active compounds nickel is an unavoidable process and whatever the temperature of the recovered metal catalyst increase, nickel will more active on its base and accumulate in various parts caused the loss of active surface area of the metal. When the primer feed containing impurities reformer catalyst poisoning is a possibility. Blocking the metal active site by materials toxic will stop cracking hydrocarbons operation. Sulfur is often a poison the catalyst, sodium but there is in hydrocarbons or steam and can not be removed easily and reacts with nickel bands that will reducing the effectiveness catalyst[6]. Other factors the loss of activity of the catalyst are carbon and coke production that soot formation mechanism depends on the temperature and composition of the gas and vapor on the catalyst. When the steam to carbon ratio decreases significantly hydrocarbon feedstock in the form of coke or soot began to break down (pyrolysis) and the soot on the catalytic reformer sitting and increases the pressure drop and the loss of activity of the catalyst. In some cases, can be separated soot from the catalyst by injecting steam and often leads to the replacement

catalytic reformer. Increase or decrease catalyst temperature hurts catalyst grains physical form and expansion and contraction due to temperature drop and increase in the launching primer reformer, shattered grains catalyst, although has not major impact on the chemical activity of the catalyst or heat transfer mode but increased pressure tubes and is reason for the replacement of the catalyst[7].

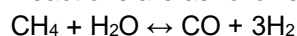
Catalyst grains with different designs for a wide range of hydrocarbons is used and a different type of catalyst used in the reformer stumbling at 1100-900°C works used. Conventional fixed bed reformer has disadvantages such as low heat transfer rate, falling resistance in the pores of the catalyst and nonisothermal. High endothermic reforming process of methane, a high impact on the efficiency of heat entering the reformer and the reformer can therefore be crucial, So choice reformer can be essential, apart from the development of heat transfer, and mass transfer rate, high uniformity catalyst bed, can be advantages to using fluid bed [8].

III. Reaction kinetics

Many equations for methane reforming reaction are presented by various researchers. In Table 2, the number of relationships listed [9].

IV. Equations and modeling

Reactions are as follows:



$$r_1 = -\frac{k_1}{\text{den}^2} \left(\frac{P_{\text{CH}_4} P_{\text{H}_2\text{O}}}{P_{\text{H}_2}^{2.5}} - \frac{P_{\text{CO}} P_{\text{H}_2}^{0.5}}{K_1} \right) \quad [1]$$

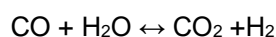
$$k_1 = 9.490 * 10^{16} \exp\left(\frac{-28870}{T}\right) \quad [2]$$

$$K_1 = 10266.76 \exp\left(\frac{-26830}{T}\right) \quad [3]$$

$$k_1 = \text{kg.h/Kmol. KPa}^{0.5}$$

$$K_1 = \text{Pa}^2$$

Second reaction:



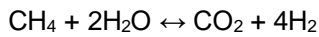
$$r_3 = -\frac{k_3}{\text{den}^2} \left(\frac{P_{\text{CH}_4} P_{\text{H}_2\text{O}}^2}{P_{\text{H}_2}^{3.5}} - \frac{P_{\text{CO}_2} P_{\text{H}_2}^{0.5}}{K_3} \right) \quad [4]$$

$$k_2 = 4.390 * 10^4 \exp\left(\frac{-8074.3}{T}\right) \quad [5]$$

$$K_2 = \exp\left(\frac{-4400}{T} - 4.036\right) \quad [6]$$

$$k_2 = \text{kg.h/ Kmol. K Pa}^{-1}$$

Third reaction:



$$r_3 = -\frac{k_3}{\text{den}^2} \left(\frac{P_{\text{CH}_4} P_{\text{H}_2\text{O}}^2}{P_{\text{H}_2}^{3.5}} - \frac{P_{\text{CO}_2} P_{\text{H}_2}^{0.5}}{K_3} \right) \quad [7]$$

$$k_3 = 2.290 * 10^{16} \exp\left(\frac{-29336}{T}\right) \quad [8]$$

$$K_3 = K_1 * K_2$$

$$k_3 = \text{kg.h/ Kmol. K Pa}^{0.5}$$

$$K_3 = \text{Pa}^2$$

$$\text{den} = 1 + K_{\text{CO}} P_{\text{CO}} + K_{\text{H}_2} P_{\text{H}_2} + K_{\text{CH}_4} P_{\text{CH}_4} + K_{\text{H}_2\text{O}} \frac{P_{\text{H}_2\text{O}}}{P_{\text{H}_2}} \quad [9]$$

$$K_{\text{CH}_4} = 6.65 * 10^{-6} \exp\left(\frac{4604.28}{T}\right) \quad [10]$$

$$K_{\text{H}_2\text{O}} = 1.77 * 10^3 \exp\left(\frac{-10666.356}{T}\right) \quad [11]$$

$$K_{\text{CO}} = 8.23 * 10^{-7} \exp\left(\frac{8497.71}{T}\right) \quad [12]$$

$$K_{\text{H}_2} = 6.12 * 10^{-11} \exp\left(\frac{9971.13}{T}\right) \quad [13]$$

V. Mass balance

$$\frac{dX_{\text{CO}_2}}{dl} = \left(\frac{\pi d_i^2}{4}\right) \frac{\rho_b r_{\text{CO}_2}}{F} \quad [14]$$

$$X_{\text{CO}_2} = 0 @ l = 0 \quad [15]$$

$$\frac{dX_{\text{CH}_4}}{dl} = \left(\frac{\pi d_i^2}{4}\right) \frac{\rho_b r_{\text{CH}_4}}{F} \quad [16]$$

$$X_{\text{CH}_4} = 0 @ l = 0 \quad [17]$$

$$r_{\text{CH}_4} = r_1 + r_3 \quad [18]$$

$$r_{\text{CO}_2} = r_2 + r_3 \quad [19]$$

$$\frac{dX_{\text{CO}}}{dl} = \left(\frac{\pi d_i^2}{4}\right) \frac{\rho_b r_{\text{CO}}}{F} \quad [20]$$

$$X_{\text{CO}} = 0 @ l = 0 \quad [21]$$

VI. Energy balance

$$\frac{dT}{dl} = \frac{1}{G C_p} \left[\frac{4U(T_{w,i} - T)}{d_i} + \rho_b \sum (\Delta H_R * r) \right] \quad [22]$$

$$T = T_{\text{in}} @ l = 0 \quad [23]$$

U

$$= 0.4 \frac{K_g}{D_p} \left[2.58 \left(\frac{D_p G}{\mu}\right)^{\frac{1}{3}} \left(\frac{C_p \mu}{K_g}\right)^{\frac{1}{3}} + 0.094 \left(\frac{D_p G}{\mu}\right)^{0.8} \left(\frac{C_p \mu}{K_g}\right)^{0.4} \right] \quad [24]$$

Arrhenius equation:

$$k_i = A_i e^{-\frac{E}{RT}}, \quad i = 1, 2, 3, \dots \quad [25]$$

Vant Hoff equation:

$$k_j = A_j e^{-\frac{E}{RT}}, \quad j = \text{H}_2, \text{CO}, \text{CO}_2, \text{CH}_4 \quad [26]$$

VII. The pressure drop in the reactor

Because the reaction in a catalytic bed, the gas pressure in the bed are facing. Pressure drop into bed it has an impact on the performance and efficiency [10].

$$\frac{\delta P_t}{\delta z} = -10^{-5} f \frac{e_g v_s^2}{d_p} \quad [27]$$

f from the following equation is obtained and also in terms of atmospheric pressure:

$$f = \frac{1 - \varepsilon_B}{\varepsilon_B} \left[a + \frac{b(1 - \varepsilon_B)}{Re} \right] \quad [28]$$

$$a = 1.75, \quad b = 150, \quad \frac{Re}{1 - \varepsilon_B} < 500 \quad [29]$$

$$a = 1.24, \quad b = 368, \quad 1000 < \frac{Re}{1 - \varepsilon_B} < 5000 \quad [30]$$

$$a = 1.75, \quad b = \frac{4}{2Re^{\frac{5}{6}}}, \quad \frac{Re}{1 - \varepsilon_B} \geq 5000 \quad [31]$$

VIII. Effectiveness factor

For industrial catalysts that are large in size due to the limited penetration velocity equation must be corrected that actual velocity of the particles obtained by multiplying the impact factor [11]. The impact factor is ratio of velocity in catalyst to velocity on the outer surface of the catalyst.

Assumptions that used in the effectiveness factor are as follows:

- Catalyst particles are assumed spherical shape.

- Catalyst particle throughout have the same temperature.

$$n_i = \frac{\int_0^v r_i(P_t) e_g \frac{dv}{v}}{r_i(P_t^s) e_g} \quad [32]$$

Using equations expression for the dependent components profiles obtained [12]:

$$P_{s,CO} - P_{CO} = (D_{e,CO2}/D_{s,CO})(P_{CO2} - P_{s,CO2}) - (D_{e,CH4}/D_{e,CO})(P_{s,CH4} - P_{CH4}) \quad [33]$$

$$P_{s,H2O} - P_{H2O} = (D_{e,CO2}/D_{e,H2O})(P_{CO2} - P_{s,CO2}) - (D_{e,CH4}/D_{e,H2O})(P_{s,CH4} - P_{CH4}) \quad [34]$$

$$P_{s,H2} - P_{H2} = (D_{e,CO2}/D_{e,H2})(P_{CO2} - P_{s,CO2}) - (3D_{e,CH4}/D_{e,H2})(P_{s,CH4} - P_{CH4}) \quad [35]$$

IX. Equations to calculate the physical properties of the fluid:

Assuming the gas will be ideal for mixed density:

$$PV = nRT \quad [36]$$

$$n = \frac{m}{M} \quad [37]$$

$$P = c_R \frac{R}{M} T \quad [38]$$

$$M = \sum_{i=1}^N y_i M_i \quad [39]$$

For an ideal gas specific heat the mixture is calculated as follows:

$$C_{pR}^{ig} = \sum_{i=1}^n y_i C_{pi}^{ig} \quad [40]$$

$$\frac{C_{pR}^{ig}}{RT} = A + BT + CT^2 + DT^{-2} \quad [41]$$

y_i = mole fraction of component i

X. The thermal conductivity of the mixture

$$\lambda_B = \sum_{i=1}^N y_i \lambda_i \quad [42]$$

λ_B = Thermal conductivity of an ideal gas mixture

λ_i = Thermal conductivity of component i

λ_i is a function of temperature and pressure and for one bar the pressure will rise about 1%.

i	A	B*10 ³	C*10 ⁶	D*10 ⁶
CH ₂	1.702	9.081	-2.164	-
H ₂	3.24	0.422	-	0.083
CO	3.376	0.557	-	-0.031
CO ₂	3.475	1.045	-	-1.157
N ₂	3.28	0.593	-	0.04
H ₂ O	3.47	1.45	-	0.121

$$\lambda_B = (1 + 0.01P) \sum_i^N y_i \lambda_i \quad [43]$$

$$\lambda_i = A + BT + CT^2 + DT^3 \quad [44]$$

λ_B = Thermal conductivity of an ideal gas mixture

XI. The viscosity of the ideal gas mixture

The viscosity of the mixture can be obtained from the following equation:

$$\mu_B = \sum_{i=1}^N y_i \mu_i \quad [45]$$

Viscosity is a function of temperature and pressure, but its dependence to the temperature is much more than pressure and only in very high pressures change noticeable [13].

From the following equation can be found on the temperature dependence of viscosity:

$$\mu_i \times AJ = [0.807 T_{ri}^{0.618} - 0.357 \exp(-0.449T_{ri}) + 0.34 \exp(-4.035T_{ri}) + 0.18] F_Q^* F_P^* \quad [46]$$

$$AJ = 0.176 \left\{ \frac{T_{ci}}{M_i^3 P_{ci}^4} \right\}^{0.166} \quad [47]$$

$$F_P^* = 1 \quad 0 < dr < 0.022 \quad [48]$$

$$dr_i = 52.46 \frac{d_i^2 P_{ci}}{T_{ci}^2} \quad [49]$$

D is momentum bipolar and F_Q^* for quantum gases are defined as follows[14]:

$$F_Q^* = 1.22 Q^{0.22} [1 + 0.00385 (T_r - 12)^2] \quad [50]$$

$$Q(\text{He}) = 1.38, \quad Q(\text{H}_2) = 0.76, \quad Q(\text{D}_2) = 0.52$$

XII. Effective diffusion coefficient of components

From the following equation can be obtaining effective penetration into the catalyst grain [15]:

$$D_{\epsilon,i} = \frac{\epsilon_i}{\tau} D_i \quad [51]$$

In relation τ is tortuosity and curvature of the catalyst and ϵ is catalyst grains porosity. D_i is calculated in accordance with the following equation:

$$\frac{1}{D_i} = \frac{1}{D_{im}} + \frac{1}{D_{ik}} \quad [52]$$

Penetration term of the Knudsen diffusion is calculated by the relationship

$$D_{ik} = a_p \frac{\varepsilon_s}{\tau} \frac{2}{3} \left[\frac{8RT}{\pi M_i} \right]^{0.5} \quad [53]$$

Molecular effusion terms:

$$\frac{1}{D_{im}} = \frac{1}{1 - y_i} \sum_{j=2}^N \frac{y_j}{D_{ij}} \quad [54]$$

Two-component diffusion coefficient obtained from the following equation:

$$D_{ij} = \frac{10^{-4} (1.084 - 0.249 \sqrt{\frac{1}{M_i} + \frac{1}{M_j}}) T^{1.5} \sqrt{\frac{1}{M_i} + \frac{1}{M_j}}}{P_t r_{ij} f \left(\frac{kT}{\varepsilon'_{ij}} \right)} \quad [55]$$

$$r_{ij} = \frac{r_i + r_j}{2} \quad [56]$$

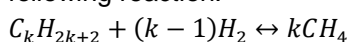
$$\varepsilon'_{ij} = \sqrt{\varepsilon'_i \varepsilon'_j} \quad [57]$$

A ker and Camo (1995)	$-r_{CH_4} = k_0 e^{\frac{-E}{RT}} P_{CH_4}$
Boudart (1967)	$-r_{CH_4} = k_0 e^{\frac{-E}{RT}} P_{CH_4}$
Allen (1975)	$-r_{CH_4} = k_0 e^{\frac{-E}{RT}} P_{CH_4}$
Rostrup-Nielsen (1973)	$-r_{CH_4} = k_0 e^{\frac{-E}{RT}} \left[P_{CH_4} - \left(1 - \left(\frac{Q_r}{K_p} \right) \right) \right]$

i	A	B	C	D
H ₂	8.099*10 ⁻³	6.689*10 ⁻⁴	-4.158*10 ⁻⁷	1.562*10 ⁻²⁰
CO	5.067*10 ⁻⁴	9.125*10 ⁻⁵	-3.524*10 ⁻⁸	8.199*10 ⁻¹²
CO ₂	-7.21*10 ⁻³	8.015*10 ⁻⁵	5.477*10 ⁻⁹	-1.053*10 ⁻¹¹
H ₂ O	7.64*10 ⁻³	-1.015*10 ⁻⁵	1.801*10 ⁻⁷	-9.1*10 ⁻¹¹
N ₂	3.919*10 ⁻⁴	8.727*10 ⁻⁵	-5.067*10 ⁻⁸	1.504*10 ⁻¹¹
CH ₄	-1.869*10 ⁻³	8.099*10 ⁻³	1.179*10 ⁻⁷	-3.614*10 ⁻¹¹

XIII. The Role of operating conditions

In reforming reaction, the reaction in this entry indicates that hydrocracking reactions occur according to the following reaction:



Compounds heavier than methane are broken at the entrance of the tube and produce methane.

0.1014	The inner diameter of the tube (m)
0.1320	The outer diameter of the tube (m)
2.12	The total length of the tube (m)
0.0172	Catalyst ring outer diameter (m)
0.01	Catalyst height (m)
2.2355	Catalyst density (Kg/m ³)
0.528	ε_s
3.54	τ

Percent of methane and carbon dioxide by increasing the inlet gas temperature decreases instead Percent of hydrogen and water vapor increases and this increase in temperature reduces the pressure. If we reduce the pressure at constant temperature the same result will happen. Also by increasing the temperature in reactor input ratios H₂O/CH₄ and CO₂/CH₄ will remain constant, but the H₂/CO ratio decreases. with increasing temperature reaction goes to the water gas shift reaction and produces H₂ and CO with a ratio of 3 and this ratio in reactor output decreases by temperature increasing and eventually turns about one. Increasing the pressure at constant temperature or reducing the inlet gas temperature at a constant pressure to increase percent of methane, and hydrogen and carbon monoxide and water vapor will decrease. In water gas shift reaction does not change the number of moles then pressure in this reaction is not impressive but in reforming reaction is impressionable and is reducing H₂/CO ratio. Due to the progress of the reaction in the presence of excess water vapor, with increasing the amount of H₂O in the input, H₂/CO ratio increases and decreases the mole percent methane output.

Operating pressure, temperature and molar ratio H₂O/CH₄ and CO₂/CH₄ are influencing parameters at the composition of the synthesis gas and for various uses can be obtained by changing these parameters different values. Basically, the reforming reaction is an endothermic reaction and therefore high temperature will improve and further conversion of methane but role of pressure is lower than other variables and therefore it can be considered as a fixed parameter.

A (k ₁)	A (k ₂)	A (k ₃)	A (K _{CO})	A (K _{H₂})	A (K _{CH₂})	A (K _{H₂O})
4.225*10 ¹⁵	1.95*10 ⁶	1.021*10 ¹⁵	8.2*10 ⁻⁵	6.2*10 ⁻⁹	6.6*10 ⁻²	1.8*10 ⁵

Because water gas shift reaction is exothermic thereupon in a high-temperature reaction suppresses so increase in temperature causes more methane into H₂ and CO, CO₂ and convert CO into CO₂ is less.

Reforming reactions are equilibrium reactions therefore composition of the feed heavily affects on the ratio of CO/H₂ production. Increasing the amount of CO₂ in the feed advances gas-water shift reaction and reduces in CO/H₂ production. By increasing the ratio of H₂O/CH₄ in input, the ratio of CO₂/CH₄ constant in input and temperature is constant and increases the ratio of CO/H₂ production. By increasing of CO₂/CH₄ input in H₂O/CH₄ constant input and constant-temperature the CO/H₂ ratio will reduce.

Table 6 -values of E and H vant Hoff and Arrhenius equation(kJ / mol)						
E ₁	E ₂	E ₃	ΔH _{CO}	ΔH _{H₂}	ΔH _{CH₄}	ΔH _{H₂O}
241	66	23	-70	-81	-38	89

XIV. Conclusions

In the process, concentration gradient of gas component is limited to a very thin layer of the active catalyst. Catalyst component concentration gradient is very steep in grain and mass transfer resistance components in the catalyst grain are process controller.

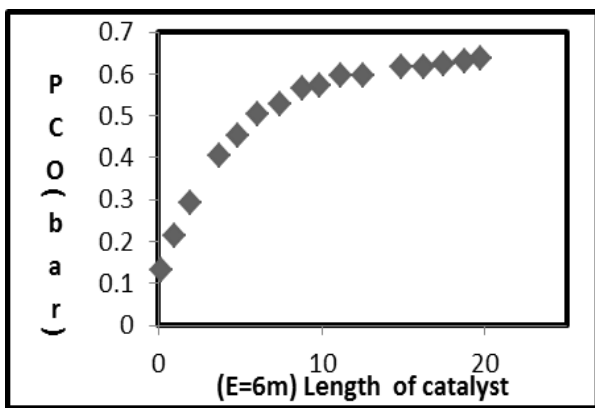


Figure 1 - CO pressure on the length of the catalyst for the (E=6m)

Figure 2. Experimental data and model prediction against the input oxygen flow for changes of methane conversion depicts. Irreversible reaction of methane to produce CO or CO₂ and water can increase the conversion of methane to increase oxygen flow.

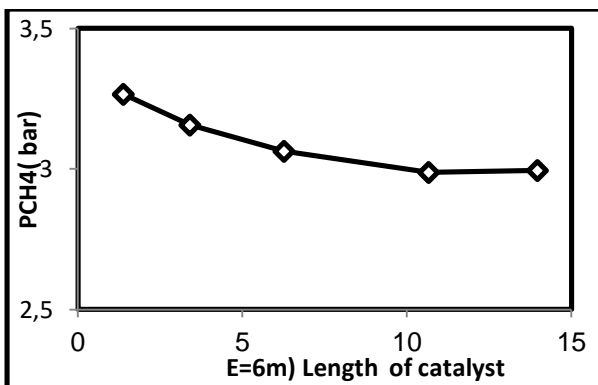


Figure 2 – CH₄ pressure on the length of the catalyst for the (E=6m)

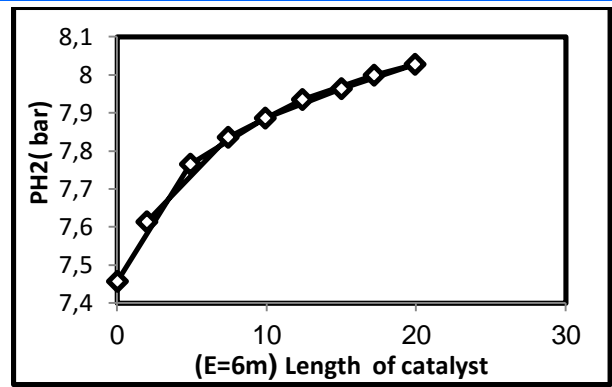


Figure 3 – H₂ pressure on the length of the catalyst for the (E= 6m)

Becomes smaller catalyst particles in the pipes are reduced catalyst loading and increases the pressure drop across the bed.

In industry has always faced pressure restriction so can built a catalyst with complex shapes in order to create more internal level, low pressure drop, or active substances in the outer layer of the catalyst, on the one hand improve the impact and also saving the use of active metal that is usually expensive.

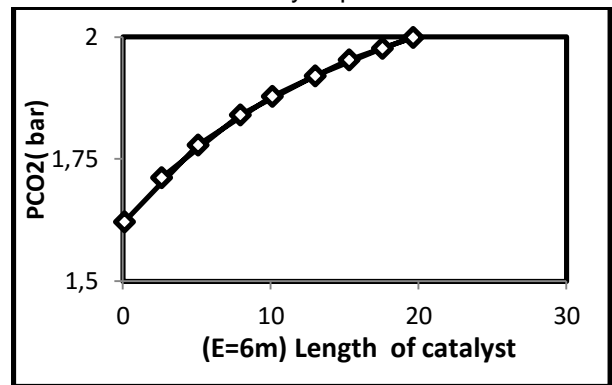


Figure 4 – CO₂ pressure on the length of the catalyst for the (E=6m)

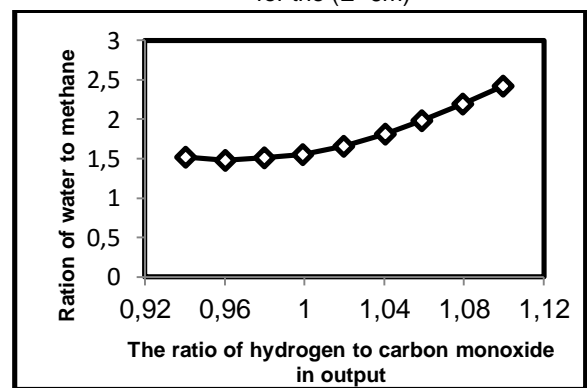


Figure 5 - the ratio of in output to ratio of water to methane

The ratio of hydrogen to carbon monoxide in reactor design as a key parameter considered so for any change in this ratio changes to the percentage of feed to be closer to the real standard.

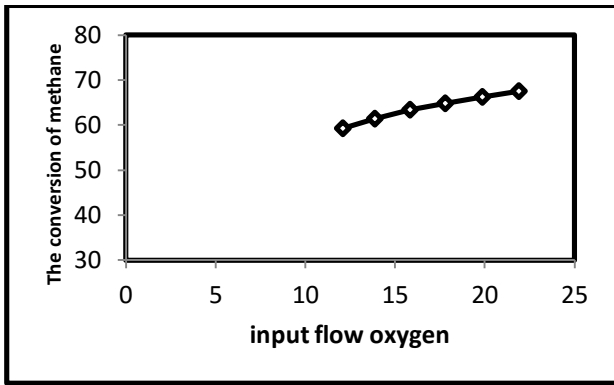


Figure 6 - Oxygen flow at input to the conversion of methane

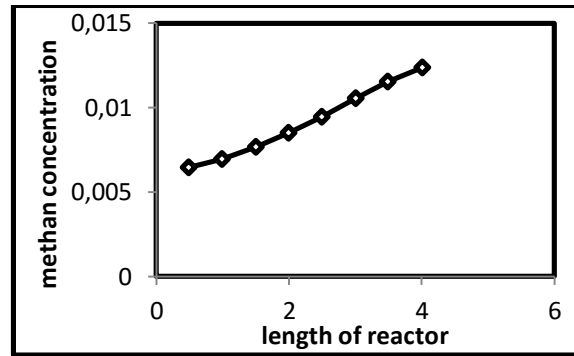


Figure10. Methane concentration changes in the reactor length

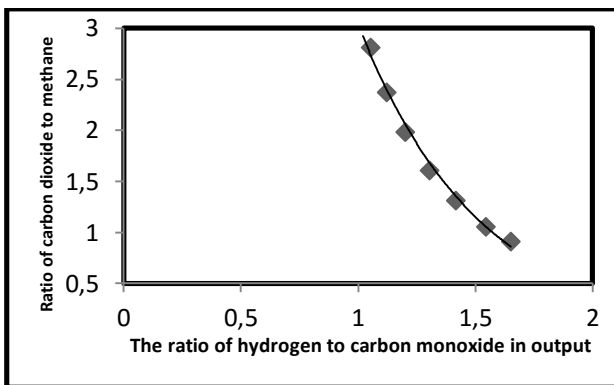


Figure 7 - The ratio of in output of carbon dioxide to methane

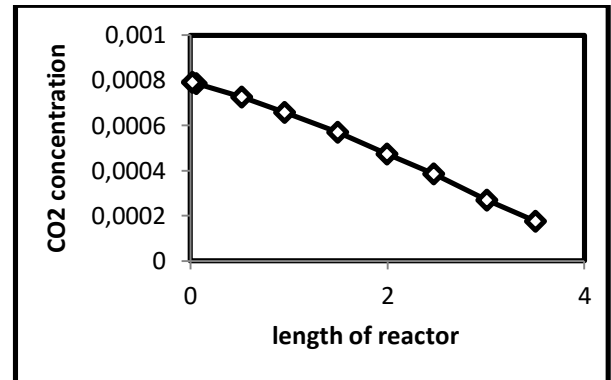


Figure9. CO₂ concentration changes in the reactor length

Feed flow rate (kmol/hr)	3392
Temperature (K)	586
Pressure (KPa)	2074

In this collection of methane in the reactor pressure decreased with increasing pressure. This process was assessed as a thermodynamically unfavorable reaction system.

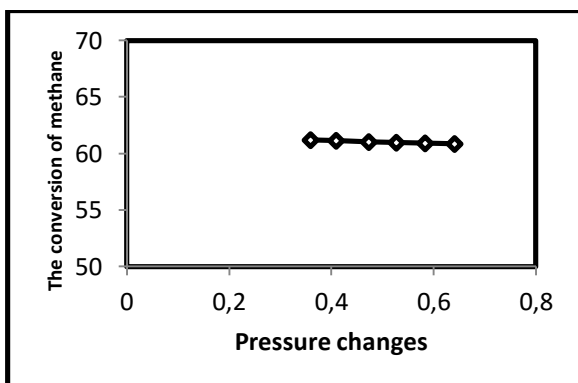


Figure 8 - Pressure changes in the conversion of

XV. Reference

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