



Numerical Investigation of the Performance of Hydrogen Production Process by Production Gas Recirculation

A. Saeedi*, N. Allahdadi

Faculty of Engineering, University of Birjand, Birjand, Iran

ABSTRACT: Clean energy sources such as hydrogen are developing because of environmental issues. Partial oxidation of methane is important among different methods of hydrogen production due to the reduction in carbon deposition, doing the process in a lower temperature range, the high conversion of methane and low energy consumption. In the present work, the numerical simulation of partial oxidation of methane with Rh/Al₂O₃ catalyst is conducted in a fixed bed flow reactor. The effect of different volumetric percentages of product gas recirculation on the hydrogen, carbon monoxide, and carbon dioxide production is calculated for various temperatures (500-900°C) and oxygen to methane ratios equal 0.4, 0.5, 0.6 and 0.7, respectively. According to that, the suitable temperature and oxygen to methane ratio are considered. The results show that in the temperature range, an increase in the product gas recirculation causes an increase in Hydrogen and CO production and a decrease in CO₂ production. Moreover, the calculated data shows that the inlet oxygen to methane ratio of 0.5 and reaction temperature of 600°C is suitable to enhance Hydrogen production performance by production gas recirculation. Also, it is demonstrated that 50% volumetric product gas recirculation in that temperature and O₂/CH₄ ratio, leads to an increase in H₂ production and a decrease in CO₂ production about 30%.

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1. INTRODUCTION

Hydrogen is produced from fossil and non-fossil fuels, and 50% of the production of hydrogen is from natural gas [1]. To reduce the possibility of carbon deposition during the hydrogen production process, the use of partial oxidation of methane is a proposed method [2]. The use of a catalyst leads partial oxidation process to a lower temperature range (600-900°C) [1]. A suitable catalyst for reforming to catalyze the reaction at low temperatures has the following specifications: resistant to coke formation, and tolerant of different concentrations of poisons (e.g. Sulphur, halogens, heavy metals, etc.) for an extended period [3]. Noble metals based catalysts are very active and selective for Polyoxometalate (POM) in terms of conversion and selectivity for H₂ and CO [4]. Therefore, hydrogen production by catalytic partial oxidation of methane is important. Cheephat et al. [5] studied the partial oxidation reactions of methane in the presence of Ni, Rh, Re metal catalysts and Al₂O₃ supported Re-Ni catalysts at 700-400°C. Their results showed that the Rh/Al₂O₃ catalyst had the highest catalytic activity. The stability test results of Re-Ni/Al₂O₃, Ni/Al₂O₃, and Rh/Al₂O₃ catalysts showed that Ni/Al₂O₃ and Rh/Al₂O₃ catalysts had higher inactivation. Eriksson et al. [6] tested the partial oxidation of methane in the presence of a supported Rh catalyst for the production of synthesized gas (H₂ and CO) at atmospheric pressure. Their experiments showed that Rh-based catalysts are active for partial oxidation of methane. Horn et al. [7] investigated the effect of pressure

*Corresponding author's email: ali.saeedi@birjand.ac.ir

changes on the partial catalytic oxidation of methane on Rh and Pt coated foams. Their study showed that pressure changes had a minor effect on the process of partial oxidation of methane in the presence of Rh catalyst. Deutschmann et al. [8] investigated the partial oxidation of methane on Rh-coated monoliths in numerical and experimental conditions. Their results showed that the CH₄/O₂ ratio of 1.8 indicated rapid O₂ consumption at the catalyst initial edge and CH₄ consumed throughout the reactor. Schwiedernoch et al. [9] studied the catalytic partial oxidation of methane on rhodium/alumina in experimentally and numerically at atmospheric pressure. Their results showed that, however, complete methane oxidation to water and carbon dioxide occurs, then selectivity of synthesis gas slowly increased with increasing temperature. The purpose of the present study is to improve the hydrogen production performance and carbon dioxide reduction during numerical simulation of catalytic partial oxidation of methane over Rh/Al₂O₃ catalyst in a fixed bed reactor by the different volumetric percentages of Production Gas Recirculation (PGR) in the reactor inlet. The present numerical simulation was performed using the Cantera software and the Python programming language for different temperatures (500-900°C) and oxygen to methane input ratios of 0.4, 0.5, 0.6, and 0.7. Hydrogen production in this temperature range and four different O₂/CH₄ input ratios and the effects of 3, 5 and 7 volumetric percentages of PGR on hydrogen, CO and CO₂ production were investigated. Based on this, the appropriate temperature and O₂/CH₄ were



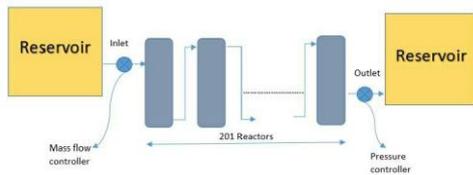


Fig. 1. Schematic design of a chain of zero-dimensional reactors for modeling the plug flow reactor

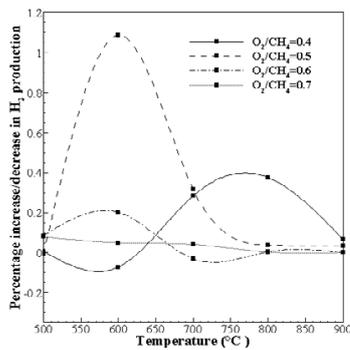


Fig. 2. Hydrogen production fluctuations at different temperatures by recirculating of product gas equals 3% volumetric

determined during the catalytic partial oxidation of methane.

2. BASICS AND METHOD OF SOLVING

Cantera software was used for the simulation and the numerical code was written in Python programming language. The Plug-Flow Reactor (PFR) at this work was simulated by a chain of NReactors stirred reactors. The plug flow reactor was represented by a linear chain of zero-dimensional reactors [10]. Fig. 1 shows the schematic design of a chain of zero-dimensional reactors for modeling plug flow reactor.

3. GOVERNING EQUATIONS

The momentum equation is ignored by assuming constant pressure and frictionless flow. The plug flow reactor is considered as isothermal. Therefore, the thermal energy equation is not needed. So governing equations are:

$$\sum_{in} \dot{m}_{in} - \sum_{out} \dot{m}_{out} + \dot{m}_{wall} = 0 \quad (1)$$

$$\sum_{in} \dot{m}_{in} (Y_{k,in} - Y_k) + \dot{m}_{k,gen} - Y_k \dot{m}_{wall} = 0 \quad (2)$$

$$\min(G(T, P, n) = \sum_{i=1}^N n_i \mu_i) \quad (3)$$

4. RESULT AND DISCUSSION

The kinetic model was used for the partial catalytic oxidation of methane in the presence of Rh/Al₂O₃ catalyst consists of 38 reactions [11]. Fig. 2 shows the hydrogen production fluctuations at different temperatures by recirculating product gas equal to 3% volumetric. At 600°C

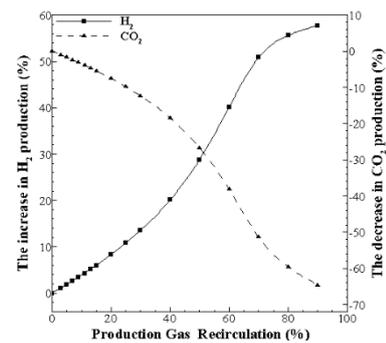


Fig. 3. The trend of hydrogen and CO₂ production at 600°C during the different volumetric percent of product gas recirculation

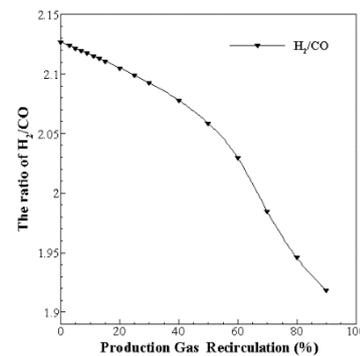


Fig. 4. The ratio of hydrogen to carbon monoxide production at 600°C for different volumes of PGR

and the O₂/CH₄ ratio equals to 0.5, hydrogen production has a peak, which is 1.1 % greater than hydrogen production without PGR. In general, it can be said that in the presence of the Rh/Al₂O₃ catalyst for the PGR process, the choice of T=600°C and O₂/CH₄=0.5 are appropriate. Fig. 3 shows the trend of hydrogen and CO₂ production at 600°C during the different volumetric percent of product gas recirculation. By increasing volumetric percentages of PGR, CO₂ production decreases and H₂ production goes up. Fig. 4 shows the ratio of hydrogen to carbon monoxide production at 600°C for different volumes of product gas recirculation. For hydrogen production purposes, up to 50% volume of PGR is suitable because it provides hydrogen to carbon monoxide ratio greater than 2.

5. CONCLUSION

In the present work, the numerical simulation of partial oxidation of methane with Rh/Al₂O₃ catalyst was conducted in a fixed bed flow reactor. The Cantera software and Python programming language were used in this work. The current simulation data was tested by valid results and the conclusions are as follows:

- Hydrogen production increases with increasing temperature and oxygen to methane input ratio.
- At all temperature ranges, hydrogen production increases with the PGR process.
- In partial oxidation of methane in the presence of Rh/

Al_2O_3 catalyst during the PGR process, $T=600^\circ\text{C}$ and $\text{O}_2/\text{CH}_4=0.5$ are selective and optimal.

- For hydrogen production purposes, up to 50% volume of PGR is suitable.
- With 50 % volumetric recovery of gaseous products at the reactor inlet at 600°C and O_2/CH_4 equals 0.5, the production of hydrogen and carbon dioxide increases and decreases by about 30%, respectively.

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