Available online at www.sciencedirect.com

SciVerse ScienceDirect

Procedia Engineering 42 (2012) 2 – 24

Procedia Engineering

www.elsevier.com/locate/procedia

20th International Congress of Chemical and Process Engineering CHISA 2012
25 – 29 August 2012, Prague, Czech Republic

A CFD model for methane autothermal reforming on Ru/γ-Al_2O_3 catalyst

S. Rowshanzamir [a], S. M. Safdarnejad[a], M. H. Eikani[b] a*

[a]*Iran University of Science and Technology, Narmak, Tehran 13114-16846 , Iran*
[b]*Iranian Research Organization for Science & Technology, Tehran 115- 13135 , Iran*

Abstract

Hydrogen is the preferred fuel for fuel cells due to high reactivity for electrochemical reaction at anode. In the present study, a three dimensional CFD (Computational Fluid Dynamics) code was developed and validated to simulate the performance of a catalytic monolith fuel processor used for hydrogen generation. Methane autothermal reforming on 5% Ru/γ-Al_2O_3 catalyst was selected as the reaction mechanism. Ruthenium catalyst is a suitable catalyst for low temperature catalytic partial oxidation (LTCPO) process. This catalyst has good reforming activity and high hydrogen yield is obtained for ruthenium/γ-alumina. This catalyst also demonstrated to be stable within the investigation time. The computational domain of the simulations was selected to be the catalytic section of the reformer. The results provided an adequate match to the experimental data from literature with respect to the outlet and maximum reactor temperature and also distribution of the products. The reactor performance was thereafter studied by numerically revealing the effects of variations of O_2/C and S/C feed molar ratios, and feed temperature on the profiles of temperature and species concentrations. Moreover, effects of using air instead of pure oxygen were also investigated. It was concluded that at higher O_2/C and S/C feed molar ratios and also at higher feed gas temperature, more hydrogen will be achieved at the reactor outlet, which is very suitable for fuel cell applications.

Keywords: Autothermal reforming; hydrogen; monolith reactor; CFD code; methane

* Corresponding author. Tel.: +98-21-7749-1223; fax: +98-21-7749-1242.
E-mail address: rowshanzamir@iust.ac.ir.

1877-7058 © 2012 Published by Elsevier Ltd. Open access under CC BY-NC-ND license.
doi:10.1016/j.proeng.2012.07.390

1. Introduction

Fuel cell technology has been a subject of vital research and development recently due to the higher energy conversion efficiency and lower amounts of emission gases in comparison with internal combustion engines [1]. For polymer electrolyte membrane fuel cells (PEMFC) which are considered as the most technologically mature of the different types of fuel cells, hydrogen is the preferred fuel due to high reactivity for the electrochemical reaction at anode [2]. In addition, hydrogen is a suitable substitution for hydrocarbons for different applications including fuel cells and internal combustion engines. However, hydrogen does not exist in pure form in nature. Furthermore, high storage pressure of hydrogen and the cost of its storage along with the lack of hydrogen distribution infrastructure at the present time restrict its direct use [3]. On board reforming of other fuels is a practical option to tackle this problem at the present time. Different fuels can be used for reforming processes. Natural gas and its surrogates such as methane are considered to be among the most attractive fuels for hydrogen production due to large existing reserves and available transportation infrastructure [4].

There are three common methods for hydrogen generation. Steam reforming (SR) process is the common industrial method for hydrogen production. However, steam reforming is highly endothermic and tends to possess large heat transfer areas. As a desirable characteristic, onboard fuel processors should be compact [5]. Considering this, steam reforming is not a suitable method for onboard fuel processing. Partial oxidation (POX) is another method for hydrogen production. Although the reaction rates are much higher for POX in comparison with SR, H_2 yield per carbon in the fuel is lower than SR. In addition, POX tends to produce coke [6]. Autothermal reforming (ATR) is the combination of steam reforming and oxidation reactions [7]. The key feature of autothermal reforming is that the thermal energy generated by POX is absorbed by SR which results a thermo neutral process. Moreover, by using steam, coke formation which is occurring in partial oxidation is reduced and it also takes advantage of high hydrogen yield in steam reforming [8]. Compact size, quick response, and inexpensive material requirements are other key features of an ATR reformer [9]. Therefore, autothermal reforming is the preferred reforming method in comparison with endothermic steam reforming and exothermic partial oxidation and has been the focal point of most of the studies in recent years as a feasible technique for hydrogen production in portable fuel cell applications.

Trimm and Lam [10] studied ATR of methane on Pt/Al_2O_3 catalyst. The kinetics of the reactions was measured at temperatures around 527 ˚C. Numaguchi and Kikuchi [11] studied methane steam reforming on 8.7 wt% Ni/Al_2O_3 catalyst in a fixed bed reactor. The temperature was in the range of 400–890 ˚C. They assumed that only CO was formed from the reaction between CH_4 and H_2O. Carbon dioxide was only produced from the water gas shift reaction (WGS). Xu and Froment [12] presented intrinsic kinetics of methane steam reforming, methanation, and the WGS reactions on Ni/Al_2O_3 catalyst for initial temperature range between 300 and 550 ˚C. The kinetics they developed was used extensively in subsequent investigations.

Packed bed reactors have been widely used in reforming processes. Dias and Assaf [13] examined methane autothermal reforming over Ni/ γ-Al_2O_3 catalyst in a packed bed reactor. The influence of addition of small amounts of noble metals to the Ni/γ-Al_2O_3 was also investigated. Temperature range in this study was between 400 and 600 ˚C. As reported, addition of noble metals led to hydrogen production enhancement. Zahedi Nezhad et al. [14] developed a model for methane autothermal reforming on $Ni/MgAl_2O_4$. The reactor they used consisted of two sections: a non-catalytic section used for partial oxidation and a catalytic packed bed section used for steam reforming. Desirable feed temperature and pressure as well as feed composition were determined to achieve higher H_2, CO, and H_2/CO ratio. Effective length of the catalyst bed was also determined.