INTERNATIONAL JOURNAL OF HYDROGEN ENERGY 39 (2014) 11583–11591

Available online at www.sciencedirect.com

ScienceDirect

journal homepage: www.elsevier.com/locate/he

Numerical investigation of components influence on characteristics of autothermal reforming of methane in micro premix chamber

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ARTICLE INFO

Article history:
Received 24 December 2013
Received in revised form
15 May 2014
Accepted 18 May 2014
Available online 14 June 2014

Keywords:
Micro premixed
Methane
Surface reaction
Autothermal reforming

ABSTRACT

This paper focuses on investigating that the influence of O_2, CO_2 and H_2O on characteristics of autothermal reforming of methane in micro premix chamber on Ni catalysts. In addition, the effect of catalytic wall temperature on autothermal reforming reaction of methane under a certain ratio of $CH_4/CO_2/H_2O/O_2$ is simulated. The results indicate that appropriately increasing O_2 concentration can increase the conversion efficiency of CH_4, so does adding CO_2 or H_2O. The positive effect of O_2, CO_2 and H_2O is more pronounced at the higher temperature. The temperature range of 650–750 K is the important transitional region in the reactions of CH_4/O_2, CH_4/H_2O and CH_4/CO_2. It also gives a guide to the available range of parameters in the high efficiency reforming process of micro-reactor.

Introduction

The rapid development of micro-electrical mechanical systems (MEMS) technology makes light weight, high energy density and long lifespan micro-energy devices to become a research hotspot in the energy and power field [1]. The micro-combustion has a remarkable characteristic of largely losing heat. Its characteristic size is smaller than flameout size of flame spread in space reaction, which leads that the burning flame cannot normally spread [2]. Therefore the space reaction will be unstable [3,4]. However, the surface catalytic reaction is not restricted by flame failure. Furthermore, hydrogen holds the characteristics of high energy/mass ratio, wide ignition range, low ignition temperature, high stability and efficiency of combustion. Owing to these favorable combustion characteristics, hydrogen becomes the first choice to realize steady combustion in the micro-combustion systems [5]. However, it is little value to directly add hydrogen into the combustor in the industrial application. Premixing of fuel and

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http://dx.doi.org/10.1016/j.ijhydene.2014.05.114

oxygen can prolong combustion time and hydrogen can strengthen combustion stability and improve combustion efficiency, which can be achieved in the premixed chamber for the catalytic reforming reaction of hydrocarbon fuel. Autothermal reforming (ATR) of hydrocarbon fuel is suggested. It is a complex thermo-chemical process in which the thermal effects of partial oxidation (POX) and steam reforming (SR) reactions are combined by feeding the hydrocarbon fuel, water and air into the micro-reactor. The two processes (POX and SR) occur simultaneously in the presence of the catalyst. ATR is a single reactor system, and hydrocarbons are completely converted into a mixture of H_2 and CO in this system. The process is called as autothermal in which the partial oxidation of a portion of hydrocarbon provides the heat required by the endothermic reforming reactions. Therefore, the whole process is thermally neutral. The temperature becomes lower in favor of the water–gas shift reaction which consumes the generated carbon monoxide to produce more hydrogen. So it is proposed that producing hydrogen technology through premixed autothermal catalytic reforming of methane.

Hydrogen is an important raw material in the chemical industries. In addition, hydrogen might become a new clean energy in the transportation and aerospace fields, especially in fuel cell [6], so hydrogen will be in great demand. Reforming of methane has been widely used to produce hydrogen in the micro-devices. However, the researches of producing hydrogen mainly focus on the effect of the reactor structure and the improvement of catalyst activity on the characteristic of reforming reaction in the micro-scale [7,8]. Dias et al. [9] studied ATR of methane with Ni/γ-Al_2O_3 catalyst on a thin layer packed bed. It indicates that the effect of noble metals will significantly increase the hydrogen production up to 25% at the temperature range of 673 K–873 K. Halabi et al. [10] presented one-dimensional model for heterogeneous autothermal reforming of methane with Ni/Al_2O_3 catalyst in the fixed-bed reactor to research the optimum conditions to obtain the maximum yield of hydrogen. Reforming reaction of methane and steam can only run smoothly at the high water–carbon ratio [11], while the speed of CH_4/O_2 laminar premixed flame can reduce because of steam in the mixed gas (methane, oxygen and hydrogen) [12]. In addition, CO_2 content also has a certain effect on the reforming characteristic of methane [13,14]. In the autothermal reforming reaction of CH_4/O_2 with H_2O and CO_2, the heat is produced by the oxidation reaction of CH_4 and O_2. And then the H_2O and CO_2 can continue to arouse the reforming reaction with CH_4. A lot of researches focus on the characteristics of methane-steam reforming and catalytic combustion between methane and oxygen [15–17]. In our previous works, we had considered the effect of molar equivalence ratio of O_2/CH_4 on the conversion of methane to hydrogen [18,19]. The results show the optimum O/C ratio is 0.2–0.225, with the mass fraction of H_2 reaching 4.52%–4.81%. However, the autothermal reforming of methane in the micro-scale is still at the primary stage, and the relevant literatures about the characteristics of autothermal reforming reaction between methane and oxygen under the existence of CO_2 or H_2O are rarely reported [20,21]. Therefore, it has an important significance to carry out the related research. In this paper, the simulation studies are performed using the different catalytic wall temperature in micro premix chamber. In addition, the reaction characteristics of autothermal reforming profile of the various mass flow ratio of $CH_4/CO_2/H_2O/O_2$ with a 3-D heterogeneous catalytic reaction model. The simulation results present the autothermal reforming characteristics of micro ATR reactor at different operating conditions.

Material and methods: physical model and mathematical model

Physical model

In order to develop and design the micro premixed chamber with the excellent premixing quality and the small flow resistance, our team has done a lot of researches. The optimal parameters, such as radian and number of swirling groove, position and diameter of fuel inlet, have been obtained [22,23]. The detailed structure is as follows. The micro premixed chamber in which the fuel and $CO_2/H_2O/O_2$ are fully mixed is useful for the autothermal reforming. It is compact and smaller than the quarter dollar. Moreover, the micro premixed reforming chamber consists of six swirling symmetrical grooves, which have arc section with 1 mm in side length. The mixed gas flows from the inlet to the circular chamber (21 mm in outer diameter, 14 mm in inner diameter and 0.5 mm in height) though swirling grooves. These swirling grooves can improve the mixing of fuel and air. During this process the relative mix coefficient strongly varies, eventually leading to over 99% [23]. The schematic geometry of the micro premixed reforming chamber is presented in Fig. 1. An inlet (1 mm in diameter) of $CO_2/H_2O/O_2$ is located in the center of premixed reforming chamber. The six fuel inlets (0.3 mm in diameter) are located at the circumference whose radius is 2 mm. The twelve circular outlets (1 mm in diameter) are evenly arranged at the bottom of premix chamber. $CO_2/H_2O/O_2$ and fuel are

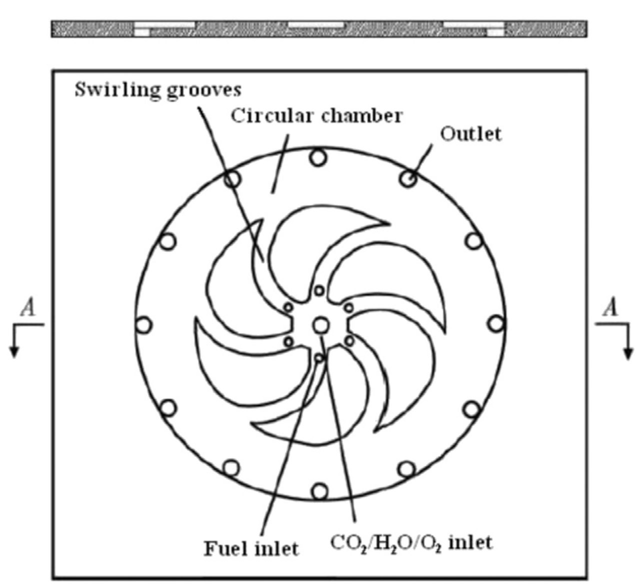

Fig. 1 – The structure of micro premixed reforming chamber.