

Available online at [www.sciencedirect.com](http://www.sciencedirect.com)

ScienceDirect

journal homepage: [www.elsevier.com/locate/ijhydene](http://www.elsevier.com/locate/ijhydene)

# Thermal and chemical effects of hydrogen addition on catalytic micro-combustion of methane–air



Yunfei Yan <sup>a,b,\*</sup>, Weimin Tang <sup>b</sup>, Li Zhang <sup>a,b</sup>, Wenli Pan <sup>b</sup>, Liya Li <sup>b</sup>

<sup>a</sup> Key Laboratory of Low-grade Energy Utilization Technologies and Systems, Chongqing University, Ministry of Education, Chongqing 400030, PR China

<sup>b</sup> College of Power Engineering, Chongqing University, Chongqing 400030, PR China

## ARTICLE INFO

### Article history:

Received 28 July 2014

Received in revised form

3 September 2014

Accepted 11 September 2014

Available online 7 October 2014

### Keywords:

Micro-reactor

Methane

Hydrogen addition

Thermal effects

Chemical effect

Ignition

## ABSTRACT

In this paper hydrogen assisted catalytic combustion of methane on rhodium is numerically modeled in steady condition. The aim of the work goes to better understand how the addition of hydrogen affects the combustion of methane–air. For this purpose, a micro flatbed channel is investigated by a three-dimensional simulation including an elementary-step surface reaction mechanism. It is clearly shown through a numerical study that appropriate hydrogen addition increases the conversion of methane and expands the lower limit of burnable equivalence ratio. In addition, the main effect of hydrogen is thermal when the mass fraction of hydrogen addition is less than 0.67%, while not only thermal effect but also chemical effect appears when the mass fraction is more than 0.67%. The sharp decreases of hydrogen fraction appear twice till hydrogen fraction increases from 0.67%. In addition, the first abrupt decline increases Rh(s) coverage to create favorable conditions for adsorption and oxidation of methane and it can suddenly reduce the ignition temperature 15 K and advance ignition distance 3%. Thanks to the second sharp decline, the adsorption–desorption equilibrium of oxygen slowly shifts towards desorption with increasing temperature to increase Rh(s) coverage.

Copyright © 2014, Hydrogen Energy Publications, LLC. Published by Elsevier Ltd. All rights reserved.

## Introduction

Micro devices have been playing a key role on human life for aviation, spaceflight, automobile, biomedicine, environmental regulation, military affair and so on, such as sensors, micro medical devices, micro-pumps, micro-motors [1]. This makes theoretical and experimental combustion work of the micro energy power system has become a hot topic [2–4]. The size of the micro combustor from millimeter to centimeter

scale is a strong result for a sharp increase in the rate of heat loss and a significant shortening of time during which fuel traverses through the channel. In many cases, the characteristics size of the combustor is less than the flame quenching distance. Thus, combustion in micro reactor is hard to maintain stability and high efficiency. Currently, catalytic reaction has been usually adopted by loading the catalyst to transform conventional combustion into wall combustion for the promotion of combustion stability and the increase of methane conversion in the micro-reactor.

\* Corresponding author. Key Laboratory of Low-grade Energy Utilization Technologies and Systems, Chongqing University, Ministry of Education, Chongqing 400030, PR China. Tel.: +86 23 65103114; fax: +86 023 65111832.

E-mail address: [yunfeiyan@cqu.edu.cn](mailto:yunfeiyan@cqu.edu.cn) (Y. Yan).

<http://dx.doi.org/10.1016/j.ijhydene.2014.09.064>

0360-3199/Copyright © 2014, Hydrogen Energy Publications, LLC. Published by Elsevier Ltd. All rights reserved.

Although the catalyst is usually a noble metal in the combustion of methane, this kind of catalyst for methane catalytic reaction has some disadvantages. Because of a higher sticking probability of  $O_2$  in comparison to that of  $CH_4$ , the surface sites Pt(s) are covered by  $O_2$ , inhibiting the adsorption of  $CH_4$  and the proceeding of reaction. It is responsible for the increase of ignition temperature and decrease of methane conversion. Adding a small amount of hydrogen into fed gas can reduce the O(s) coverage of the wall surface to improve the combustion. As a result, investigating the hydrogen addition into hydrocarbon fuel is increasingly favored by the researcher [5–8].

Yan et al. [9] have performed a numerical research to study the combustion of methane under both catalytic and non-catalytic conditions. Results indicate that hydrogen addition obviously boosts the conversion rate of methane, especially with catalytic. Furthermore, the O(s) coverage decreases with hydrogen addition and the methane combustion moves toward the more stabilized reaction as hydrogen is added [10]. Ran et al. [11] studied the catalytic oxidation of fuel-lean  $CH_4-H_2$  mixtures over Pt in a micro-channel. The results show that the surface sites Pt(s) is covered by O(s), inhibiting the adsorption of methane and resulting in a high ignition temperature. The hydrogen addition can decrease the initial reaction temperature and the ignition temperature of methane. Zheng et al. [12] conducted an experiment of  $CO/O_2$  and  $CO/H_2/O_2$  catalytic combustion in comparison with numerical simulation. In the ignition phase, hydrogen addition inhibits  $O_2$  from adsorbing, reduced the number of O(s) on the wall. Zhong et al. [13,14] found that the effect of hydrogen on the catalytic ignition process and temperature of n-butane depends on the percentage of hydrogen added. When a small amount of hydrogen is added, it has a thermal effect, while adding more hydrogen gas causes it to have a chemical effect.

There is no unambiguous answer to the specific impact of hydrogen addition on catalytic micro-combustion of methane. In order to elucidate the mutual effect between hydrogen and fuel, a methane–air premixed combustion adding hydrogen formed in micro channel is investigated. Furthermore, the thermal and chemical effects of hydrogen addition on combustion of methane–air are studied numerically by a three-dimensional simulation including an elementary-step surface reaction mechanism of rhodium (Rh). This reaction calculates the mass fluxes at the wall due to surface reactions and the surface coverage with species adsorbed by Arrhenius models.

## Problem model and control equation

The micro flatbed combustor ( $20\text{ mm} \times 4\text{ mm} \times 1\text{ mm}$ , wall thickness of  $0.2\text{ mm}$ ) is utilized in this paper. The dimension of the combustion chamber is very small and the gas velocity is slow. So the Reynolds number based on the averaged inlet gas velocity, gas kinematic viscosity at ambient conditions and the characteristic length is approximately 10. Kuo et al. [15] have suggested that laminar model more appropriately estimates the characteristics in micro-reactor when the Reynolds number is below 500, so laminar model is selected in our work. This means that the volume force and dissipative effect in the

flow can be neglected. Meanwhile, the gas radiation is ignored here. The fluid flow is described by the Navier–Stokes equations, an energy conservation equation and additional conservation equations for each chemical species, so mathematical equations are as follows.

$$\text{Continuity equation : } \frac{\partial(\rho u_j)}{\partial x_j} = 0 \quad (1)$$

where  $\rho$  is the density of the gas mixture,  $u$  is the velocity.

$$\text{Composition equation : } \rho u_j \frac{\partial Y_s}{\partial x_j} = \frac{\partial}{\partial x_j} \left( D \rho \frac{\partial Y_s}{\partial x_j} \right) + R_s \quad (2)$$

where  $Y_s$  corresponds to the mass fraction of species  $s$  in micro premix chamber,  $D$  is diffusion coefficient and  $R_s$  represents the consumption or decomposition rate. Both gas phase species and surface species can be generated and consumed by surface reactions. The consumption or decomposition rate  $R_s$  is written as below,

$$R_s = \sum_{k=1}^{K_s} \nu'_{sk} k_{sk} \prod_{i=1}^{N_g+N_s} [X_i]^{\nu''_{ik}} \quad (3)$$

where  $K_s$  is the total number of elementary surface reactions,  $\nu'_{sk}$  is stoichiometric coefficient in forward direction of the reaction  $k$ ,  $\nu''_{ik}$  is stoichiometric coefficient in negative direction of the reaction  $k$ ,  $k_{sk}$  is forward rate coefficient of the reaction  $k$ .  $N_g$  is the number of gas phase species and  $N_s$  is the number of surface species.  $[X_i]$  is the molar concentration of surface species  $i$ .  $k_{sk}$  is calculated by Arrhenius reaction source of the reaction  $k$  as below,

$$k_{sk} = A_k T^{\beta_k} \exp\left(-\frac{E_a}{RT}\right) \prod_{i=1}^{N_s} \Theta_i^{\mu_{ik}} \exp\left[\frac{\epsilon_{ik} \Theta_i}{RT}\right] \quad (4)$$

Here,  $A_k$  is the pre-exponential factor,  $\beta_k$  is the temperature exponent,  $E_a$  is the activation energy of reaction  $k$ ,  $\Theta_i$  is surface coverage rate of species  $i$ ,  $\mu_{ik}$  and  $\epsilon_{ik}$  are surface coverage parameter. In addition,  $[X_i]$  is defined as below.

$$[X_i] = \Gamma \Theta_i \quad (5)$$

Here,  $\Gamma$  is the surface site density of the catalyst. The value of  $2.72 \times 10^{-9} \text{ mol} \cdot \text{cm}^{-2}$  is used for the Rh catalyst in this study. The surface of the catalytic is described by its coverage with adsorbed species and its temperature. The temperature is based on the governing equation for enthalpy as below.

$$\text{Energy equation : } \rho u_j \frac{\partial h}{\partial x_j} = \frac{\partial}{\partial x_j} \left( \lambda \frac{\partial T}{\partial x_j} \right) + \frac{\partial}{\partial x_j} \left( \sum_{s=1}^{N_g} h_s \rho D_s \frac{\partial Y_s}{\partial x_j} \right) + q \quad (6)$$

with  $h$  as enthalpy,  $q$  as heat of reaction and  $\lambda$  as thermal conductivity.

$$\text{Momentum equation : } \frac{\partial}{\partial x_j} (\rho u_j u_i) = -\frac{\partial p}{\partial x_j} \left[ \mu \left( \frac{\partial u_i}{\partial x_j} + \frac{1}{3} \frac{\partial u_j}{\partial x_i} \right) \right] \quad (7)$$

with  $\mu$  as viscosity. This equation system is closed by the ideal gas law.

$$\text{Closure equation : } \sum_{s=1}^{N_g} Y_s = 1 \quad (8)$$

Download English Version:

<https://daneshyari.com/en/article/7717752>

Download Persian Version:

<https://daneshyari.com/article/7717752>

[Daneshyari.com](https://daneshyari.com)