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Optimization of a counter-flow microchannel reactor using hydrogen assisted catalytic combustion for steam reforming of methane

Seung Won Jeon, Won Jae Yoon, Min Woo Jeong, Yongchan Kim*

Department of Mechanical Engineering, Korea University, Anam-Dong, Sungbuk-Ku, Seoul 136-713, Republic of Korea

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ABSTRACT

The objective of this study is to optimize a microchannel reactor using hydrogen assisted catalytic combustion for steam reforming of methane. Hydrogen assisted catalytic combustion does not require preheating because the catalytic combustion of hydrogen occurs at room temperature. After start-up by hydrogen catalytic combustion, fuels of hydrogen and methane were changed to methane. The geometric configuration of the counter-flow reactor was optimized by the simulation model under steady state condition. The hydrogen flow rate in the counter-flow reactor was also optimized by transient simulations using the response surface methodology. As a result, the counter-flow reactor showed extremely short start-up time because of the optimized configuration and the optimized hydrogen flow rate. Hot spots were avoided because of the hydrogen shut-off after start-up. The operating characteristics of the counter-flow reactor were compared with those of the co-flow reactor.

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Introduction

Hydrogen assisted catalytic combustion has been used in the catalytic combustors of gas turbines because of its low light-off temperature [1–4]. Catalytic combustion of hydrogen normally occurs at room temperature [4–6], and that of hydrocarbon occurs at elevated temperatures of 413 K–703 K [7]. Hydrogen addition not only decreases ignition temperature [3,4], but also reduces NO_x emission [3,8,9]. Hydrogen assisted catalytic combustion is also used in portable micro burners [10,11]. In addition, it can be used in a steam reforming reactor for hydrogen production [12–19]. When hydrogen is used as a

fuel for catalytic combustion in a steam reforming reactor, neither preheating devices nor preheating energy is required, unlike in hydrocarbon combustion.

The catalytic combustor operates without additional hydrogen, once ignited by hydrogen [4,11]. However, most steam reforming reactor used hydrogen only as a fuel of catalytic combustion [12–18]. Hwang et al. [19] measured the performance of a steam reforming reactor by changing the fuel from hydrogen to a mixture of methane and hydrogen after start-up. To reduce the energy consumption, the minimum amount of hydrogen should be used by shutting off the hydrogen supply after start-up [20]. The steam reforming reactor should be investigated for various geometric flow

* Corresponding author. Tel.: +82 2 3290 3366; fax: +82 2 921 5439.

E-mail address: yongckim@korea.ac.kr (Y. Kim).

| Nomenclature | | | |
|--------------------|--|------------------|--|
| C_i | molar concentration, kmol m^{-3} | x^* | dimensionless length in x direction |
| $D_{\text{eff},i}$ | effective mass diffusion coefficient of species i, $\text{m}^2 \text{s}^{-1}$ | X_1 | reactor length, m |
| h | enthalpy of mixture, kJ kg^{-1} | X_2 | ratio of uncoated length to reactor length |
| J_i | diffusion flux of species i, $\text{kg m}^{-2} \text{s}^{-1}$ | X_3 | initial H_2/CH_4 |
| k | thermal conductivity, $\text{W m}^{-1} \text{K}^{-1}$ | X_4 | equivalence ratio |
| k_{eff} | effective thermal conductivity, $\text{W m}^{-1} \text{K}^{-1}$ | X_5 | H_2/CH_4 just after start-up |
| $K_{e,j}$ | equilibrium constant | X_6 | slope of H_2/CH_4 after start-up |
| K_i | adsorption constant of species i | y | coordinates in lateral direction, m |
| k_j | reaction rate constant | Y_1 | sum of reforming and combustion conversions |
| p | pressure, N m^{-2} | Y_2 | maximum temperature, K |
| p_i | partial pressure, N m^{-2} | Y_3 | start-up time, s |
| R | gas constant, $\text{kJ kmol}^{-1} \text{K}^{-1}$ | Y_4 | hydrogen consumption during start-up, g |
| R_i | net rate of production of species i by chemical reaction, $\text{kg m}^{-3} \text{s}^{-1}$ | Y_5 | minimum reforming conversion |
| r_j | reaction rate, $\text{kmol m}^{-3} \text{s}^{-1}$ | Y_6 | maximum temperature, K |
| S_h | chemical reaction heat production rate, $\text{kJ m}^{-3} \text{s}^{-1}$ | Y_i | mass fraction of species i |
| S_i | momentum source in porous medium, N m^{-3} | y_i | mole fraction of species i |
| T | temperature, K | Greek | |
| t | time, s | β | coefficient of the second-order response surface model |
| u_i | velocity, m s^{-1} | ΔH_{298} | standard heat of reaction, kJ kmol^{-1} |
| x | coordinates in axial direction, m | μ | viscosity, $\text{N m}^{-2} \text{s}^{-1}$ |
| | | ρ | density, kg m^{-3} |
| | | φ | equivalence ratio |

configurations such as co- and counter-flow because the heat transfer characteristics of co- and counter-flow reactors are completely different; the reaction zones overlap in a co-flow reactor [21], but reactant depletions occur in the opposite direction in a counter-flow reactor [22].

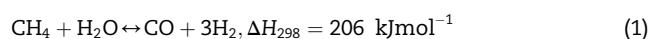
Rapid start-up is a very important issue especially in hydrogen assisted catalytic combustion because the reactant is fed into the reactor at room temperature. The reduction of the start-up time of the steam reforming reactor has been studied using hydrogen [14–16,19] and hydrocarbon fuels [23–25]. Park et al. [15] and Kim [16] investigated start-up time reduction by increasing the hydrogen flow rate. However, the increase in the hydrogen flow rate led to the formation of hot spots [14–16]. Therefore, the hydrogen flow rate should be optimized to reduce the start-up time while minimizing hot spot formations.

In this study, hydrogen assisted catalytic combustion was applied in a microchannel reactor designed for steam reforming of methane. The reactant was fed into the reactor at room temperature without preheating. Hydrogen assisted combustion was used during reactor start-up, and after start-up, hydrogen inflow was shut off. The configuration of the counter-flow reactor was optimized by a simulation model under steady state condition. The optimal hydrogen flow rate was determined to reduce the start-up time in the counter-flow reactor, while satisfying stable fuel transition by using a transient simulation model. The response surface methodology was used for the optimizations. The temperature of the reactor and the reforming conversion were analyzed for various operating conditions. In addition, the performance of the co-flow reactor was compared with that of the counter-flow reactor.

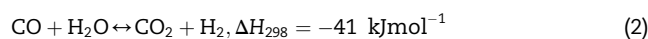
Kinetics of steam reforming and catalytic combustion

A microchannel reactor used for steam reforming of methane can be divided into two channels; namely, the steam reforming channel for hydrogen production and the catalytic combustion channel for heat supply. Methane and hydrogen are used as fuels for catalytic combustion. The reaction in the reforming and the combustion channel is as follows:

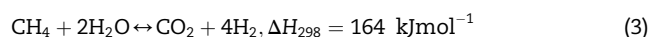
Steam reforming of methane



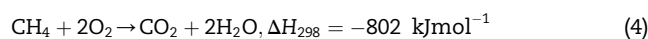
Water-gas shift



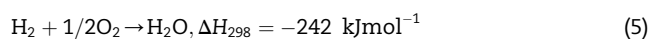
Reverse methanation



Catalytic combustion of methane



Catalytic combustion of hydrogen



The reaction kinetics developed by Xu and Froment [26] on the Ni catalyst was applied in the reforming channel.

$$r_1 = \frac{k_1}{p_{\text{H}_2}^{2.5}} \left(p_{\text{CH}_4} p_{\text{H}_2\text{O}} - \frac{p_{\text{H}_2}^3 p_{\text{CO}}}{K_{e,1}} \right) \quad (6)$$

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