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Numerical simulation of exhaust reforming characteristics in catalytic fixed-bed reactors for a natural gas engine

Zunhua Zhang [a,b], Pengpeng Jia [b], Shangsheng Feng [c], Junjie Liang [b], Yanxiang Long [b], Gesheng Li [a,b,*]

[a] Key Laboratory of High Performance Ship Technology (Wuhan University of Technology), Ministry of Education, Wuhan, Hubei 430063, PR China
[b] School of Energy and Power Engineering, Wuhan University of Technology, Wuhan 430063, PR China
[c] State Key Laboratory for Strength and Vibration of Mechanical Structure, Xi'an Jiaotong University, Xi'an 710049, PR China

HIGHLIGHTS

- The fixed bed reactor is used to produce on-board hydrogen for natural gas engine.
- Molar ratio of H_2/CO first increases and then decreases with the increasing GHSV.
- Average temperature of the mixed gas decreases with the increase of methane content.
- Wall temperature has notable effects on methane conversion and molar ratio of H_2/CO.

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ABSTRACT

Coupled with detailed catalytic reaction mechanism, the exhaust reforming process in a fixed bed reactor was simulated using a porous media model to investigate the reforming characteristics under different initial conditions. The effects of gas hourly space velocity (GHSV), feed component, steam addition and wall temperature on the methane conversion rate, hydrogen yield and other characteristic parameters were analyzed. The simulation results show that the oxygen is consumed rapidly when the reforming gas enters the reaction zone and the steam reforming plays a dominant role in the latter part. The exhaust reforming process mainly involves oxidation reaction, steam reforming reaction and water gas shift reaction. The methane conversion and hydrogen production decrease with the rise of GHSV, while the molar ratio of H_2 to CO first increases and then decreases and reaches its peak value when GHSV ranges from 30,000 h^{-1} to 35,000 h^{-1}. As the ratio of methane to exhaust increases, a higher molar fraction of hydrogen at the outlet can be achieved owing to a bigger proportion of partial reforming reaction of methane, and methane conversion rate decreases. It also suggests that a modest steam addition is preferable for both the reforming performance and the life span of the fixed bed.

1. Introduction

The increasingly stringent emission regulations for internal combustion engines and the gap between supply and demand of petroleum resources make it important to explore more economical and cleaner alternative energy sources. Compared with traditional fuels such as gasoline and diesel, natural gas has the advantages of low sulfur content and carbon intensity, and CO_2 and SO_x emissions are significantly reduced when the natural gas is used to drive the internal combustion engine. In addition, a higher compression ratio can be achieved to obtain a higher thermal efficiency (Zeng et al., 2006). As a result, the natural gas engine has been widely investigated (He et al., 2016; Ma et al., 2008; Ma and Wang, 2008). However, due to the narrow flammability limit of methane and its high ignition energy, the lean burn performance of the natural gas engine becomes worse as a result of the misfire and cycle-by-cycle variations in the engine, which directly affects the stable operation of the engine (Ma et al., 2008; Ma and Wang, 2008). Hydrogen has a fast combustion speed, low ignition energy and wide flammability limits, and therefore hydrogen addition is a good choice for improving the combustion characteristics of natural gas engines (Tartakovsky and Sheintuch, 2018). The previous studies (Ma et al., 2007; Wang et al., 2007; Wang et al., 2008) show that hydrogen addition in natural gas engines can improve the peak of in-cylinder pressure and

* Corresponding author at: School of Energy and Power Engineering, Wuhan University of Technology, Wuhan, Hubei 430063, PR China.
E-mail address: gsliwh@gmail.com (G. Li).

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Nomenclature

A	pre-exponential factor (s^{-1} or m^2/mol s)
C_2	inertial loss factor (m^{-1})
c_i	species concentrations (mol/m^3 or mol/m^2)
$D_{i,m}$ or $D_{i,T}$	mass diffusion coefficient or thermal diffusivity (m^2 s or kg/(m s))
$E_{a,n}$	activation energy (kJ/mol)
h	explicit enthalpy (J/kg)
$\triangle H$	reaction enthalpy (kJ mol^{-1})
$J_{i,j}$	diffusion flux of the species i in j direction (kg/s)
K	permeability coefficient (m^2)
k_{eff}	effective heat transfer coefficient
$k_{f,n}$	dynamic constant of elementary reactions (s^{-1} or m^2/(mol s))
M_i	molecular weight of the species i (kg/mol)
m	mass (kg)
N_g or N_s	species number of gas phase or adsorption phase
p	fluid pressure (Pa)
R	ideal gas constant (J/(K mol))
R_i^{het}	net generation rate of species i (kg/(m^3 s))
Re	Reynolds number
S_V	catalytic surface area of the pore walls per unit volume (m^{-1})
s_i	molar net production rate (mol/(m^3 s))
T	fluid temperature (K)
u_i	velocity in i direction (m/s)
v	stoichiometric coefficient
x_i	coordinate in i direction (m)
Y_i	mass fraction of the specie i
GHSV	gas hourly space velocity (h^{-1})
REGR	Reformed Exhaust Gas Recirculation

Greek symbols

ε	the porosity of the fixed beds
ρ	fluid density (kg/m^3)
μ	dynamic viscosity (Pa s)
Θ	surface coverage
φ	exponent of surface coverage
ξ	activation energy modified by surface coverage (kJ/mol)
γ	initial sticking coefficient
Γ	catalyst active base density (kmol/m^2)

shorten the combustion duration to obtain a better performance on thermal efficiency and emission characteristics.

Through the reforming reaction of the gas mixture consisting of exhaust gas and extra fuel in the presence of catalyst, Reformed Exhaust Gas Recirculation (REGR) technology provides a good alternative way to achieve on-board hydrogen production to improve in-cylinder combustion. It has been widely concerned owing to the increase of overall fuel economy (Golunski, 2010; Leung et al., 2010; Fennell et al., 2015; Liao and Horng, 2016; Moazami et al., 2013). The feasibility of this technology was confirmed by some studies on the closed loop exhaust-gas fuel reforming, and it suggested that REGR can reduce NO_x emissions and expand the lean burning limits (Yap et al., 2006; Zhang et al., 2017). In addition, this technology can be applied with widely-used fossil fuels such as gasoline, diesel and natural gas (Zhang et al., 2017; Poran and Tartakovsky, 2017; Sall et al., 2013).

The main components of exhaust gas include CO_2, O_2, H_2O and N_2, and it means exhaust reforming process may involve dry reforming, steam reforming and oxidation reforming of methane. The exhaust reforming of natural gas in the microchannel reactor has been simulated with Langmuir-Hinshelwood-Hougen-Watson (LHHW) reaction model (Peucheret et al., 2005; Bulutoglu et al., 2016), which conducted a parametric investigation on effects of the initial conditions on the hydrogen production characteristics. However, the parameters of the reaction kinetics are questionable (Wehinger et al., 2014; Wehinger et al., 2015). It is suggested that reliable kinetics is essential due to the interaction between the chemistry and transport phenomenon. Coupled with detailed surface catalytic reactions, the exhaust reforming process in a reforming tube with catalyst coated on the inner wall was reported (Zhang et al., 2017), which gave insight into the effects of EGR ratio and molar ratio of water to methane on the reforming characteristics. Besides the microchannel reactor, typical reactors for the catalytic reforming reaction include fixed beds, foams, microchannel reactors and fluidized-bed reactors. The fixed-bed reactor is the most common type of reactors in the catalytic industry, in which steam reforming reaction, dry reforming reaction and autothermal reforming of methane for hydrogen production can be performed with catalyst coated on the surface of the pellets (Wehinger et al., 2015; Halabi et al., 2008; Dixon, 2017; Benguerba et al., 2015), and obviously it is also a feasible choice for the exhaust reforming reaction. However, there are few studies focusing on the hydrogen production purpose for the Reformed Exhaust Gas Recirculation technology.

The fixed-bed catalytic reactor, which is applied to the exhaust reforming process, has a complex arrangement for the randomly packed catalytic pellets and it is difficult to generate three-dimensional grids due to the irregular porous structure and the contact points between pellets for the reactor. In addition, the computational resources for the full-size reactor are expensive, and it is unnecessary for the study on the reforming characteristics under different boundary conditions. Therefore in this work, the reactor was represented with a 2-D model and the catalytic pellets were simulated using the porous media model in the ANSYS-Fluent 15.0. The reforming reaction in the reactor was described with a detailed catalytic reaction mechanism. The aim of this study is to investigate the effects of GHSV, methane to exhaust ratio, steam addition and other parameters on the reforming performance, which may lead to a better operating strategy for the reformer and serve as a reference for the further research on the Reformed Exhaust Gas Recirculation technology.

2. Model establishment and validation

2.1. Model description

The exhaust gas of natural gas engine includes CO_2, H_2O, O_2, N_2, NO_x, CO and many other compounds, and the content of each component in the exhaust and its energy are different under various conditions. This simulation selected the emission data of a modified YC6MK 200 N natural gas engine at 75% propulsion load to carry out the correlation simulation. The exhaust gas was simplified as CO_2, H_2O, O_2 and N_2 according to the experimental emission test of the natural gas engine, and their contents were 6.71%, 13.42%, 6.15% and 72.72%, respectively. The content of the remaining components including NO_x, CO and unburned HC was less than 0.5% so that it was negligible in the subsequent simulations.

The catalysts in the exhaust reforming reaction are divided into noble and non-noble metal catalyst. The extensively used Ni-based