

Performance analysis of a multi-tube exhaust waste heat recovery methanol reforming to hydrogen reactor

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Abstract. To address hydrogen storage and transport challenges, a methanol reforming hydrogen reactor featuring multi-pipe exhaust heat recovery was simulated using Fluent. Initial simulations focused on microscopic flow, heat transfer, and reaction dynamics within a catalyst particle stack. Subsequent macroscopic analyses of a comprehensive reactor model evaluated how reactant inlet velocity, molar ratio of steam to methanol vapor (S/M), exhaust temperature, and flow impact reforming performance. Findings indicate that smaller catalyst particles enhance heat transfer and reaction at the cost of increased pressure drop, with an optimal size of 4.4 mm. Methanol conversion correlates positively with S/M, exhaust temperature, and flow, but negatively with inlet velocity. Hydrogen production increases then decreases with S/M, showing positive correlation with inlet velocity, exhaust temperature, and flow. Carbon monoxide selectivity is inversely related to inlet velocity and S/M, and positively to exhaust temperature and flow.

Keywords: Exhaust waste heat recovery; Methanol reforming to hydrogen; Multi-pipe reactor; Numerical simulation.

1. Introduction

Hydrogen is recognized as a "zero-emission fuel" with advantages such as rapid flame propagation, low ignition energy, and swift diffusion rate, making it an excellent alternative fuel^[1,2]. However, hydrogen's characteristics, including flammability, explosiveness, and propensity for leakage, pose significant challenges for its storage and transportation, thus impeding the development of hydrogen fuel^[3]. The integration of methanol reforming technology with internal combustion engines—leveraging exhaust waste heat for methanol reforming to produce hydrogen—not only enhances energy utilization but also facilitates on-site hydrogen production^[4,5]. This can be used for hydrogen-enhanced combustion in engines to improve performance, effectively addressing the issue of hydrogen storage and transport. Liao et al.^[6] investigated the on-line methanol reforming system based on exhaust waste heat recovery, finding that engine exhaust temperature and flow were sufficient for on-line methanol reforming for hydrogen production. Srivastava et al.^[7] explored the impact of parameters such as reactant temperature, velocity, S/M, and heating-side exhaust temperature on the reactor's reforming performance, identifying heat as the most influential factor in enhancing methanol conversion. Wu et al.^[8] examined a packed bed reactor equipped with various internal helical fins, demonstrating that the installation of helical fins significantly improves energy utilization efficiency in the hydrogen production process. Presently, most simulation studies on methanol reforming reactors employ porous media models to simplify the catalyst component, which does not accurately reflect the actual state of the catalyst bed. Moreover, while most reactors used for methanol reforming are single-tubular^[9,10] or involve new structures^[11-13], few studies have focused on multi-tubular reactors, which offer larger heat transfer coefficients, compact design, and minimal space requirements. This study aims to design a multi-tube process exhaust waste heat recovery methanol reforming to hydrogen reactor and to examine the influence of key parameters on reforming performance. Initially, a catalyst particle stacking model was developed to simulate and analyze gas flow between catalyst particles, heat exchange, and surface chemical reactions from a microscopic perspective. Subsequently, a full reactor model was constructed to investigate the effects of reactant inlet velocity, S/M, exhaust temperature, and flow on the reforming performance from a macroscopic viewpoint. The findings of this study provide a reference for the application of multi-tube process

reactors in utilizing exhaust waste heat reforming for hydrogen production to achieve hydrogen doping combustion in engines.

2. Methodology

2.1 Geometry

Exhaust data from a water-doped methanol engine, converted from a naturally aspirated 186FA diesel engine under varying load conditions, were acquired through experimentation. Table 1 presents the exhaust data across different load scenarios.

Table 1. Exhaust data under different engine conditions

Speed (rpm)	Engine load (%)	Water content (%)	Exhaust temperature (K)	Exhaust flow (kg/s)
3000	50	20	536	0.0106
3000	75	20	595	0.0101
3000	100	20	659	0.0099

Utilizing the exhaust energy at three distinct loads, the optimal hydrogen doping ratio was identified, and the reactor's design was tailored to fulfill the energy requirements for methanol reforming. Initially, the reactant inlet velocity was set at 1.5 m/s, with methanol and water molar fractions both at 50%, the exhaust temperature at 593 K, and the exhaust flow at 0.01 kg/s.

Fig. 1(a) illustrates the schematic of the catalyst particle stacking model, featuring an inlet section length of 20 mm, a catalyst stacking length of 30 mm, and an outlet section length of 30 mm. Six particle models of varying sizes were developed to assess the impact of catalyst particle size and to ascertain the optimal size. Fig. 1(b) depicts the schematic of the complete multi-tube range reactor model, with a reactor diameter of 20 mm, a total length of 2500 mm, a shell length of 515 mm, and a wall thickness of 2 mm.

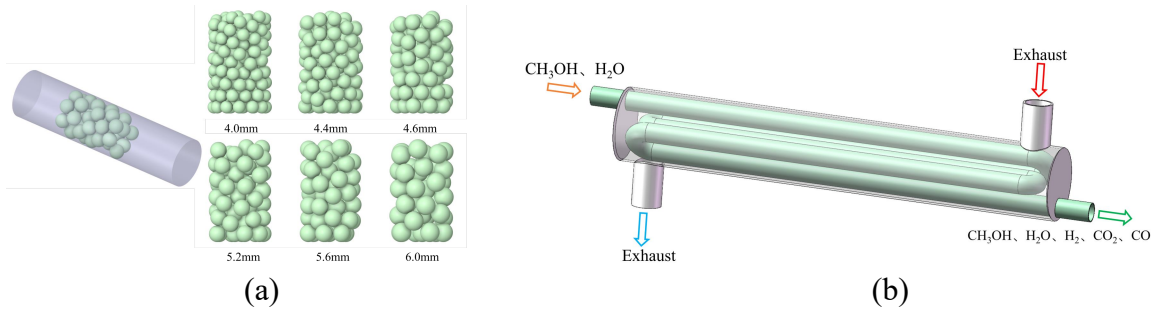


Fig. 1. Reactor geometry

2.2 Governing equations and simulation settings

The simulation study was predicated on the assumptions that all gases behave as ideal gases; gas flow is laminar, incompressible, and maintains a steady state; the impacts of volume and gravity are disregarded; radiative heat transfer is excluded; and the catalyst bed is isotropic and homogeneous with uniform morphological properties. Furthermore, it is assumed that chemical reactions occur exclusively within the catalyst.

The governing equations for the simulation include the continuity equation, momentum equation, species concentration equation, and energy equation, as delineated below:

Continuity equation:

$$\nabla \cdot \vec{V} = 0$$

Momentum equation:

$$\rho_f \varepsilon (\vec{V} \cdot \nabla) \vec{V} = -\varepsilon \nabla p + \varepsilon \mu \nabla^2 \vec{V} + S_m$$

Species concentration equation:

$$\varepsilon (\vec{V} \cdot \nabla) C_i = D_{eff} \nabla^2 C_i + \varepsilon S_R$$

Energy equation:

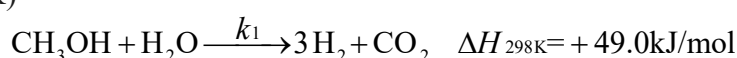
$$(\rho_f c_p)(\vec{V} \cdot \nabla)T = \lambda_{eff} \nabla^2 T + \varepsilon S_t$$

ε represents the bed porosity; ρ_f denotes the catalyst density, kg/m³; μ is the kinetic viscosity, kg/(m·s);

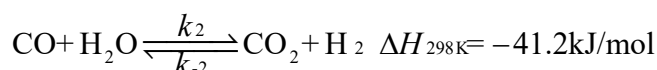
S_m signifies the momentum source term induced by the porous catalyst, N/m³; C_i is the concentration of the i^{th} species, mol/m³; D_{eff} is the effective diffusion coefficient, m²/s; S_R represents the source term of the components generated by the chemical reaction, mol/(m³·s); c_p denotes the specific constant pressure heat capacity, J/(kg·K); λ_{eff} is the effective thermal conductivity, J/(kg·K); T stands for temperature, K; S_t is the heat of reaction, W/m³.

This study utilized a double rate model to simulate the methanol steam reforming reaction, encompassing both the methanol steam reforming (SR) and reversible water gas shift (WGS) reactions, described by the respective chemical equations. Chemical kinetic parameters are provided in Table 2.

Steam reforming (SR)



Water gas shift(WGS)



For the evaluation of reaction rates, the Arrhenius model was employed^[14,15], with rate expressions for the SR and WGS reactions presented in Eqs (1) and Eqs (2) In these equations, R is the universal gas constant, k the pre-exponential factor, and E_a the activation energy.

$$R_{\text{SR}} = k_1 C_{\text{CH}_3\text{OH}}^{0.6} C_{\text{H}_2\text{O}}^{0.4} \exp\left(-\frac{E_{a1}}{RT}\right) \quad (1)$$

$$R_{\text{WGS}} = k_2 C_{\text{CO}} C_{\text{H}_2\text{O}} \exp\left(-\frac{E_{a2}}{RT}\right) - k_{-2} C_{\text{CO}_2} C_{\text{H}_2} \exp\left(-\frac{E_{a-2}}{RT}\right) \quad (2)$$

Table 2. Chemical kinetics parameters

Parameter	Unit	Value
Pre-exponential factor for SR, k_1	s ⁻¹	8×10^8
Pre-exponential factor for forward WGS, k_2	m ³ /(mol·s)	4×10^8
Pre-exponential factor for reverse WGS, k_{-2}	m ³ /(mol·s)	4×10^8
Activation energy for SR, E_{a1}	J/mol	7×10^8
Activation energy for forward WGS, E_{a2}	J/mol	1×10^8
Activation energy for reverse WGS, E_{a-2}	J/mol	1×10^8

2.3 Mesh independence verification and model verification

Numerical simulations were conducted utilizing a pressure-based steady-state solver in ANSYS Fluent, with the governing equations discretized via the finite volume method (FVM). Species transport models, inclusive of finite-rate kinetic models or turbulence-free chemical reaction (TCI) models, were selected for the reactions. The SST k-omega model was employed to account for potential turbulence or disturbances within the curved channel. Boundary conditions were established as velocity inlet and pressure outlet. Pressure-velocity coupling was achieved through the SIMPLE-C scheme, with the PRESTO! scheme applied for pressure term discretization, and a second-order upwind scheme utilized for momentum, component, and energy equations. A convergence criterion maintained all residual values below 10⁻⁶. An initial grid-independence validation was conducted, with Table 3 illustrating the impact of grid count on simulation outcomes under identical conditions. Observations with grid number 581,429 demonstrated a variance of less than 0.5% compared to those with grid number 932722, leading to the selection of the 581429 grid model for further study.

Table 3. The effect of the number of grids on the simulation

Number of grids	Mole fraction of CH ₃ OH (%)	Mole fraction of H ₂ (%)
403596	13.23	54.04

581429	13.08	54.24
932722	13.97	54.39

The model's accuracy was substantiated through comparison with the findings of Debanik Bose et al.^[16]. Input parameters were aligned with those specified in the original study, and both methanol conversion and hydrogen mass fraction were compared against original data as depicted in Fig. 2, with discrepancies within 5%. This comparison validates the accuracy of the developed model.

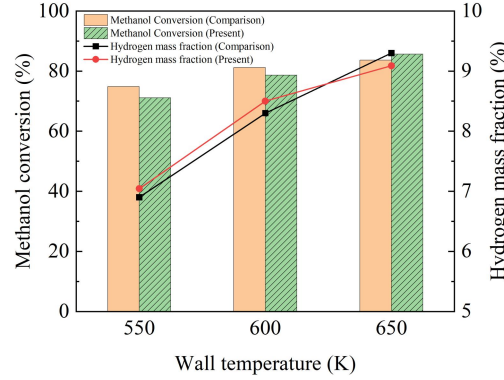


Fig. 2. Validation of the formulated numerical model

3. Results and discussion

To better analyze and explain the results, the following evaluation indicators are defined.

Methanol conversion: $\eta_{\text{CH}_3\text{OH}}$, %.

$$\eta_{\text{CH}_3\text{OH}} = \frac{MF_{\text{CH}_3\text{OH},\text{in}} - MF_{\text{CH}_3\text{OH},\text{out}}}{MF_{\text{CH}_3\text{OH},\text{in}}} \times 100\%$$

$MF_{\text{CH}_3\text{OH},\text{in}}$: reactant inlet methanol mass fraction, %;

$MF_{\text{CH}_3\text{OH},\text{out}}$: reactant outlet methanol mass fraction, %.

Hydrogen production: Y_{H_2} , mol/h:

$$Y_{\text{H}_2} = Q_{v,\text{out}} \times C_{\text{H}_2}$$

$Q_{v,\text{out}}$: volume flow of hydrogen at the reactant outlet, m³/h;

C_{H_2} : molar concentration of hydrogen at the reactant outlet, mol/m³.

Hydrogen production per unit of methanol: $IOR_{\text{CH}_3\text{OH}}$, mol/kg:

$$IOR_{\text{CH}_3\text{OH}} = \frac{Y_{\text{H}_2}}{M_{\text{CH}_3\text{OH},\text{in}}}$$

$M_{\text{CH}_3\text{OH},\text{in}}$: mass flow of methanol at the inlet of the reactants, kg/h.

Carbon monoxide selectivity: X_{CO} , %:

$$X_{\text{CO}} = \frac{mf_{\text{CO},\text{out}}}{mf_{\text{CO},\text{out}} + mf_{\text{CO}_2,\text{out}}} \times 100\%$$

$mf_{\text{CO},\text{out}}$: mole fraction of carbon monoxide at the reactant outlet, %;

$mf_{\text{CO}_2,\text{out}}$: mole fraction of carbon dioxide at the reactant outlet, %.

3.1 Effect of particle size

The particle stacking model was employed to assess the impact of particle size on reforming performance, with the necessary reaction heat provided by a thermostatic wall surface at 593 K. Fig. 3 and Fig. 4 illustrate the schematic diagrams for the variation curves of catalyst bed pressure drop and wall convective heat transfer coefficient across different particle sizes, alongside the pressure variation cloud diagram of the catalyst bed, respectively. The figures indicate a decrease in pressure drop within the catalyst bed with an increase in particle size, where the pressure drop declines from 1.18 Pa to 0.50 Pa as particle size expands from 4.0 mm to 6.0 mm. This reduction is attributed to

larger particles allowing for a greater gas circulation volume, thereby reducing flow resistance and consequent pressure drop.

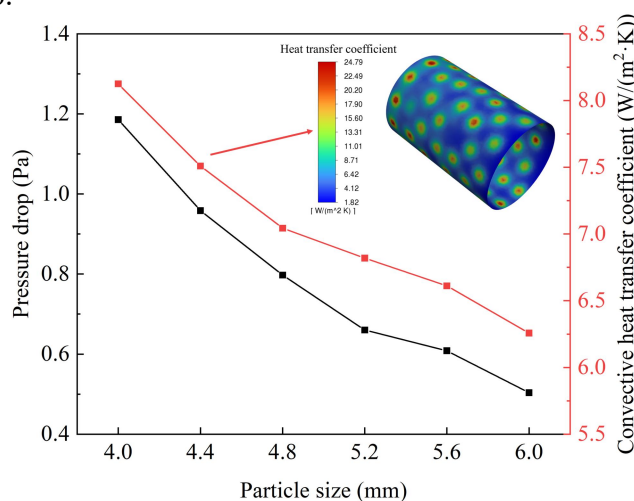


Fig. 3. Pressure drop and wall convection heat transfer coefficients for different particle sizes

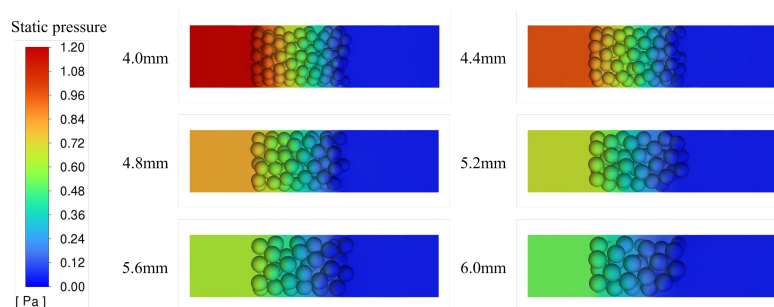


Fig. 4. Pressure in the catalyst bed for different particle sizes

Fig. 5 depicts the variation in chemical reaction rate per unit area and hydrogen mass fraction at the catalyst bed outlet for differing particle sizes. A higher reaction rate per unit area signifies improved catalyst utilization and economic efficiency. As demonstrated in Fig. 5, the reaction rate per unit area initially decreases then increases with particle size enlargement, while the hydrogen mass fraction at the outlet diminishes with particle size growth. Specifically, the hydrogen mass fraction decreased from 6.79‰ to 4.62‰ as the particle size was enlarged from 4.0 mm to 6.0 mm. This trend is due to larger catalyst particles reducing the number of catalyst particles per unit mass or volume, consequently diminishing the catalyst's specific surface area and the number of available active sites for the reaction, thereby reducing the reaction rate. Nonetheless, a continual increase in catalyst particle size also raises the porosity of the catalyst bed, enhancing gas flow and reactant diffusion, which in turn augments the reaction rate.

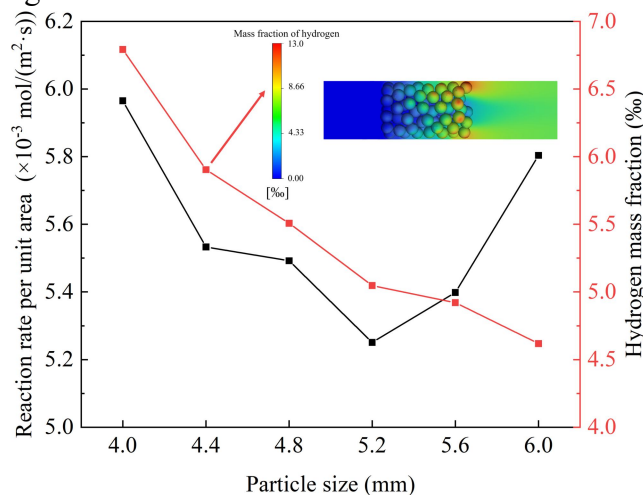


Fig. 5. Chemical reaction rates and hydrogen mass fraction at the outlet for different particle sizes

Given these observations, catalyst particles with a size of 4.4 mm were chosen for further investigation, taking into account factors such as pressure drop, heat transfer efficiency, reaction rate, and economic considerations of the catalyst bed. To conserve computational resources, the porous medium model was utilized to simplify the representation of the catalyst bed layer composed of solid particles in the simulation of the full model of the multi-pipe reactor.

3.2 Effect of reactant inlet velocity

As depicted in Fig. 6.(a), methanol conversion experienced a decline from 66.42% to 51.90% with an increase in the inlet velocity of reactants from 1.2 m/s to 1.8 m/s. This reduction was attributed to a decrease in the residence time of the reactants within the reactor, falling from 1.28 s to 0.92 s, which limited the reactants' ability to sufficiently absorb heat from the exhaust, subsequently lowering the temperature and reducing methanol conversion. Moreover, the elevated inlet velocity resulted in an increased mass flow of the reactants, thereby imposing a higher demand on the same mass of catalyst for catalyzing an augmented quantity of reactants, which diminished catalytic efficiency and, consequently, the methanol conversion.

Fig. 6.(b) illustrates that with the elevation of inlet velocity, the hydrogen production ascended from 37.84 mol/h to 44.87 mol/h, while the hydrogen production per unit of methanol decreased from 60.51 mol/kg to 47.84 mol/kg. This outcome was due to the higher inlet velocity facilitating a greater quantity of methanol to engage in the reforming reaction, leading to an enhanced hydrogen production and a reduced hydrogen production per unit of methanol.

From Fig. 6.(c), it is observable that an increase in inlet velocity from 1.2 m/s to 1.8 m/s corresponded with a decrease in carbon monoxide selectivity from 8.10% to 4.72%. This phenomenon is primarily because the reactants of the reverse water gas shift (rWGS) reaction, hydrogen and carbon dioxide, decrease in mole fractions with an increase in inlet velocity, resulting in a diminished mole fraction of produced carbon monoxide and, thus, a lower carbon monoxide selectivity.

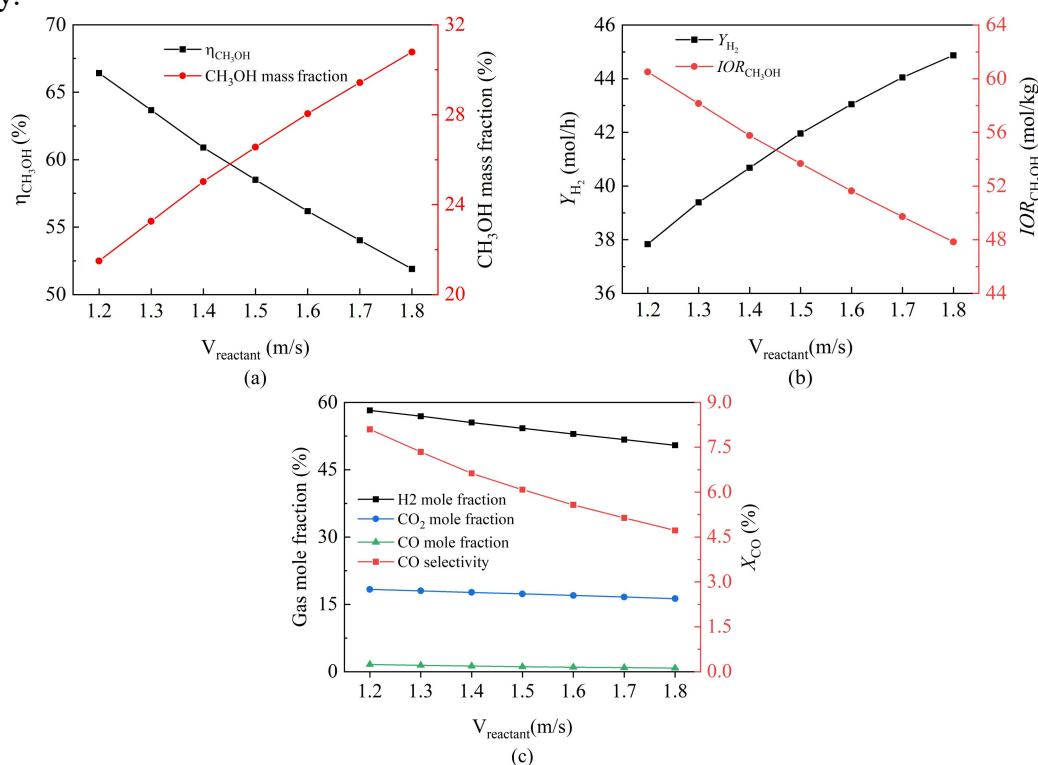


Fig. 6. Effect of reactant inlet velocity on reforming performance

3.3 Effect of S/M

Fig. 7.(a) demonstrates that methanol conversion is almost linearly correlated with the S/M. At lower S/M, the water mole fraction in the reactants is less than that of methanol, hindering complete reaction of methanol and resulting in a reduced conversion rate at these lower ratios. Conversely, at higher S/M, the water mole fraction surpasses the methanol mole fraction, enabling methanol molecules to interact with more water molecules and promoting reaction participation, leading to an increase in methanol conversion from 48.56% to 65.68% as the S/M rose from 0.7 to 1.3.

As indicated in Fig. 7.(b) and Fig. 7.(c), increasing the S/M from 0.7 to 1.3 resulted in a combination of rising methanol conversion and decreasing methanol mass flow in the reactants, culminating in an initial increase followed by a decrease in hydrogen production, peaking at 41.95 mol/kg at a S/M of 1.0. This adjustment in the S/M from 0.7 to 1.3 also led to an increase in hydrogen production per unit of methanol from 44.23 mol/kg to 60.51 mol/kg. The initial rise and subsequent fall in hydrogen production, alongside the decrease in methanol mass flow, contributed to a gradual deceleration in the growth trend of the hydrogen production per unit of methanol.

As revealed in Fig. 7.(d), lower S/M are associated with higher carbon monoxide selectivity, peaking at 8.16% and reaching a minimum of 4.87%. This trend is attributed to the reduced water mole content at lower ratios, which diminishes the inhibition of the rWGS reaction compared to higher water mole content at elevated ratios, thereby generating more carbon monoxide at lower S/M and resulting in greater carbon monoxide selectivity.

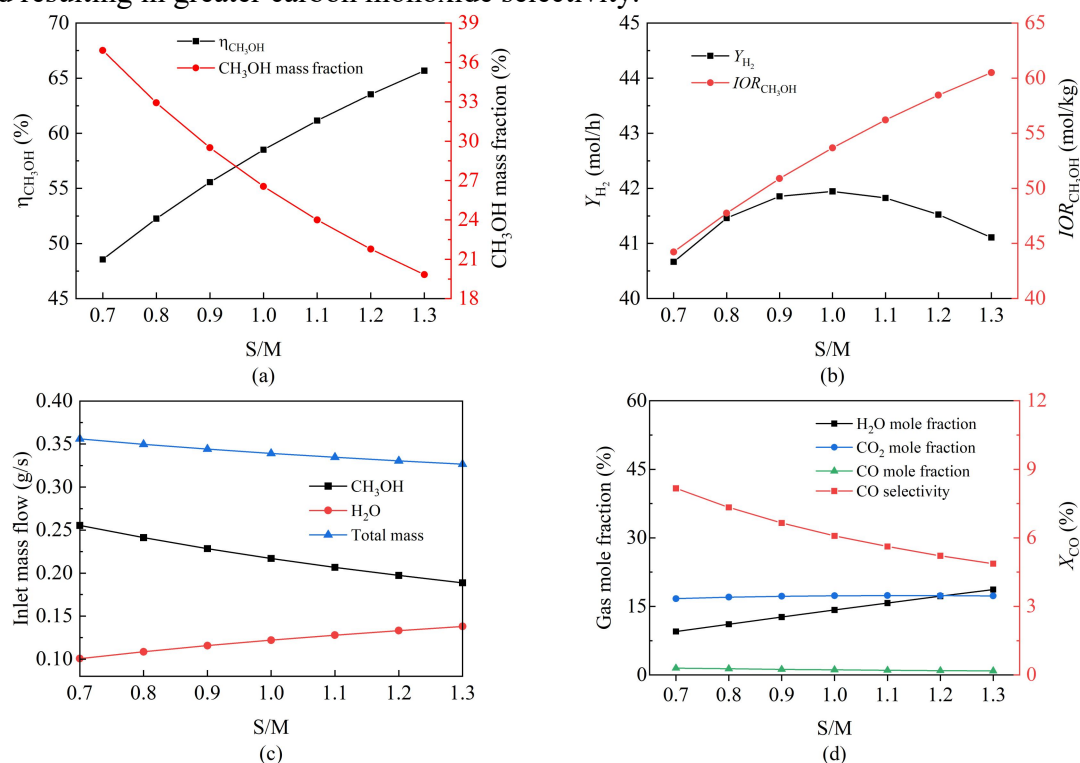


Fig. 7. Effect of S/M on reforming performance

3.4 Effect of exhaust temperature

Fig. 8.(a) and Fig. 8.(b) reveal that methanol conversion, hydrogen production, and hydrogen production per unit of methanol exhibit an upward trend with an increase in exhaust temperature from 533K to 653K. The methanol conversion escalated from 38.94% to 71.24%; the hydrogen production climbed from 28.39 mol/h to 49.38 mol/h; and the hydrogen production per unit of methanol increased from 36.33 mol/kg to 63.17 mol/kg. Enhancing the temperature gradient between the hot and cold fluids boosts heat exchange, elevating the reactants' temperature. An increase in reactants' temperature augments the average kinetic energy of molecules and the effective collision number, accelerating the SR reaction rate, consuming more methanol, and

generating additional hydrogen. Furthermore, a rise in exhaust temperature also elevates the reaction zone temperature and catalyst activity, reducing reaction activation energy and further accelerating the reaction rate, significantly improving methanol conversion, hydrogen production, and hydrogen production per unit of methanol.

Fig. 8(c) indicates that carbon monoxide selectivity increases with the rise in exhaust temperature. Elevated reactant temperatures enhance the SR reaction rate, increasing carbon dioxide and hydrogen content while decreasing water content. Moreover, WGS reaction is exothermic, and the rWGS reaction is endothermic. Based on the principle of reversible reaction equilibrium shift, changes in external conditions prompt the reaction to counteract these changes; hence, an increase in exhaust temperature favors the rWGS reaction, producing more carbon monoxide and thus raising carbon monoxide selectivity.

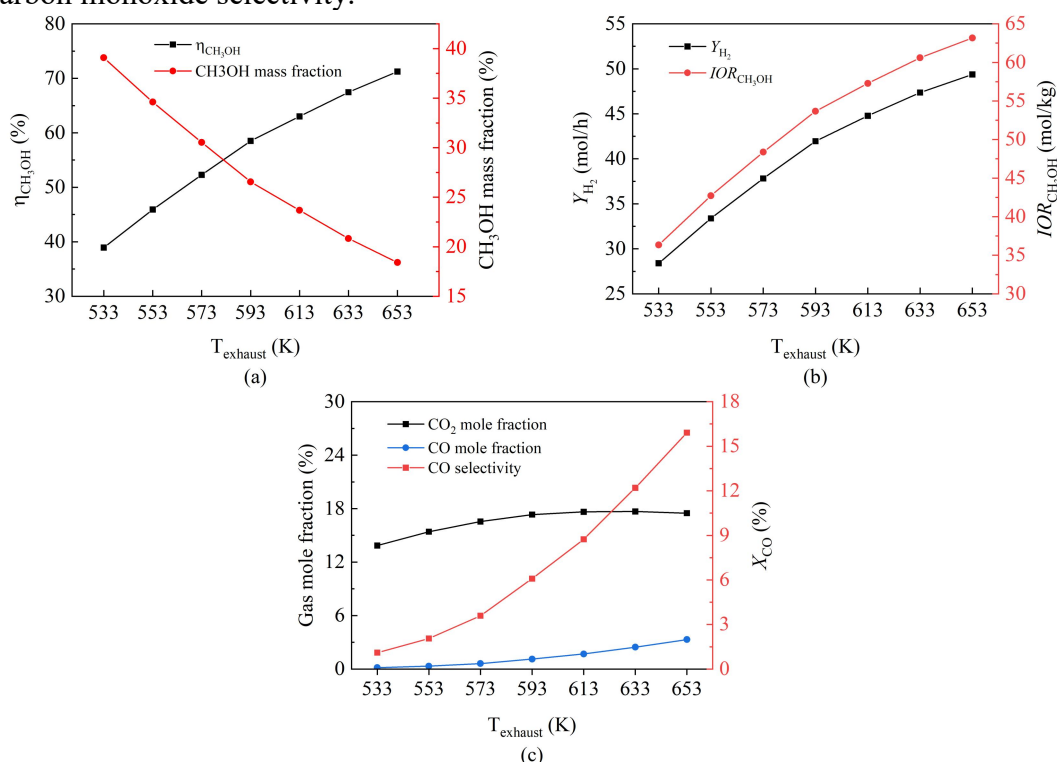


Fig. 8. Effect of exhaust temperature on reforming performance

3.5 Effect of exhaust flow

As demonstrated in Fig. 9(a) and Fig. 9(b), methanol conversion, hydrogen production, and hydrogen production per unit of methanol increase with the rise in exhaust flow. When the exhaust flow escalated from 0.007 kg/s to 0.013 kg/s, the methanol conversion grew from 53.74% to 61.30%, the hydrogen production from 38.79 mol/h to 43.76 mol/h, and the hydrogen production per unit of methanol from 49.62 mol/kg to 55.98 mol/kg. The augmented exhaust flow enhances the convective heat transfer coefficient on the exhaust side, thereby improving heat transfer between exhaust and reactants. Consequently, reactants absorb more heat from the exhaust, leading to increased temperatures that promote the SR reaction and methanol conversion. Simultaneously, the increased exhaust flow also facilitates the occurrence of the rWGS reaction, with a corresponding increase in carbon monoxide selectivity.

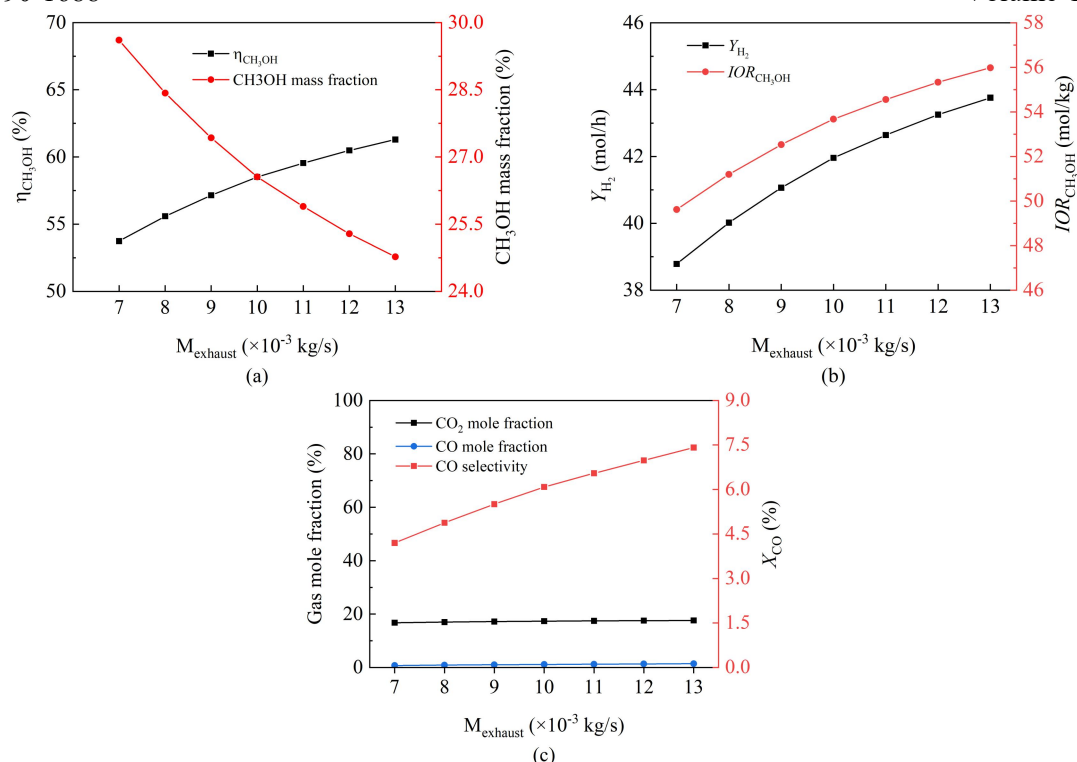


Fig. 9. Effect of exhaust flow on reforming performance

4. Summary

A multi-tubular methanol reforming reactor for hydrogen production, utilizing exhaust waste heat recovery, was designed to investigate the impact of various key parameters on reforming performance. The findings indicate that:

1. Smaller catalyst particle sizes enhance heat exchange and hydrogen production from the reforming reaction, albeit at the cost of increased pressure drop, with an optimal particle size determined to be 4.4 mm.
2. The methanol conversion declined from 66.42% to 51.90% with an elevation in reactants' inlet velocity, whereas the hydrogen production escalated from 37.84 mol/h to 44.87 mol/h as inlet velocity increased.
3. An increase in the S/M positively influenced methanol conversion and the hydrogen production per unit of methanol; the hydrogen production demonstrated a pattern of initial increase followed by a decrease with an augmentation in S/M, peaking at 41.95 mol/kg at a ratio of 1.0.
4. Elevating the exhaust temperature and flow contributed to improvements in methanol conversion, hydrogen production, and hydrogen production per unit of methanol for the reactor, albeit concurrently resulting in an augmented carbon monoxide selectivity.

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