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Effect of Divergent Fuel Channel on the Performance of a Plate-Shape Methanol Steaming Reformer

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Keywords: divergent fuel channel, methanol steaming reformer, hydrogen production

ABSTRACT

A three-dimensional numerical analysis is performed to estimate the effect of divergent fuel channel on the reforming performance and non-isothermal reactant transport in a plate-shape reactor under methanol steam reforming and water-gas-shift reactions. The hydrogen yield was then employed to evaluate the net power output from a proton exchange membrane (PEM) fuel cell. These results display that the application of divergent fuel channel to plate-shape reactors plainly betters the hydrogen production from a reformer and net PEMFC power output. As compared to an ordinary reactor, the novel reactor with the divergent fuel channel of H_{out}/H_{in} = 4.0 creates the most notable improvement in the hydrogen production of 24.03 %. Furthermore, this novel reactor also gains the greatest advance of 6.39 % in the net power output form a PEMFC under implementing the pressure drop for the reactor channel.

INTRODUCTION

A PEMFC power source utilizes hydrogen and oxygen as the fuel to provide the advantages of zero-carbon emission, low-operating temperature, and high-energy efficiency (Wu, 2016; Yang et al., 2019). About the hydrogen fuel, methanol owns significant benefits of higher hydrogen-to-carbon ratio and lower-reforming temperature (Ouyang et al., 2017). Huang et al. (2013) integrated microfluidic channels and Pt catalytic layers in a co-fired ceramic methanol reformer to study the effect of Pt catalyst thickness. A PEMFC stack combined with a miniature plate methanol steam reformer (MSR) is generally regarded as a major portable power source (Zhang et al., 2018). Accordingly, designing a plate MSR is significant to advance the hydrogen yield from MSR and raise the net power output from PEMFC.

The plate-shape wall-coated reactor possesses the benefits of higher ratio of surface area to volume, uniform flow distribution, and lower pressure drop as compared with the packed-bed cylindrical reactor (Ke et al., 2019). Therefore, the plate-shape reactor is developed to augment the heat transfer inside the reactor to promote the catalytic reaction. The application of divergent channel to the reactor had been mentioned for improving chemical reaction within the catalyst in numerous literatures. Chowdhury and Akansu (2017) applied novel convergent and divergent serpentine channels to PEMFC bipolar plates. The experimental results revealed that this novel channel can enhance PEMFC performance. Perng et al. (2017) investigated numerically the heat and fluid flow, methanol conversion, hydrogen production, as well as net power of fuel cell through installing a diffuser in front of the reactor. Their results showed that the diffuser evidently improves methanol conversion and hydrogen production of a cylindrical MSR, because of increasing the residence time of fuel and enhancing the mixing of fuel within the reactor. From the findings of the cited literatures, the divergent channel can clearly perform superior catalytic reaction as compared to the ordinary straight-channel due to longer residence time of fuel and better mixing of fuel.

For lowering the cost of experiments, the scholars commonly performed numerical methods to simulate the catalytic reaction and flow filed within the reactor during fuel reforming. A CFD model (Ji et al., 2018) was developed to simulate the catalytic reaction of methane steam reforming inside a sorption enhanced membrane reactor. Perng et al. (2019) estimated numerically the influences of bottomed ribs along the fuel channels on the gained hydrogen and net PEMFC power. Fluent software was applied to evaluate how the operating and the geometric parameters influenced the

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reforming performance of a micro reactor under methanol steam reforming (Jang et al., 2010). Chang et al. (2019) solved mass balance, chemical reactions, and conjugated heat transfer in a double-pipe type of reformer with CFD code. The above cited literatures revealed that the numerical methods could simulate reasonably the thermal-flow fields and the catalytic reaction processes inside the reactor. However, they have never explored how a divergent fuel channel advanced the hydrogen yield and evaded a larger pressure drop to better the net PEMFC power output.

Consequently, the motive of this study is to evaluate how a divergent fuel channel affects the hydrogen yield, the CO emission, and the net PEMFC electric-power output from various reactors during methanol steam reforming.

CFD SIMULATING MODEL

This CFD simulating model, which concerns continuity, momentum, energy, and species equations, is employed to simulate the thermal-flow fields along the fuel channels inside various plate-shape reactors during methanol steam reforming (SR) and water-gas-shift (WGS) reactions. In Figure 1, the novel plate-shape reactor is invented through arranging a divergent channel in an ordinary plate-shape reactor. The main geometric parameter is the ratio of channel-outlet height to channel-inlet height (H_{out}/H_{in}). This parameter is considered as 4.0, 2.0, 1.33, and 1.0 with constant channel-outlet height and space velocity (SV). H_{out}/H_{in} = 1.0 indicates an ordinary channel. The catalyst layer is mantled above the fuel channel and contains stationary catalyst within the reactor. The CH_3OH blended with H_2O is supplied into the fuel channel at z = 0, and the positive z means the stream-wise direction along the channel.

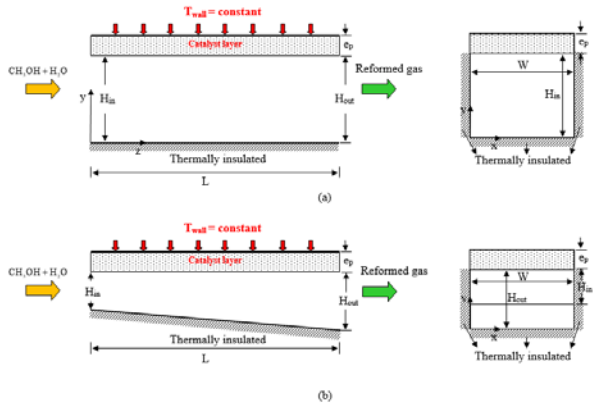

Fig. 1. Computational domains of (a) ordinary fuel channel and (b) divergent fuel channel of a reactor.

The assumptions of this simulating model for this reactor are listed as follows.

(1) The reacting fluid is ideal gas.
(2) The thermal flow field is laminar, incompressible, and steady.
(3) A homogeneous and isotropic catalyst is considered in this study.
(4) All chemical reactions carry out merely within the catalyst layer.
(5) As compared with the convection, the radiation and conduction is disregarded at the gas state.

The principle equations of this 3D simulating model for this reactor are depicted through the subsequent equations.

Continuity equation:
$$\nabla \bullet \vec{u} = 0 \quad (1)$$

Momentum equation:
$$\varepsilon\left(\vec{u}\bullet\nabla\right)\vec{u} = -\frac{\varepsilon}{\rho_{mix}}\nabla p + \frac{\varepsilon\mu_{mix}}{\rho_{mix}}\nabla^2\vec{u} - \frac{\mu_{mix}}{\rho_{mix}\kappa}\vec{u} - \frac{\beta\vec{u}}{2}\left|\vec{u}\right| \quad (2)$$

Species equation:
$$\varepsilon\left(\vec{u}\bullet\nabla\right)c_i = D_{eff}\nabla^2 c_i + \varepsilon\sum_{r=1}^{n} M_{w,i}R_{i,r} \quad (3)$$

where κ, ε, and β mean separately the permeability, the porosity, and the inertial loss coefficient of the catalyst layer. ρ_{mix} and μ_{mix} denote the density and viscosity of mixed gas shown as Eqs. (4)-(6). c_i means the molar concentration of i-th species. D_{eff} indicates the effective diffusivity. The last term of Eq. (3) represents the source arisen from the catalytic reactions.

$$\rho_{mix} = \frac{p}{RT}\sum_{i=1}^{n} M_i M_{w,i} \quad (4)$$

$$\mu_{mix} = \sum_{i=1}^{n}\frac{M_i\mu_i}{\sum_{j=1}^{n} M_j\phi_{ij}} \quad (5)$$

$$\phi_{ij} = \sum_{i=1}^{n}\frac{\left[1+\left(\frac{\mu_i}{\mu_j}\right)^{1/2}\left(\frac{M_{w,j}}{M_{w,i}}\right)^{1/4}\right]^2}{\left[8\left(1+\frac{M_{w,i}}{M_{w,j}}\right)\right]^{1/2}} \quad (6)$$

Catalytic reaction equations：

In this simulating model, methanol SR as well as WGS catalytic reactions are merely considered as Eqs. (7) and (8) because the decomposition of methanol needs greater heat and carries out more slowly than SR of methanol (Sá et al., 2011).

$$CH_3OH + H_2O \xrightarrow{k_1} CO_2 + 3H_2 \quad \Delta H_{298} = 49.5\frac{kJ}{mol} \quad (7)$$

$$CO + H_2O \xleftrightarrow{k_2} CO_2 + H_2 \quad \Delta H_{298} = -41.2\frac{kJ}{mol} \quad (8)$$

In view of Arrhenius chemical model (Wan et al., 2016), the rates of above principle reactions are estimated from the next equations:

SR:
$$R_{SR} = k_1 c_{CH_3OH}^{0.6} c_{H_2O}^{0.4}\exp\left(-\frac{E_{a1}}{RT}\right) \quad (9)$$

Table I. Major parameters for this simulating modeling

Quantity	Value
Channel length, L (mm)	30.0
Inlet height, H_{in} (mm)	0.375~1.5
Outlet height, H_{out} (mm)	1.5
Channel width, W (mm)	1.5
Thickness of catalyst layer, e_p (mm)	0.5
Space velocity, SV (h^{-1})	4000
Inlet temperature, T_o (°C)	120
Heating temperature, T_{wall} (°C)	300
Operated pressure (atm)	1
Catalyst density (kg m^{-3}) (Lü et al., 2018)	1480
Thermal conductivity (W m^{-1} K^{-1}), K_S (Palma et al., 2018)	1.0
Permeability, κ (m^2) (Sá et al., 2011)	2.379×10^{-12}
Mass diffusion coefficient (m^2 s^{-1}) (Sá et al., 2011)	6.8×10^{-5}
Universal gas constant, R (J mol^{-1} K^{-1})	8.314
Porosity of catalyst, ε (Sá et al., 2011)	0.38
Tortuosity of catalyst, τ (Sá et al., 2011)	1.5
Activation energy of SR, E_{a1} (J mol^{-1}) (Palma et al., 2018)	7.0×10^{4}
Activation energy of WGS, E_{a2} (J mol^{-1}) (Palma et al., 2018)	1.0×10^{5}
Reaction constant of SR, k_1 (Palma et al., 2018)	8.0×10^{8}
Reaction constant of WGS, k_2 (Palma et al., 2018)	4.0×10^{8}

WGS:

$$R_{WGS} = k_2 c_{CO_3} c_{H_2} \exp\left(-\frac{E_{a2}}{RT}\right) - k_2 c_{CO} c_{H_2O} \exp\left(-\frac{E_{a2}}{RT}\right) \quad (10)$$

where k_1 and k_2 mean the reaction constants.

Energy equation：

$$(\rho_f C_P)(\vec{u} \bullet \nabla) T = K_{eff} \nabla^2 T + \varepsilon S_t \quad (11)$$

where C_P means the specific heat. K_{eff} indicates the effective thermal conductivity of the porous catalyst layer and is evaluated from $K_{eff} = \varepsilon K_f + (1-\varepsilon) K_S$. Moreover, the source term of Eq. (12) is originated from all catalytic reactions within the catalyst layer, and evaluated from

$$S_t = -\sum_{i=1}^{N} \left(\frac{h_i^0}{M_i} + \int_{T_{ref}}^{T} C_{P,i} dT \right) (R_i) \quad (12)$$

where h_i^0 means the enthalpy of the i-th species.

The boundary conditions of this simulating model are displayed in Fig. 1. On the inlet face, the temperature of uniform inlet flow is 120°C. On the outlet face, the atmospheric pressure is specified as the outlet condition. The no-slip condition is designated for all walls of a reactor. On the heated-wall surface, the temperature is controlled at 300°C. On the interface between reactor fuel channel and catalyst layer, all physical quantities are specified to be continuous. The space velocity (SV) is fixed at 4000 h^{-1}. The molar ratio of H_2O/ CH_3OH (S/C) is 0.9 at the inlet. The operated pressure is fixed at 1 atm within the reactor. The catalyst considered for reforming is $CuO\text{-}ZnO/Al_2O_3$. Moreover, other major parameters are cataloged in Table I.

NUMERICAL METHODS

This solving procedure is based on the SIMPLE-C algorithm (Van Doormaal and Raithby, 1984) linked with an improved conjugate gradient method (Kershaw, 1978; Van Der Vorst, 1992) for predicting the three-dimensional flow fields and all catalytic reactions about SR of methanol and WGS in the divergent fuel channel of a reactor. Under specifying 0.5 to be the under-relaxation factors of all principle equations, this computational program will be terminated while the residuals of major physical quantities are lower than 10^{-6}.

RESULTS AND DISCUSSION

The present study explored numerically the effect of divergent fuel channel on the reforming performance and non-isothermal reactant transport in a plate-shape reactor under methanol SR and WGS reactions. In addition, the yielded hydrogen from the reactor can be used to evaluate the net power output from a PEMFC. The main geometric parameter (H_{out}/H_{in}) of the fuel channel is specified as 4.0, 2.0, 1.33, and 1.0 with constant channel-outlet height and space velocity (SV). The present study performed the independent process for deciding the mesh density of computational domain. Three densities of structural hexahedron mesh system are tested and the related results are displayed in Fig. 2(a). The relative variances of major reforming performances between the two denser mesh densities for various heating temperatures are all smaller than 0.05 %. Accordingly, the second mesh density ($38\times80\times750$) is counseled for all reactor domains. In addition, as shown Fig.2(b), our previous study (Perng and Wu, 2019) had utilized the experimental data (Kim and Kwon, 2006) to certify that our solving program could be carried out for simulating the methanol SR and WGS catalytic reactions within a plate-shape reactor.

Figs. 3 and 4 demonstrate how the divergent effect of fuel channel acts on the species conveyance and the fluid velocity. Inside the catalyst layer (the region upon the dash-line), the reactants (CH_3OH and H_2O) lower progressively and the products (H_2 and CO) grow gradually downstream because the methanol SR and WGS catalytic reactions undergo inside the catalyst layer. A higher hydrogen contour signifies a greater hydrogen output while a lower methanol contour stands

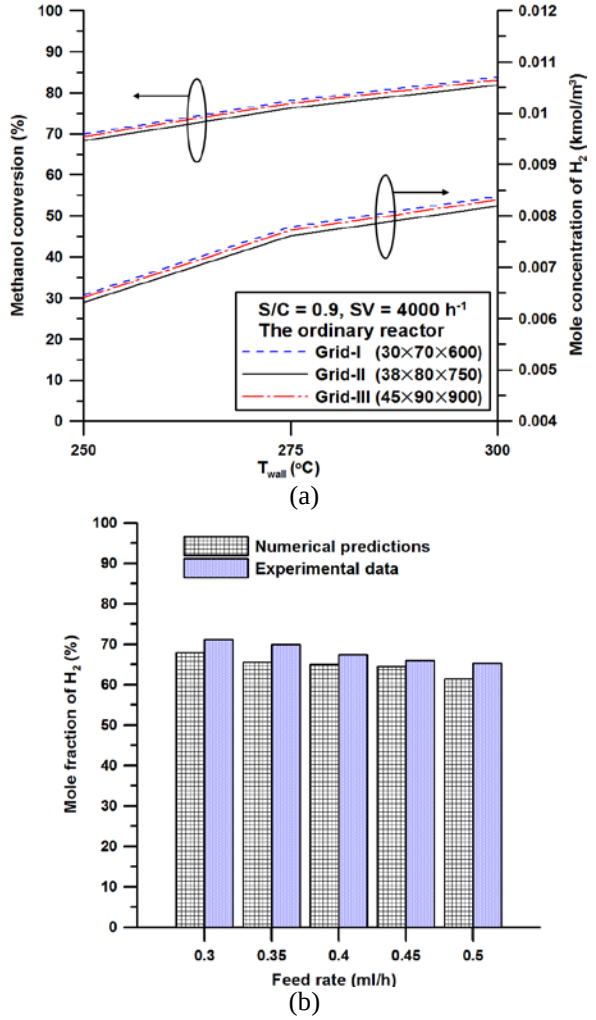

Fig. 2. (a) Mesh-independent tests of major reforming performances in an ordinary reactor; (b) Comparison of our numerical predictions (Perng and Wu, 2019) with the experimental data (Kim and Kwon, 2006).

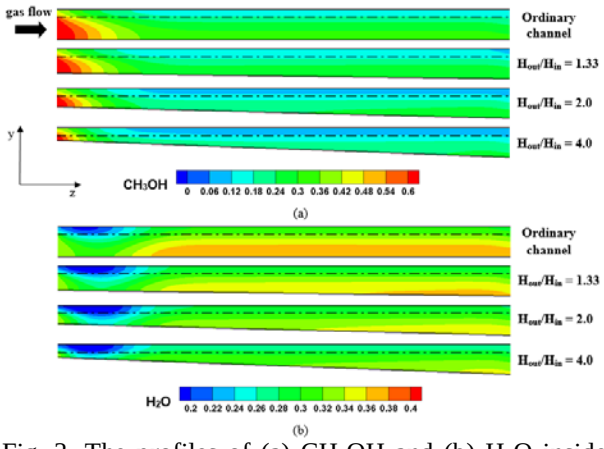

Fig. 3. The profiles of (a) CH_3OH and (b) H_2O inside various reactor channels on the y-z plane (x = 0.75 mm).

for better reforming of methanol. Consequently, the divergent fuel channels obviously bring about higher

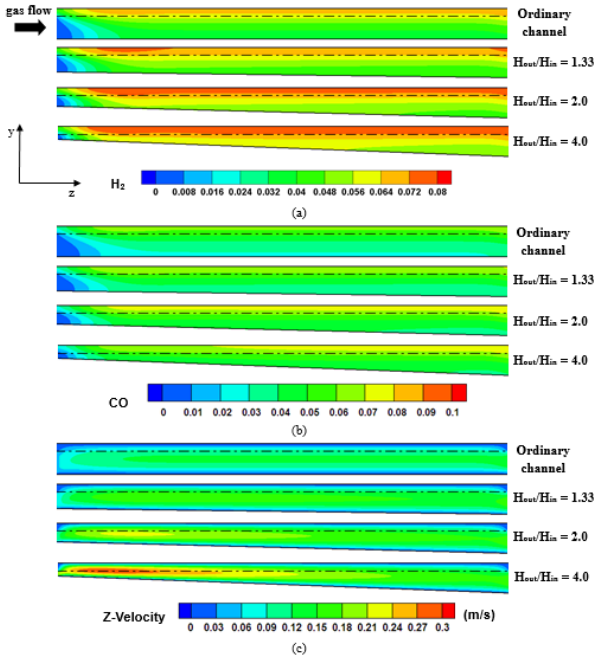

Fig. 4. The profiles of (a) H_2, (b) CO, and (c) z-velocity inside various reactor channels on the y-z plane (x = 0.75 mm).

product and lower reactant concentrations than an ordinary one. This is since a diffused channel retards the fluid velocity to longer the residence of reactants inside the catalyst layer and enhances the mixing of fuel within the reactor, and then obtains better catalytic reactions for advancing the hydrogen output. Additionally, larger variations of species and velocity profiles are caused from a greater H_{out}/H_{in}, which strengthens the diffusing effect to enhance the methanol SR and WGS reactions to advance the conversion of methanol, the hydrogen output, as well as carbon monoxide emission. The present work plotted Fig. 5 to value the application of divergent fuel channel to a reactor through related reforming performance. It obviously reveals that three divergent channels bring about higher methanol conversion and mass fraction of yielded hydrogen than the ordinary channel (H_{out}/H_{in} = 1.0), and higher H_{out}/H_{in} acquires more notable improvement in reforming performance. Compared with an ordinary channel, the divergent channel of H_{out}/H_{in} = 4.0 applied to the novel reactor results in the greatest enhancement of the hydrogen yield of 24.03 %. However, ameliorated hydrogen yield accompanies trade-off rise of CO emission from a reactor. These CO emissions are all less than 6 ppm from three novel reactors with various divergent channels. Accordingly, this still avoids the critical problem of PEMFC catalyst poisoning.

Because a larger pumping power of transporting the fluid is induced from a greater pressure drop within the reactor, the pressure drop needs to be properly studied in obtaining the net power of PEMFC system.

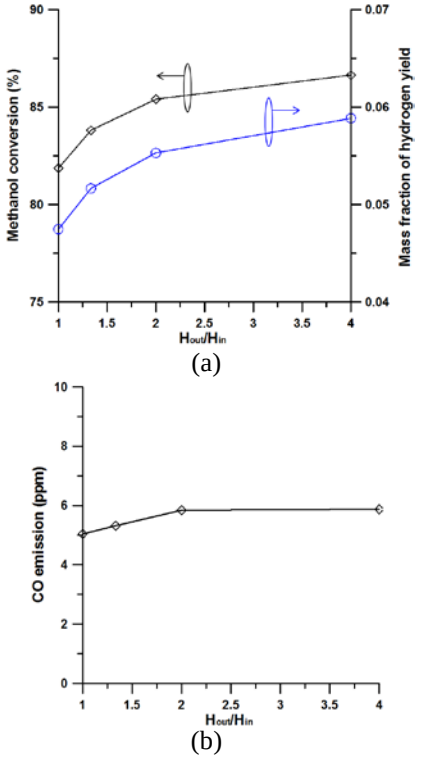

Fig. 5. Influence of H_{out}/H_{in} on (a) methanol conversion and hydrogen yield and (b) CO emission from a reactor.

The pressure drop is decided by the variation of pressure from the inlet to the outlet of reactor, expressed as $\Delta P = (P_{inlet} - P_{outlet})$, where P_{inlet} symbolizes the averaged pressure over the inlet surface, and P_{outlet} the averaged pressure over the outlet. Furthermore, the net power (W_{net}) is determined by Eqs. (13) and (14) under weighing the supplementary pumping power arising from this increased pressure drop.

$$W_{net} = W_{PEMFC} \times \eta_{H_2} \times \eta_{PEMFC} - W_P \quad (13)$$

$$W_P = \Delta P \times A_{ch} \times u_{in} \quad (14)$$

where W_{PEMFC}, the theoretic PEMFC power, is nearly 67 W while the fueled hydrogen is about 375 sccm (Nehe et al., 2015). Moreover, the efficiency of PEMFC (η_{PEMFC}) operation is commonly 60%, and the fair depletion of hydrogen (η_{H_2}) is close to 80% (Nehe et al., 2015). W_P signifies the supplemental pumping power of transporting the fluid through a reactor, A_{ch} the inlet area of reactor, and u_{in} the uniform velocity at the inlet. It can be found from Fig. 6 that higher H_{out}/H_{in} brought on greater pressure drop within the reactor, which requires larger pumping power of transporting the mixture inside the reactor. However, the greatest H_{out}/H_{in} (= 4.0) channel brings about the largest pressure drop, and the scheme of excessively expanded channel is thereby unsuitable for improving the net PEMFC power.

As a result, the greatest net PEMFC power is brought from the divergent channel of H_{out}/H_{in} = 4.0 under weighing the impact of pressure drop on the net PEMFC power output. As compared to an ordinary reactor, the novel reactor with this divergent fuel channel of H_{out}/H_{in} = 4.0 earns the greatest advance of 6.39 % in the net PEMFC power output.

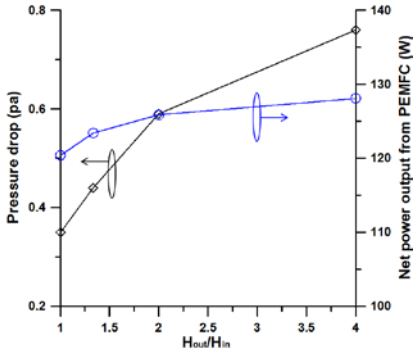

Fig. 6. Influence of H_{out}/H_{in} on the pressure drop within a reactor and the net PEMFC power output.

CONCLUSION

This numerical study carried out the 3D simulations to estimate the effect of divergent fuel channel on the reforming performance and non-isothermal reactant transport in a plate-shape reactor under methanol SR and WGS reactions. In addition, the yielded hydrogen would be utilized to evaluate the net PEMFC power output. The major conclusions excerpted from the above results are described as follows. The divergent channel brings a reactor better reforming performance and a PEM cell greater net power output than the ordinary channel. This is since a diffused channel retards the fluid velocity to longer the residence of reactants inside the catalyst layer and enhances the mixing of fuel within the reactor, and then obtains better catalytic reactions for advancing the hydrogen output. Moreover, a greater H_{out}/H_{in} divergent channel strengthens the diffusing effect to improve the methanol SR and WGS reactions, and thereby the divergent channel of H_{out}/H_{in} = 4.0 brings a reactor greater hydrogen yield than other fuel channels. As compared to the ordinary channel, the above divergent fuel channel brings about the most notable improvement in the hydrogen production of 24.03 %. Furthermore, this divergent channel produces a PEM cell the greatest advance of 6.39 % in the net power output under implementing the pressure drop through the reactor channel.

ACKNOWLEDGEMENTS

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漸擴燃料流道對平板型甲醇蒸汽重組器性能之效應研究

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摘要

本數值研究係利用三維模擬，評估漸擴燃料流道對平板型甲醇蒸汽重組器之重組性能與熱質傳遞現

象之影響。此外，根據甲醇重組器之氫氣產量及燃料流道內之壓力降，計算質子交換膜燃料電池之淨輸出功率。模擬結果顯示，採用漸擴燃料流道之甲醇重組器明顯可以提升氫氣產量，而且也能增加燃料電池之淨功率輸出。經與普通燃料流道之比較，採用流道出入口高度比4.0(H_{out}/H_{in} = 4.0)之漸擴燃料流道的新式甲醇重組器可以獲得最大的產氫提升率24.03 %。在考量甲醇重組器燃料流道內之壓力降，與普通燃料流道比較，採用流道出入口高度比4.0(H_{out}/H_{in} = 4.0)之漸擴燃料流道的新式甲醇重組器可以獲得最大的燃料電池淨輸出功率之增加率6.39 %。