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Numerical Study of Plate-Shape Methanol Steaming Reformer with a Convergent Fuel Channel

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Keywords：methanol steaming reformer, convergent fuel channel, hydrogen production.

ABSTRACT

This study performed three-dimensional numerical simulations to observe the effect of convergent fuel channel on the non-isothermal reactant transport and reforming performance in a plate-shape reactor under methanol steam reforming. Furthermore, the hydrogen production was utilized to estimate the net power output from a PEMFC. The results reveal that the application of convergent fuel channel to a plate-shape reactor evidently improves the hydrogen yield from reformer and estimated net power output from PEMFC. Compared with a common reactor, the novel reactor with the convergent fuel channel of H_{out}/H_{in} = 0.25 produces the greatest improvement in the hydrogen yield of 34.85 %. However, the novel reactor with the convergent fuel channel of H_{out}/H_{in} = 0.5 earns the maximum rise of 8.84 % for the estimated net power output form a PEMFC under considering the pressure drop through the channel.

INTRODUCTION

A PEMFC power source possesses the benefits of low-operating temperature, zero-carbon emission, and high-energy efficiency using hydrogen and oxygen as the fuel (Chung et al., 2007; Wu, 2016). About the hydrogen source, methanol has a significant advantage of greater hydrogen over carbon ratio converted into hydrogen through reforming processes under relatively low-operating temperature (Ouyang et al., 2017). A PEMFC stack integrated with a miniature metanol steam reformer (MSR) is generally regarded as an important portable power source (Zhang et al., 2018). Consequently, optimizing the miniature MSR is important to improve the hydrogen production from MSR and augment the net power output from PEMFC.

As compared to the packed-bed cylindrical reactor, the wall-coated plate-shape reactor has the advantages of greater ratio of heat transfer surface area to reactor volume, more uniform flow, and lower pressure drop (Ke et al., 2019; Dolanc et al., 2014). Accordingly, the plate-shape reactor is considered to be designed for enhancing the heat transfer from the heat source to improve the catalytic reaction. Numerous studies have addressed the application of convergent channel to the reactor for better chemical reaction in the catalyst. Chowdhury and Timurkutluk (2018) explored numerically the transport phenomena in the convergent and divergent serpentine channel of PEMFC. Their results indicated that the convergent channel leads to better reactant transport and higher current density. Perng and Wu (2011) investigated numerically the PEMFC performance enhancement through installing a baffle plate in a tapered flow channel. Their results displayed that the tapered flow channel facilitates better convection heat transfer and higher cell performance. From the results of the cited studies, it is clear that the convergent channel can facilitate better catalytic reaction than the common straight-channel because of better heat transfer performance.

For lowering the experimental cost, the scholars ordinarily performed numerical simulations to predict the catalytic reaction and thermal flow characteristics inside the reactor under fuel reforming. Ji et al. (2018) developed a CFD model to predict the chemical reaction process of methane steam reforming in a sorption enhanced membrane reactor. Perng et al. (2019) investigated numerically the effects of transverse ribs bottomed in the reactor channels on the hydrogen yield and estimated PEMFC net power. Jang et al. (2010) made use of Fluent software to explore how the geometric and the operating

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conditions influenced the performance of a micro-methanol steam reformer. Jiwanuruk et al. (2016) used COMSOL software to perform 3D simulations of a micro membrane reactor with methane reforming reaction for exploring the influences of various reactor arrangements on the conversion efficiency. The above cited numerical studies revealed that the numerical simulations could predict precisely the thermal-flow characteristics and the catalytic reactions in the reactor. However, they have never investigated how a convergent fuel channel improved the hydrogen yield and avoided a greater pressure drop to obtain a better estimated PEMFC net power.

Accordingly, the objective of this paper is to numerically examine how convergent fuel channel affects the thermal-flow characteristics, hydrogen yield, CO emission, and the estimated PEMFC net electric-power output within various reactors under methanol steam reforming.

CFD SIMULATION MODEL

This 3D simulation model, which consists of continuity, momentum conservation, energy conservation, and specie conservation equations, is used to study the thermal-flow characteristics for various fuel channels of plate-shape reactor under methanol steam reforming. Fig. 1 depicts that the plate-shape reactor is invented through a common plate-shape reactor by utilizing a convergent channel. The principal geometrical parameter is the ratio of outlet area and inlet area (H_{out}/H_{in}). This ratio is considered as 0.25, 0.5, 0.75, and 1.0. H_{out}/H_{in} = 1.0 indicates a common channel. The catalyst layer is covered above the fuel channel, and comprises fixed catalyst volume in the reactor. The CH_3OH mixed with H_2O is fed into the reactor at z = 0, and the positive z represents the streamwise direction along the fuel channel.

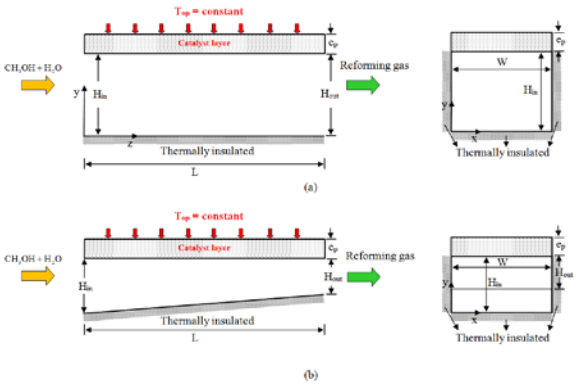


Fig. 1. Computational domains of (a) common reactor fuel channel and (b) convergent fuel channel of reactor.

The assumptions of this simulation model for the methanol reforming reactor are given as follows.

(1) The reacting gas is ideal.
(2) The thermo-flow field is laminar, incompressible, and steady.
(3) The catalyst is homogeneous and isotropic.
(4) All chemical reactions undergo merely within the catalyst layer.
(5) As compared to the convection, the radiation and conduction could be negligible at the gas state.

The governing equations of this 3D model for the methanol reforming reactor are expressed by the following equations.

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

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

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

where κ and ε denote the permeability and porosity of catalyst layer. β is the inertial loss coefficient within the catalyst layer. ρ_{mix} and μ_{mix} indicate the density and viscosity of gas mixture. c_i is the molar concentration of specie i. D_{eff} is the effective diffusivity. The final term of Equation (3) stands for the source resulted from the catalytic reactions within the catalyst layer.

Catalytic reaction equations：

In this model, the catalytic reactions of steam reforming (SR) as well as reverse water-gas-shift (WGS) are merely taken in Equations (4) and (5) because the decomposition reaction needs higher heat and undergoes more slowly than SR reaction (Sá et al., 2011).

$$CH_3OH + H_2O \xrightarrow{k_1} CO_2 + 3H_2 \quad \Delta H_{298} = 49.5 \frac{kJ}{mol} \quad (4)$$

$$CO + H_2O \xrightarrow{k_2} CO_2 + H_2 \quad \Delta H_{298} = -41.2 \frac{kJ}{mol} \quad (5)$$

$$H_2 + CO_2 \xrightarrow{k_3} CO + H_2O \quad \Delta H_{298} = 41.2 \frac{kJ}{mol} \quad (6)$$

On the basis of Arrhenius model (Wan et al., 2016), the rates of above catalytic reactions are evaluated in the following equations:

SR:
$$R_{SR} = k_1 c_{CH_3OH}^{0.6} c_{H_2O}^{0.4} \exp\left(-\frac{E_a}{RT}\right) \quad (7)$$

WGS:
$$R_{WGS} = k_2 c_{CO_3} c_{H_2} \exp\left(-\frac{E_a}{RT}\right) - k_3 c_{CO} c_{H_2O} \exp\left(-\frac{E_a}{RT}\right) \quad (8)$$

where k_1, k_2, and k_3 are the constants of catalytic reaction rate.

Energy equation：

$$(\rho_f C_P)(\vec{u} \bullet \nabla)T = K_{eff}\nabla^2 T + \varepsilon S_t \quad (9)$$

where C_P is the specific heat of mixture. K_{eff} is the effective thermal conductivity of solid state coupled with that of fluid state for the porous catalyst and calculated from $K_{eff} = \varepsilon K_f + (1-\varepsilon)K_S$. Furthermore, the source term of energy equation, S_t, is originated from the catalytic reaction inside the catalyst layer, so it can be calculated by

$$S_t = -\sum_{i=1}^{N}\left(\frac{h_i^0}{M_i} + \int_{T_{ref}}^{T} C_{P,i}dT\right)(R_i) \quad (10)$$

where h_i^0 symbolizes the enthalpy of specie i.

The boundary conditions of simulation model are illustrated in Fig. 1. At the inlet, the temperature of uniform fuel flow is 120°C. At the outlet, the atmospheric pressure is designated as the boundary condition. The no-slip condition is considered for the wall of reactor. On the heated-surface, the temperature is operated and fixed at 300°C. On the interface between catalyst layer and reactor channel, all physical quantities are continuous. The space velocity (SV) is 4000 h^{-1}. The molar ratio of H_2O/ CH_3OH of fueled mixture (S/C) is 0.9. The operating pressure is specified at 1 atm in the reactor. Moreover, other principal parameters are summarized as shown in Table I.

NUMERICAL METHODS

This numerical study applies the SIMPLE-C algorithm (Van Doormaal and Raithby, 1984) integrated with a refined conjugate gradient method (Kershaw, 1978; Van Der Vorst, 1992) as the solver for predicting the 3D laminar thermal flow and the catalytic reactions of methanol steam reforming in the convergent fuel channel of reactor. Under adopting 0.5 as the under-relaxation factors of all equations, this CFD code will be stopped when the residuals of all physical quantities are less than 10^{-6}.

RESULTS AND DISCUSSION

This numerical study explored the effect of convergent fuel channel on the non-isothermal reactant transport and reforming performance in a plate-shape reactor under methanol steam reforming. In addition, the hydrogen yielded from the reactor is used to estimate the net power output from a PEMFC. The ratio H_{out}/H_{in} of the convergent fuel channel is considered as 0.25, 0.5, 0.75, and 1.0. The grid-independence procedure was established for this CFD study. The refinement of three structural hexahedron grid systems is indicated in Fig. 2(a). The relative variation of evaluated reforming performance between the two denser grid systems for various operating temperatures is smaller than 0.05 %. Accordingly, the second grid system ($38\times80\times750$) is taken for all simulations. In addition, the experimental data (Kim and Kwon, 2006) were utilized to verify that this solving code could be performed to simulate the catalytic reactions inside a plate-shape reactor under methanol steam reforming.

Table I. Principal parameters for this modeling

Quantity	Value
Channel length, L (mm)	30.0
Inlet height, H_{in} (mm)	1.5
Outlet height, H_{out} (mm)	0.375~1.5
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
Operating temperature, T_{op} (°C)	300
Operating 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 reverse WGS, E_{a2} (J mol^{-1}) (Palma et al., 2018)	1.0×10^{5}
Activation energy of backward WGS, E_{a3} (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 reverse WGS, k_2 (Palma et al., 2018)	4.0×10^{8}
Reaction constant of backward WGS, k_3 (Palma et al., 2018)	4.0×10^{8}

Figs. 3 and 4 exhibit how the convergent fuel channel affects the species transportation and the streamwise velocity profile in a reactor. Within the catalyst layer (the area above the dash-line), the reactants (CH_3OH and H_2O) lessen gradually and the products (H_2 and CO) rise gradually downstream, owing to the reactants transformed into the products via the chemical catalytic reactions. A higher hydrogen concentration represents a greater hydrogen yield while a lower methanol concentration means a

superior

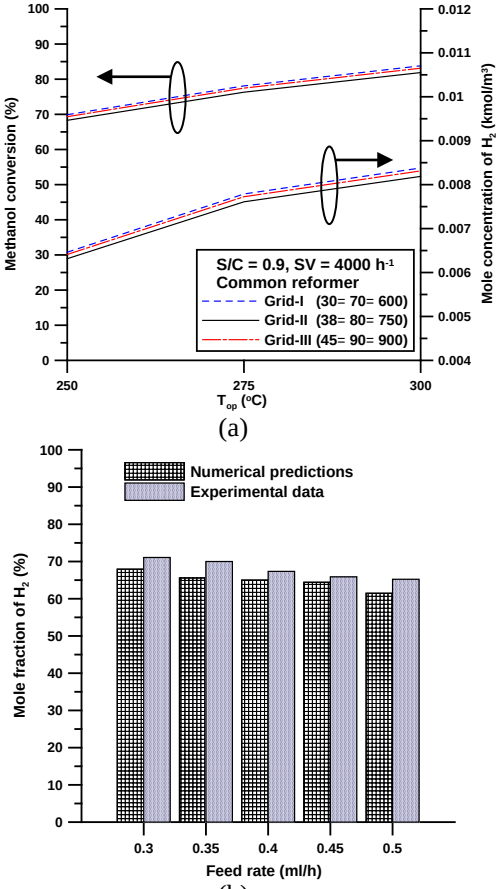


Fig. 2. (a) Grid-independence procedure of reforming performance in a common reactor; (b) Comparison of this numerical predictions with the experimental data (Kim and Kwon, 2006).

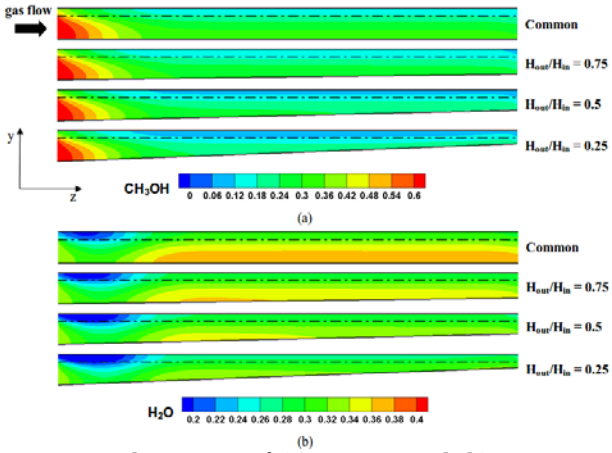


Fig. 3. Local contours of (a) CH_3OH and (b) H_2O in the reactor channels with various H_{out}/H_{in} on the y-z plane (x = 0.75 mm).

methanol conversion. The convergent fuel channels result in lower reactant and higher product profiles than a common one since contracting channel squeezes products effectively from the catalyst layer to avoid obstructing reactants. Moreover, the convergent channel can compress and accelerate the reacting fluid to feed more reactant into the catalyst layer for increasing the hydrogen yield.

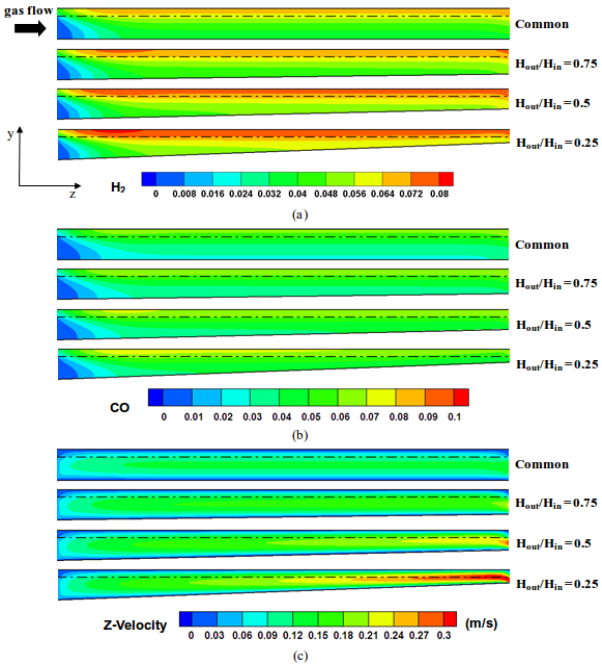


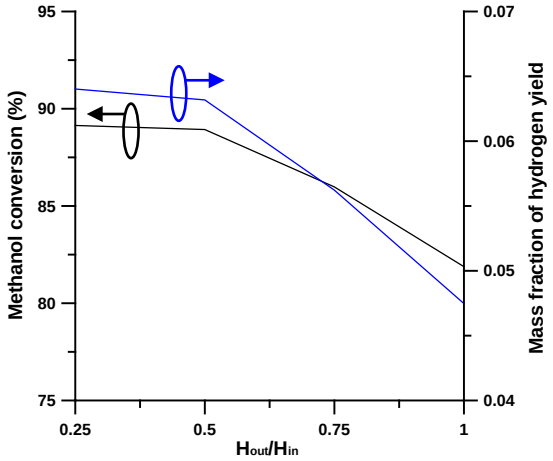
Fig. 4. Local contours of (a) H_2, (b) CO, and (c) z-velocity in the reactor channels with various H_{out}/H_{in} on the y-z plane (x = 0.75 mm).

Accordingly, greater variations of reactant, product, and velocity distributions are induced by a lower H_{out}/H_{in}, which enhances the catalytic reforming reactions to facilitate the methanol conversion, the hydrogen yield, and carbon monoxide emission. This study plotted Fig. 5 to estimate the effect of convergent fuel channel applied to a reactor on the reforming performance. It can be found that three convergent channels induce higher methanol conversion and mass fraction of hydrogen yield than the common channel (H_{out}/H_{in} = 1), and a lower H_{out}/H_{in} acquires the greater enhancement of reforming performance. As compared to a common reactor, the novel reactor with the convergent channel of H_{out}/H_{in} = 0.25 produces the greatest improvement in the hydrogen yield of 34.85 %.

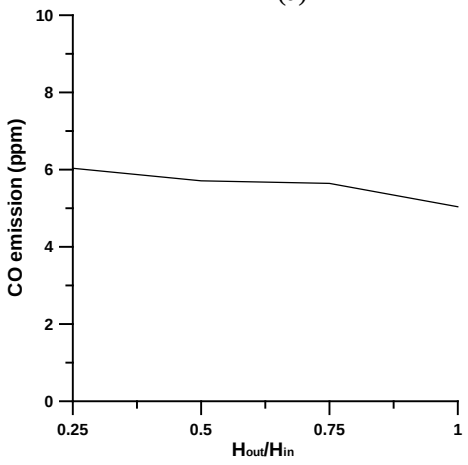
However, an improved reforming performance accompanies a trade-off rise of carbon monoxide emission from the reactor. These carbon monoxide emissions are lower than 6 ppm from the novel reactors with the convergent channel. Consequently, this could avoid the serious poisoning problem of PEMFC catalyst.

A larger pressure drop through the fuel channel results in a higher pumping power to deliver the reacting gas, so the pressure drop must be properly considered in estimating the net power output from a PEMFC system. The pressure drop is determined through the difference of pressure between the inlet and the outlet of channel, expressed by ,

$\Delta P = (P_{inlet} - P_{outlet})$ where P_{inlet} represents the averaged pressure on the inlet surface, and P_{outlet} the averaged pressure on the outlet surface. In addition, the net power (W_{net}) is estimated from Equations. (11) and (12) under considering the supplementary pumping power brought from the pressure drop.



(a)



(b)

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

$$W_{net} = W_{PEMFC} \times \eta_{H_2} \times \eta_{PEMFC} - W_P \quad (11)$$

$$W_P = \Delta P \times A_{ch} \times u_{in} \quad (12)$$

where W_{PEMFC}, the theoretical PEMFC power, is almost 67 W as the hydrogen yield is nearly 375 sccm (Nehe et al., 2015). Furthermore, the working efficiency of PEMFC (η_{PEMFC}) is normally 60%, and the acceptable consumption of hydrogen (η_{H_2}) is nearly 80% (Nehe et al., 2015). W_P represents the supplemental pumping power to deliver the reacting gas through the channel, A_{ch} the inlet area of channel, and u_{in} the uniform velocity on the inlet. It can be observed from Fig. 6 that smaller H_{out}/H_{in} results in larger pressure drop through reactor channel, which requires a greater pumping power to deliver the reacting gas through the reactor. However, the smallest H_{out}/H_{in} (= 0.25) channel results in the largest pressure drop, and thereby the utilization of excessively contracted channel is unsuitable for the practical improvement of PEMFC performance.

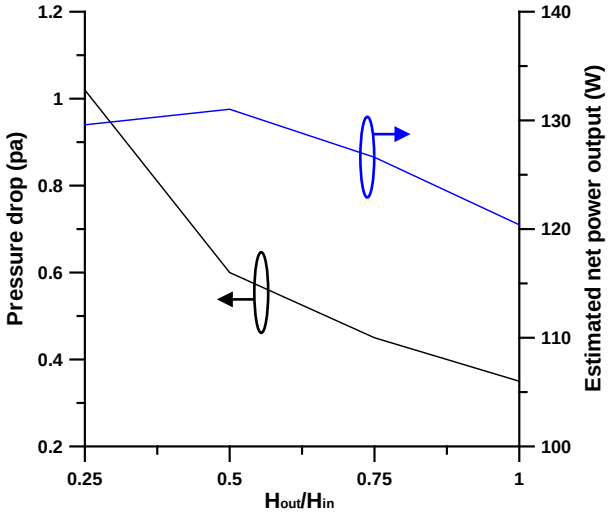


Fig. 6. Effect of H_{out}/H_{in} on the pressure drop through a reactor and the estimated net power output from a PEMFC.

Consequently, the greatest estimated power is generated from the convergent channel of H_{out}/H_{in} = 0.5 under weighting the influence of pressure drop on evaluating the net power output from PEMFC. As compared to the common reactor, the novel reactor with this convergent fuel channel of H_{out}/H_{in} = 0.5 earns the maximum rise of 8.84 % for the net power output from PEMFC.

CONCLUSION

This paper carried out the 3D numerical simulations to evaluate how the convergent fuel channel affects the non-isothermal reactant transport and reforming performance in a plate-shape reactor under methanol steam reforming. In addition, the hydrogen yield was utilized to estimate the net power output from a PEMFC. The principal conclusions summing up the above discussions are as follows. The novel reactor with the convergent channel creates the better reactor performance and greater estimated net power output from PEMFC than the common reactor. This is since the contracting channel squeezes products effectively from the catalyst layer to avoid obstructing reactants, and it can compresses and accelerates the reacting fluid to feed more reactant into the catalyst layer for increasing the hydrogen yield. Moreover, a lower H_{out}/H_{in} convergent channel produces the stronger squeezing and compressing effects on the reacting fluid for improving the reactor performance, and thereby the

novel reactor with the convergent channel of H_{out}/H_{in} = 0.25 creates greater hydrogen yield than other reactors. As compared to the common reactor, applying the above convergent fuel channel to a reactor acquires the maximum increase in the hydrogen yield by 34.85 %. Furthermore, under weighting the pressure drop through the reactor channel, the reactor with the convergent channel of H_{out}/H_{in} = 0.5 brings about the maximum rise of 8.84 % for the estimated net power output form a PEMFC.

ACKNOWLEDGEMENTS

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應用漸縮燃料流道於平板型甲醇蒸汽重組器之數值研究

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摘 要

本研究利用三維數值模擬，探討漸縮燃料流道對平板型甲醇蒸汽重組器之熱質傳遞現象與重組性能之影響。進一步，根據重組器之氫氣產量，估算質子交換膜燃料電池之實質淨功率輸出。模擬結果顯示，應用漸縮燃料流道之重組器明顯可以改善氫氣產量，而且也能提升燃料電池之實質淨功率輸出。經與一般平板型重組器之比較，採用出入口面積比0.25(H_{out}/H_{in} = 0.25)之漸縮燃料流道的新式重組器可以獲得最大的產氫改善率34.85 %。在考量重組器燃料流道內之壓力降下，與一般平板型重組器比較，採用出入口面積比0.5(H_{out}/H_{in} = 0.5)之漸縮燃料流道的新式重組器可以獲得最大的燃料電池估算淨功率輸出之提升率8.84 %。