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Article

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Performance analysis of direct steam reforming of methane in SOFC with SDC-based electrolyte

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Abstract

This work aims to study on the performance of the SDC-based SOFC with a direct internal reforming of methane at lower temperature in the range of 600–750 °C. The model of SDC-based SOFC with direct internal reforming mode is simulated and validated. The predicted results are in a good agreement with experimental data. The simulated results indicate that the molar flow rate of the hydrogen produced from the methane steam reforming reaction is higher as an increasing temperature. Moreover, the cathode activation overpotential is obviously reduced. Thus, the increase of temperature from 600 to 700 °C can enhance the average power density of SOFC from 0.19 to 0.42 A/m².

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Keywords: Solid oxide fuel cell; Low temperature; Direct reforming; Steam reforming of methane

1. Introduction

Among the various types of fuel cell, a solid oxide fuel cell (SOFC) is the most attractive fuel cell technology that can be used in a wide range of commercial applications. The high-temperature operation of SOFC (800–1000 °C) has many advantages. A high-temperature waste heat from the SOFC can be recovered for using in other heat-demanding units in the SOFC system. However, high-temperature SOFC requires the highly durable materials for SOFC fabrication. In general, materials used for high-temperature SOFC are all ceramic and high-chrome alloys as interconnect which are rather expensive. Other significant problems of the high-temperature SOFC are the slow startup or shutdown operation and the high thermal gradient [1]. Some research groups have concentrated

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on alternative electrolyte materials with high ionic conductivity at lower temperatures instead of yttria-stabilized zirconia (YSZ). Samarium doped ceria (SDC) is promising electrolyte material for lower temperature of SOFC (500–800 °C) because the oxide-ion conductivity of SDC is higher than that of YSZ [2,3]. The suitable materials for electrode of low-temperature SOFC have been considered by some studies. Hong et al. [4] investigated the performance of Nickel-samarium doped ceria (Ni-SDC) as anode for low-temperature SOFC. The results showed that the use of Ni-SDC anode having a high porosity resulted in a low resistance within the cell. Ni can tolerate CO and use impure hydrogen, resulting in its fuel flexibility. In addition, Ni is commonly used as catalyst for steam reforming of hydrocarbon fuel to produce hydrogen-rich gas. Thus, hydrocarbon fuels are possible to be internally reformed into a hydrogen-rich gas (synthesis gas) within SOFC (referred to as an internal reforming SOFC or IR-SOFC) because the operating temperature of SOFC is in the same range as that of the reforming reactions. IR-SOFC offers not only the simplest and most cost-effective design for an SOFC system due to the elimination of pre-reformer, but also it is the high thermal efficient operation resulting from the utilization of heat released from an exothermic electrochemical oxidation for endothermic steam reforming [5,6]. Numerous researchers studied the direct reforming on Ni/YSZ. Nowadays, there are limited studies on the design of low-temperature SOFC with SDC-based electrolyte under the direct internal reforming of methane. To improve the performance of low-temperature SOFC with SDC-based electrolyte, an in-depth understanding of the relations between performance and relevant microstructure parameters is acquired. Thus, the model of SDC-based SOFC with direct internal reforming is simulated to analyze and improve its performance at low temperature in this work.

2. Modeling of direct reforming SOFC

SOFC operated under direct reforming mode shows in Fig. 1. In the configuration of SOFC with SDC-based electrolyte, materials of electrolyte, anode, and cathode are samarium-doped ceria (SDC), nickel-samarium-doped ceria (Ni/SDC), and lanthanum strontium cobalt ferrite (LSCF), respectively. When feeding the methane as fuel and steam as reagent, methane can be converted to synthesis gas consisting of CH₄, H₂, CO, CO₂ and H₂O through the methane steam reforming reaction and the water gas shift reaction as follows:

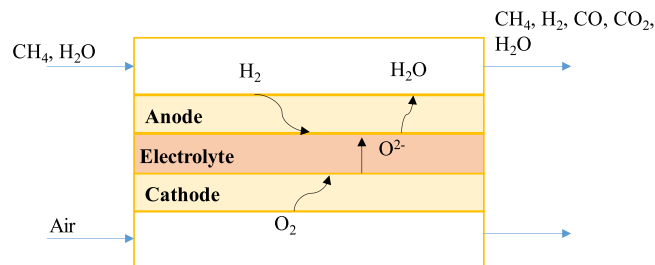
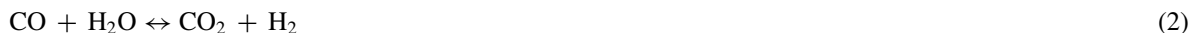


Fig. 1. Working principle SOFC with SDC-based electrolyte under direct reforming of methane.

Steam reforming:



Water gas shift:



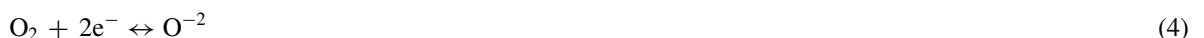
Hydrogen produced from steam reforming and water-gas shift reactions is oxidized at the anode side.

Oxidation reaction:



In SOFC operation, air is continuously fed into the air channel as an oxidant. Oxygen in air is reduced to oxygen ion as shown in Eq. (4).

Reduction reaction:



Overall electrochemical reaction of cell is as follows:



The SOFC model was simulated based on the mass balance and electrochemical equations.

2.1. Mass balance

The configuration of the SOFC is a planar design. The direct reforming SOFC was considered as one-dimensional model. The mass balance in the fuel and air channels can be shown in terms of the component concentration and can be described as follows:

Fuel channel:

$$\frac{dC_{i,f}}{dx} = \frac{1}{u_f} \sum_{k \in \{(i),(ii),(iii)\}} v_{i,k} R_k \frac{1}{h_f} \quad (6)$$

Boundary condition for fuel channel:

$$C_{i,f}|_{x=0} = C_{i,f}^0 \quad (7)$$

Air channel:

$$\frac{dC_{i,a}}{dx} = \frac{1}{u_a} v_{i,(iii)} R_{(iii)} \frac{1}{h_a} \quad (8)$$

Boundary condition for air channel:

$$C_{i,a}|_{x=0} = C_{i,a}^0 \quad (9)$$

where $C_{i,f}$ and $C_{i,a}$ are the concentrations of species i in the fuel and air channels, respectively; u_f and u_a are the inlet velocities of fuel and air, respectively; h_f and h_a are the heights of the fuel and air channels, respectively; and R_k is the reaction rates. All reaction rates in SOFC are referred from Brus et al. [7].

2.2. Electrochemical model

SOFC can produce power which can be calculated from Eq. (10).

$$P = iVA \quad (10)$$

where i is the current density; A is the active area; and V is operating voltage of cell.

The operating voltage of cell can be calculated from the open-circuit voltage (E^{OCV}) deducted by voltage losses within the internal cell, which is described by Eq. (11). There are three main overpotential losses namely ohmic losses (η_{ohm}), activation overpotentials (η_{ac}), and concentration overpotentials (η_{con}).

$$V = E^{\text{OCV}} - (\eta_{\text{ohm}} + \eta_{\text{ac}} + \eta_{\text{con}}) \quad (11)$$

The open-circuit voltage can be expressed by the Nernst equation:

$$E^{\text{OCV}} = E^0 - \frac{RT}{2F} \ln\left(\frac{P_{\text{H}_2\text{O}}}{P_{\text{H}_2} P_{\text{O}_2}^{0.5}}\right) \quad (12)$$

Ohmic loss can be expressed by Ohm's law.

$$\eta_{\text{ohm}} = iR_{\text{ohm}} \quad (13)$$

The sluggishness of reaction at the electrode–electrolyte interfaces is the cause of the activation overpotentials which are described by the Butler–Volmer equation.

$$i = i_{0,\text{anode}} \left[\frac{P_{\text{H}_2,\text{TPB}}}{P_{\text{H}_2,\text{f}}} \exp\left(\frac{\alpha n F}{RT} \eta_{\text{ac},\text{anode}}\right) - \frac{P_{\text{H}_2\text{O},\text{TPB}}}{P_{\text{H}_2\text{O},\text{f}}} \exp\left(-\frac{(1-\alpha)n F}{RT} \eta_{\text{ac},\text{anode}}\right) \right] \quad (14)$$

$$i = i_{0,\text{cathode}} \left[\exp\left(\frac{\alpha n F}{RT} \eta_{\text{act},\text{cathode}}\right) - \exp\left(-\frac{(1-\alpha)n F}{RT} \eta_{\text{act},\text{cathode}}\right) \right] \quad (15)$$

where n is the number of electrons exchanged in reaction step; α is the apparent transfer coefficient; $i_{0,\text{anode}}$ and $i_{0,\text{cathode}}$ are the exchange current densities of the anode and cathode; and $p_{i,\text{TPB}}$ is the partial pressure of species i at triple phase boundary that can be calculated from gas transport model in porous media.

The exchange current density of the electrode ($i_{0,\text{electrode}}$) is given by the following

$$i_{0,\text{electrode}} = \frac{RT}{nF} k_{\text{electrode}} \exp\left(-\frac{E_{\text{electrode}}}{RT}\right) \quad (16)$$

where $E_{\text{electrode}}$ is the activation energy of electrode exchange current density and $k_{\text{electrode}}$ represents the pre-exponential factors of the exchange current densities. Due to the reduction of reactant concentration at the electrode, the mass transport of species i decreases, and leading to concentration overpotentials.

$$\eta_{\text{con}} = \frac{RT}{2F} \ln\left(\frac{p_{\text{H}_2\text{O,TPB}} p_{\text{H}_2,\text{f}}}{p_{\text{H}_2\text{O,f}} p_{\text{H}_2,\text{TPB}}}\right) + \frac{RT}{4F} \ln\left(\frac{p_{\text{O}_2,\text{a}}}{p_{\text{O}_2,\text{TPB}}}\right) \quad (17)$$

The electrochemical parameters of SOFC with SDC-based electrolyte are referred from literature [8,9].

3. Results and discussion

The characteristic of direct reforming SOFC with SDC-base electrolyte was studied in this work. The predicted results of direct reforming SOFC model were validated with the experimental results of Brus et al. [7]. The specific surface area for model validation is 4.4 m²/g of catalyst. Table 1 shows the comparison of product compositions from methane steam reforming on Ni/SDC between the simulation and experimental data at various temperatures. The SOFC was operated at the pressure of 1 bar. From Table 2, the product compositions from predicted results are closed to the experimental data. Thus, the results from the model simulation are acceptable. For the electrochemical characteristics of SDC-based SOFC was validated in our previous work [9].

Table 1. Comparison of experimental and simulation data.

Temperature (°C)	Results	Mole fraction (-)			
		CH ₄	CO ₂	H ₂	CO
650	Experiment	0.53	0.13	0.3	0.02
	Simulation	0.55	0.13	0.31	0.01
	Error (%)	3.77	0	3.33	50.00
724	Experiment	0.28	0.14	0.52	0.06
	Simulation	0.28	0.15	0.54	0.04
	Error (%)	0	7.14	3.85	33.33

Table 2. Cell configuration.

Parameters	Value	Parameters	Value
Cell length (cm)	0.4	Anode thickness (μm)	500
Fuel channel height (m)	0.001	Electrolyte thickness (μm)	40
Air channel height (m)	0.001	Cathode thickness (μm)	40

The performance of direct reforming of SOFC was studied at low-temperature operation in the range of 600–750 °C. The configuration of SOFC is shown in Table 2. The SOFC operates at inlet methane to carbon ratio of 1.8. Fig. 2 shows the profile of hydrogen molar flow rate along the cell length at fuel channel with various temperatures. From Fig. 2, the molar flow rate of hydrogen considerably increases at the cell inlet and increases until maximum point. Then, the molar flow rate of hydrogen continuously decreases. This can explain that the rate of methane steam reforming reaction is high; hence, hydrogen is substantially produced at the cell inlet. The molar flow rate of hydrogen decreases because it is consumed in the hydrogen oxidation reaction. At a lower operating temperature, the rate of produced hydrogen is slower. This is because the reduction of operating temperature has effect on the decrease in the reaction rate of methane steam reforming. Moreover, the steam reforming reaction is highly endothermic reaction. Thus, the amount of the produced hydrogen reduces at lower temperature.

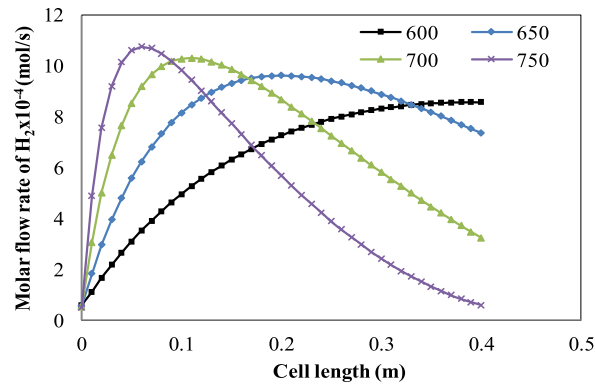


Fig. 2. Effect of operating temperature on molar flow rate of hydrogen along the cell length.

Fig. 3 presents the influence of operating temperature on the average power density of direct reforming SOFC. It can be seen that the power density would be raised as the operating temperature increased. The increment of power density results from the increment of the produced hydrogen at a higher temperature. Additionally, the operating temperature has a direct effect on the electrochemical performance. The main internal voltage loss of SOFC with SDC-based electrolyte is the activation overpotential. Fig. 4 shows the average anode and cathode activation overpotentials at different temperatures. The anode activation overpotential slightly decreases as increasing the cell temperature. Meanwhile, the increase of the cell temperature results in the obvious reduction of cathode activation overpotential. This leads to increase of the cell power density. The average power density enhances from 0.19 A/m² to 0.42 A/m² when an increasing temperature from 600 to 700 °C. The increment of power density reduces when an increasing temperature above 700 °C.

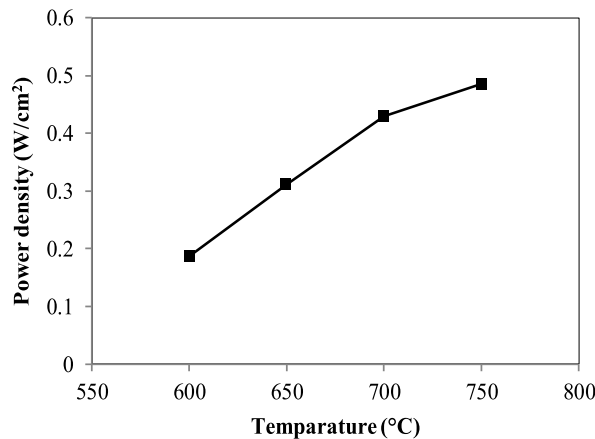


Fig. 3. Effect of operating temperature on average cell power density.

4. Conclusion

The performance of direct reforming SOFC with SDC-based electrolyte at low temperature in the range of 600–750 °C is studied in this work. The model of direct methane SOFC with SDC-based electrolyte is validated with the experimental data. The model predictions are in a good agreement with experimental data. The validated model is used to study the influence of operating temperature on the produced hydrogen from the steam reforming reaction along the cell length and electrochemical characterization of SOFC with SDC-based electrolyte. The results indicate that the increase of operating temperature raises the reaction rate of methane steam reforming and the amount of the produced hydrogen at fuel channel. In addition, the cathode activation overpotential considerably reduces

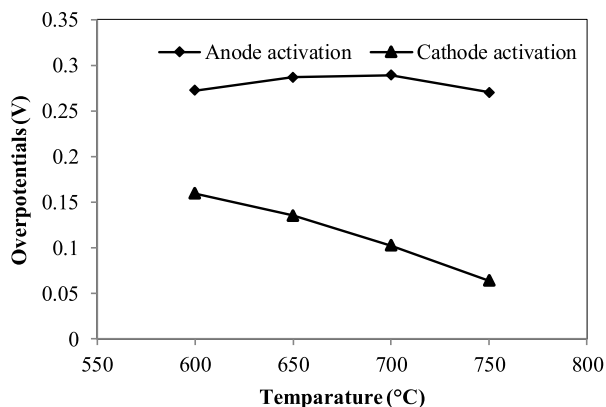


Fig. 4. Effect of operating temperature on average anode and cathode activation overpotentials.

with increasing temperature while the temperature has an insignificant effect on anode activation overpotential. Therefore, the power density of SOFC enhances from 0.19 to 0.42 A/m² with increasing temperature of 600 to 700 °C. However, the increment of power density reduces at higher temperature above 700 °C.

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