



## Short communication

## Electrochemical modeling of ammonia-fed solid oxide fuel cells based on proton conducting electrolyte

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## ABSTRACT

An electrochemical model was developed to study the  $\text{NH}_3$ -fed and  $\text{H}_2$ -fed solid oxide fuel cells based on proton conducting electrolyte (SOFC-H). The modeling results were consistent with experimental data in literature. It is found that there is little difference in working voltage and power density between the  $\text{NH}_3$ -fed and the  $\text{H}_2$ -fed SOFC-H with an electrolyte-support configuration due to an extremely high ohmic overpotential in the SOFC-H. With an anode-supported configuration, especially when a thin film electrolyte is used, the  $\text{H}_2$ -fed SOFC-H shows significantly higher voltage and power density than the  $\text{NH}_3$ -fed SOFC-H due to the significant difference in concentration overpotentials. The anode concentration overpotential of the  $\text{NH}_3$ -fed SOFC-H is found much higher than the  $\text{H}_2$ -fed SOFC-H, as the presence of  $\text{N}_2$  gas dilutes the  $\text{H}_2$  concentration and slows down the transport of  $\text{H}_2$ . More importantly, the cathode concentration overpotential is found very significant despite of the thin cathode used in the anode-supported configuration. In the SOFC-H,  $\text{H}_2\text{O}$  is produced in the cathode, which enables complete fuel utilization on one hand, but dilutes the concentration of  $\text{O}_2$  and impedes the diffusion of  $\text{O}_2$  to the reaction sites on the other hand. Thus, the cathode concentration overpotential is the limiting factor for the  $\text{H}_2$ -fed SOFC-H and an important voltage loss in the  $\text{NH}_3$ -fed SOFC-H. How to reduce the concentration overpotentials at both electrodes is identified crucial to develop high performance SOFC-H.

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## 1. Introduction

Ammonia ( $\text{NH}_3$ ) emerges as a promising fuel for fuel cells because it is relatively cheap, easy to store and transport, and relatively safe due to any leakage being easily detectable by its pungent odor. In addition, the infrastructure of ammonia technology has been well established. Therefore, there is increasing interest in using ammonia in fuel cells, especially solid oxide fuel cells (SOFCs) [1–17].

Conventionally, an SOFC is based on oxygen ion-conducting electrolyte (SOFC-O) and works at a high temperature of about 1273 K in order to achieve a high ionic conductivity. However, the high operating temperature limits the choice of materials used for SOFCs and also causes catalyst sintering and thermal expansion mismatch of SOFC components. In order to resolve these material and stability problems, great research efforts have been done to reduce the operating temperature to be about 773 K. However, the ionic conductivity of oxygen ion-conducting ceramics decreases considerably with decreasing temperature, leading to high ohmic loss at the electrolyte. For comparison, some proton conducting

ceramics show good ionic conductivity at intermediate temperatures and are thus good candidates for use as SOFC electrolyte [14]. Therefore, there is increasing interest in SOFC based on proton conducting electrolyte (SOFC-H) [2]. The most frequently used proton conducting electrolytes are  $\text{BaCeO}_3$ -based ceramics, such as  $\text{BaCeO}_3$  doped with Gd or Nd [2,11,14]. Promising anodes are Ni based composite materials consisting of Ni phase and ionic conducting phase, such as  $\text{Ce}_{0.8}\text{Gd}_{0.2}\text{O}_{1.9}$  (CGO)-Ni [2]. Compared with the conventional SOFC-O, the SOFC-H is advantageous in converting ammonia fuel to electricity. In the SOFC-H, steam is produced in the cathode, eliminating the need of gas separation in the anode. Recently, a thermodynamic analysis has been conducted to compare the theoretical performance of  $\text{NH}_3$ -fed SOFC with different electrolytes [18]. The study considers the Nernst potentials while the overpotentials are not included. It has also been demonstrated that the  $\text{NH}_3$ -fed SOFC-H had higher maximum energy efficiency than the SOFC-O [18].

However, the present literature is lacking electrochemical modeling on  $\text{NH}_3$ -fed SOFC-H, which is important for understanding and quantifying the actual performance of the  $\text{NH}_3$ -fed SOFC-H. In this short communication, an electrochemical model was developed to predict the current density–voltage ( $J$ – $V$ ) characteristics of the  $\text{NH}_3$ -fed SOFC-H. All overpotentials are included in the electrochemical model. The performance of  $\text{H}_2$ -fed SOFC-

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H is also investigated for comparison. The analyses presented in this paper provide better understanding on the working mechanisms of SOFC-H and can be a useful reference for SOFC-H design optimization.

## 2. Electrochemical model

In the NH<sub>3</sub>-fed SOFC-H (Fig. 1(a)), NH<sub>3</sub> undergoes decomposition over a Ni catalyst in the anode chamber as



The N<sub>2</sub> is removed from the anode chamber by the gas stream, while the H<sub>2</sub> produced undergoes electrochemical reactions at the anode–electrolyte interface to produce protons and electrons. The electrons traveling from the anode to the cathode through an external circuit yield an electrical power output. The protons are transported through dense electrolyte to the cathode chamber and undergo electrochemical reactions with oxygen molecules and electrons to form H<sub>2</sub>O at the cathode–electrolyte interface. For comparison, the working mechanisms of the H<sub>2</sub>-fed SOFC-H are shown in Fig. 1(b).

Although NH<sub>3</sub> thermal decomposition is sluggish at low temperatures without catalyst, it is favorable at high temperature with Ni catalyst. Several experimental works have reported that NH<sub>3</sub> could achieve 100% conversion at a temperature of above 873 K in the SOFC-H anode [11,12,14,16]. In a recent study on the NH<sub>3</sub>-fed electrolyte-support SOFC-H, 100% NH<sub>3</sub> decomposition was achieved in a thin anode layer with thickness of only a few tens of microns [11]. At a higher temperature ( $T > 873\text{K}$ ), the rate of NH<sub>3</sub> decomposition will be much higher, thus complete conversion of NH<sub>3</sub> to N<sub>2</sub> and H<sub>2</sub> can be achieved in a much thinner layer of the anode. Compared with the large anode thickness (0.5–1.0 mm) in the anode-supported SOFC-H, the anode layer involved in NH<sub>3</sub> decomposition is negligibly small. Therefore, in the present study, it can be safely assumed that complete decomposition of NH<sub>3</sub> is attained at the surface of the Ni-based anode. This assumption can greatly simplify the electrochemical model without seriously

decreasing the model accuracy, especially at a temperature higher than 873 K.

Considering all the overpotentials in the NH<sub>3</sub>-fed SOFC-H, the external voltage ( $V$ ) can be expressed as

$$V = E - \eta_{\text{con,a}} - \eta_{\text{con,c}} - \eta_{\text{act,a}} - \eta_{\text{act,c}} - \eta_{\text{ohm}} \quad (2)$$

where  $E$  is the equilibrium (reversible) potential of the SOFC-H, which can be calculated with the Nernst equation [19,20];  $\eta_{\text{act,a}}$  and  $\eta_{\text{act,c}}$  are the activation overpotentials at the anode and cathode, respectively, which can be calculated with the Butler–Volmer equation [20];  $\eta_{\text{ohm}}$  is the ohmic overpotential at the electrolyte and can be analytically obtained by Ohm's law [20];  $\eta_{\text{con,a}}$  and  $\eta_{\text{con,c}}$  are the concentration overpotentials at the anode and cathode, respectively. Concentration overpotential measures the resistance of the porous electrode to transport of reactants to and the products from the electrochemical reaction sites. In the NH<sub>3</sub>-fed SOFC-H, the concentration overpotentials can be expressed in terms of the gas concentration difference between the electrode surface and the electrode–electrolyte interface,

$$\eta_{\text{con,a}} = \frac{RT}{2F} \ln \left( \frac{P_{\text{H}_2}^{\text{S}}}{P_{\text{H}_2}^{\text{I}}} \right) \quad (3)$$

$$\eta_{\text{con,c}} = \frac{RT}{2F} \ln \left[ \frac{(P_{\text{O}_2}^{\text{S}})^{0.5} P_{\text{H}_2\text{O}}^{\text{I}}}{(P_{\text{O}_2}^{\text{I}})^{0.5} P_{\text{H}_2\text{O}}^{\text{S}}} \right] \quad (4)$$

where  $P_{\text{H}_2}$ ,  $P_{\text{H}_2\text{O}}$  and  $P_{\text{O}_2}$  are the partial pressure of H<sub>2</sub>, H<sub>2</sub>O, and O<sub>2</sub>; the superscripts I and S represent the electrode–electrolyte interface and the electrode surface, respectively.

The dusty gas model (DGM) is used to model the multi-component mass transfer within the anode and cathode as it can predict the mass transfer in porous media more accurately than the Fick's model and Stefan-Maxwell model [21,22]. According to DGM, the transport of gas species in both anode and cathode is governed by

$$\frac{N_i}{D_{i,k}^{\text{eff}}} + \sum_{j=1, j \neq i}^n \frac{y_j N_j - y_i N_j}{D_{ij}^{\text{eff}}} = -\frac{1}{RT} \left[ P \frac{dy_i}{dx} + y_i \frac{dP}{dx} \left( 1 + \frac{B_0 P}{D_{i,k}^{\text{eff}} \mu} \right) \right] \quad (5)$$

where  $N_i$  is the flux of gas species  $i$ ;  $y_i$  is the molar fraction of gas species  $i$ ;  $D_{i,k}^{\text{eff}}$  is the effective Knudsen diffusion coefficient of species  $i$ ;  $D_{ij}^{\text{eff}}$  is the effective binary molecular diffusion coefficient of species  $i$  and  $j$ ;  $x$  is the depth in electrode measured from the electrode surface;  $B_0$  is the permeability of the porous electrode; and  $\mu$  is the viscosity of the gas mixture. The calculation procedures of the diffusion coefficients, permeability, as well as the viscosity of gas mixture can be found elsewhere [23–25].

At the electrochemical reaction sites (electrode–electrolyte interface), the fluxes of reacting species can be written as:  $N_{\text{H}_2}|_{x=d_a} = (J/2F)$ ,  $N_{\text{H}_2\text{O}}|_{x=d_c} = -(J/2F)$ , and  $N_{\text{O}_2}|_{x=d_c} = (J/4F)$ . In the above boundary conditions,  $J$  is the current density;  $d_a$  and  $d_c$  are the thickness of anode and cathode, respectively. N<sub>2</sub> is not involved in electrochemical reactions and thus its flux is equal to zero. With given gas composition at the electrode surface and given fluxes at the electrode–electrolyte interface, the governing equation (Eq. (5)) for mass transfer in porous electrodes can be solved with finite difference method. After obtaining the molar fraction/partial pressure of each gas species inside the porous electrodes, the concentration overpotentials can thus be calculated with Eqs. (3) and (4). Including the equilibrium potential, ohmic and activation overpotentials, the working voltage of the NH<sub>3</sub>-fed SOFC-H can thus be obtained. The model development and validation for the H<sub>2</sub>-fed SOFC-H have been presented in a previous publication [23].

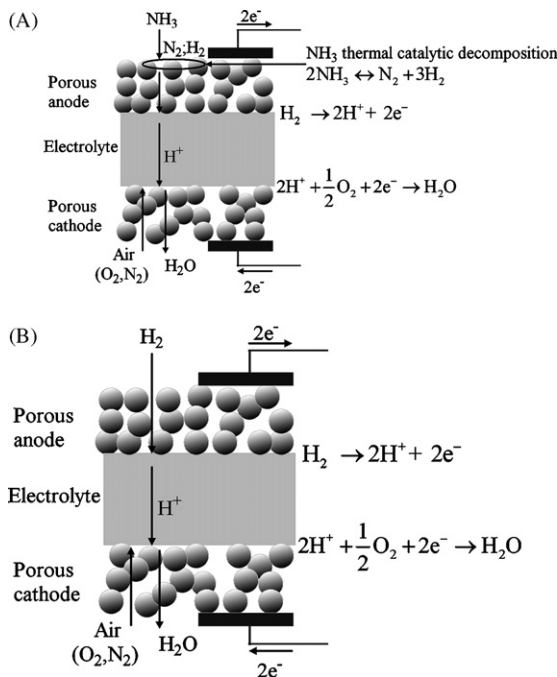


Fig. 1. Working mechanisms of the SOFC-H (a) NH<sub>3</sub>-fed SOFC-H and (b) H<sub>2</sub>-fed SOFC-H.

**Table 1**  
Input parameters for the present electrochemical model of the NH<sub>3</sub>-fed SOFC-H and H<sub>2</sub>-fed SOFC-H

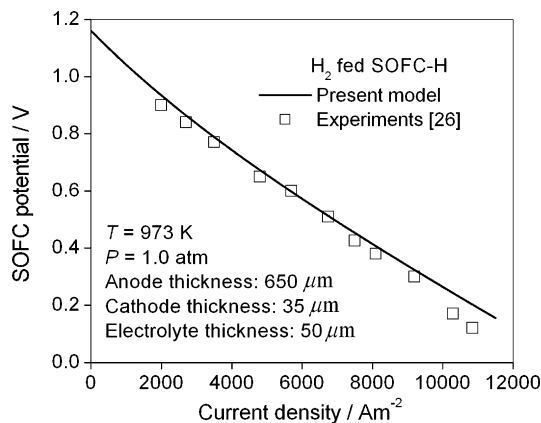
Parameter	Value
Operating temperature, <i>T</i> (K)	1073
Operating pressure, <i>P</i> (atm)	1.0
Gas composition (%) at the electrode surface	
Anode of NH <sub>3</sub> fed SOFC-H (molar fraction of H <sub>2</sub> /N <sub>2</sub> )	66.7/33.3 (see text)
Anode of H <sub>2</sub> fed SOFC-H	Pure H <sub>2</sub>
Cathode (molar fraction of H <sub>2</sub> O/O <sub>2</sub> /N <sub>2</sub> )	3.0/18.0/79.0 [23]
Exchange current density of anode (Am <sup>-2</sup> )	5300.0 [19,24]
Exchange current density of cathode (Am <sup>-2</sup> )	2000.0 [19,24]
Electrode porosity	0.4 [23]
Electrode tortuosity	5.0 [23]
Electrode pore radius (μm)	0.5 [23]
Conductivity of electrolyte (i.e. Sm-doped BaCeO <sub>3</sub> ) at 1073 K	1.22 S m <sup>-1</sup> (estimated from [26])
Component thickness (μm)	
Electrolyte-supported SOFC-H	
Electrolyte	500.0
Anode	50.0
Cathode	50.0
Anode-supported SOFC-H	
Electrolyte	50.0
Anode	500.0
Cathode	50.0
Anode-supported SOFC-H with thin film	
Electrolyte	10.0
Anode	500.0
Cathode	50.0

**3. Results and discussion**

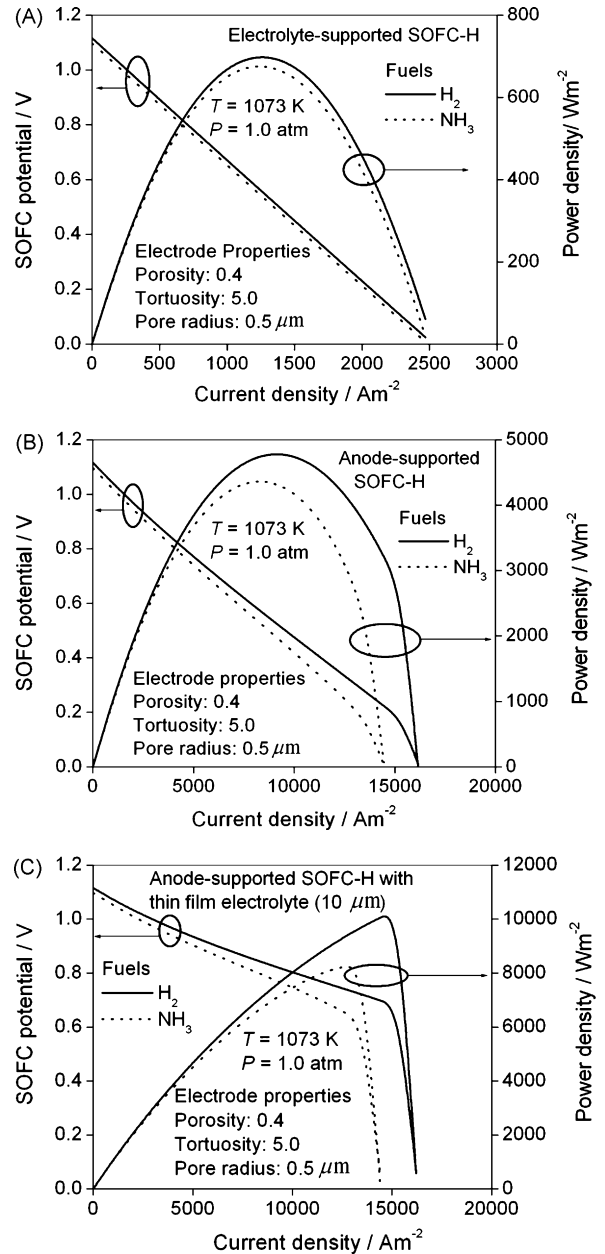
In this section, the performances of the NH<sub>3</sub>-fed and H<sub>2</sub>-fed SOFC-H are studied and compared. The values of input parameters are summarized in Table 1. A comparison between the present modeling results and experimental data from reference [26] is shown in Fig. 2. Good agreement between the modeling results and experimental data validates the present model.

**3.1. Working voltage and power density of the NH<sub>3</sub>-fed and H<sub>2</sub>-fed SOFC-H with different support configurations**

From the previous analyses on the H<sub>2</sub>-fed SOFC-O, it is known that the fuel cell component thickness greatly influences the elec-



**Fig. 2.** Comparison between present modeling results and experimental data from literature for model evaluation.



**Fig. 3.** Comparison in working voltage and power density between the NH<sub>3</sub>-fed SOFC-H and the H<sub>2</sub>-fed SOFC-H: (a) electrolyte-supported SOFC-H, (b) anode-supported SOFC-H, and (c) anode-supported SOFC-H with advanced thin electrolyte.

trical performance of the SOFC-O [19,20]. In this section, the working voltage and power density of the NH<sub>3</sub>-fed and the H<sub>2</sub>-fed SOFC-H are investigated and compared with different component thickness. Being consistent with experimental studies [1–17], the NH<sub>3</sub>-fed SOFC-H always shows lower working voltage and power density than the H<sub>2</sub>-fed SOFC-H for both the electrolyte-support and electrode-support configurations (Fig. 3(a)–(c)). However, there is only little difference between the NH<sub>3</sub>-fed SOFC-H and the H<sub>2</sub>-fed SOFC-H with an electrolyte-supported configuration (Fig. 3(a)). With an anode-supported configuration, the H<sub>2</sub>-fed SOFC-H shows about 10% higher maximum power density than the NH<sub>3</sub>-fed SOFC-H (Fig. 3(b)). It is also found that in the advanced configuration of the anode-supported SOFC-H with a thin film electrolyte (10 μm), the difference between the H<sub>2</sub>-fed SOFC-H and the NH<sub>3</sub>-fed SOFC-H becomes much more significant, i.e. the maximum

power density of the H<sub>2</sub>-fed SOFC-H is about 25% higher than the NH<sub>3</sub>-fed SOFC-H.

In an experimental research conducted by Maffei et al. [9], little difference (less than 10%) in maximum power density was observed between the NH<sub>3</sub>-fed and the H<sub>2</sub>-fed SOFC-H with an electrolyte-supported configuration (electrolyte thickness: 1.3 mm). In another study conducted by Meng et al. [13], the maximum power density of the NH<sub>3</sub>-fed SOFC-H was found to be 36% lower than the H<sub>2</sub>-fed SOFC-H with an anode-supported configuration (electrolyte thickness: 10 μm). The difference in maximum power density between the H<sub>2</sub>-fed SOFC-H and the NH<sub>3</sub>-fed SOFC-H in Meng's experiments is comparatively high partly due to the use of liquid ammonia in their experiment, which could result in lower cell temperature and even incomplete decomposition of NH<sub>3</sub>. Nevertheless, they also mentioned that the overpotentials between the NH<sub>3</sub>-fed SOFC-H and the H<sub>2</sub>-fed SOFC-H could be high in their experiments. In general, the modeling results in the present study are consistent with experimental data from literature.

### 3.2. Ohmic and activation losses in SOFC-H

The higher voltage and power density of the H<sub>2</sub>-fed SOFC-H is partly due to a higher H<sub>2</sub> partial pressure in the anode layer. In the H<sub>2</sub>-fed SOFC-H, H<sub>2</sub> is the only gas species present in the anode thus its pressure can be always maintained at 1.0 atm. For comparison, in the NH<sub>3</sub>-fed SOFC-H, H<sub>2</sub> is diluted by the presence of N<sub>2</sub> produced from NH<sub>3</sub> decomposition. A lower H<sub>2</sub> partial pressure in the NH<sub>3</sub>-fed SOFC-H can thus lead to lower open-circuit voltage as well as lower voltage and power density. In order to better understand the electrical performance of the SOFC-H, the ohmic and activation overpotentials are investigated and shown in Fig. 4(a) and (b),

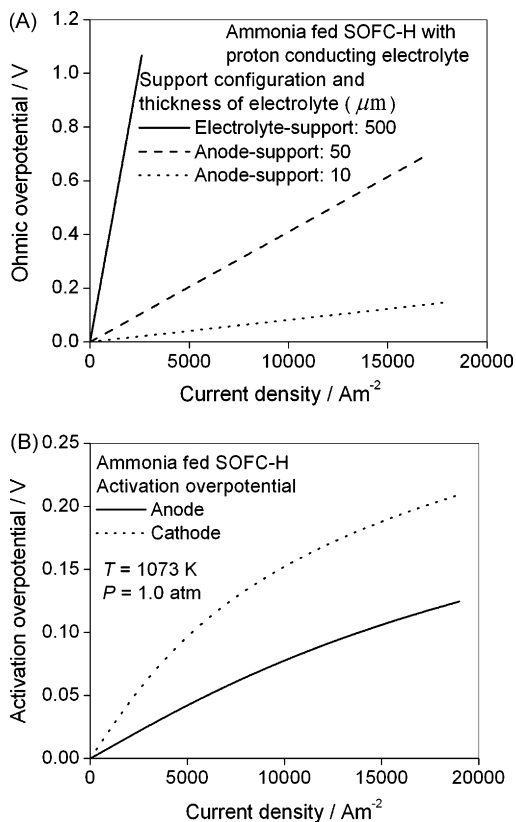


Fig. 4. Ohmic and activation losses in the SOFC-H fed with NH<sub>3</sub> and H<sub>2</sub>: (a) ohmic overpotential and (b) activation overpotential.

while the concentration overpotentials are discussed more in detail in the next section. As expected, the ohmic overpotential plays an extremely important role in governing the current density–voltage (*J*–*V*) characteristics of the SOFC-H due to low proton conductivity of the existing electrolyte materials. The high ohmic overpotential can be greatly reduced by fabricating thin film electrolyte for SOFC-H (Fig. 4(a)). Similar to the conventional SOFC-O, the NH<sub>3</sub>-fed SOFC-H has higher cathode activation overpotential than anode activation overpotential due to slower electrochemical reaction taking place at the cathode–electrolyte interface (Fig. 4(b)).

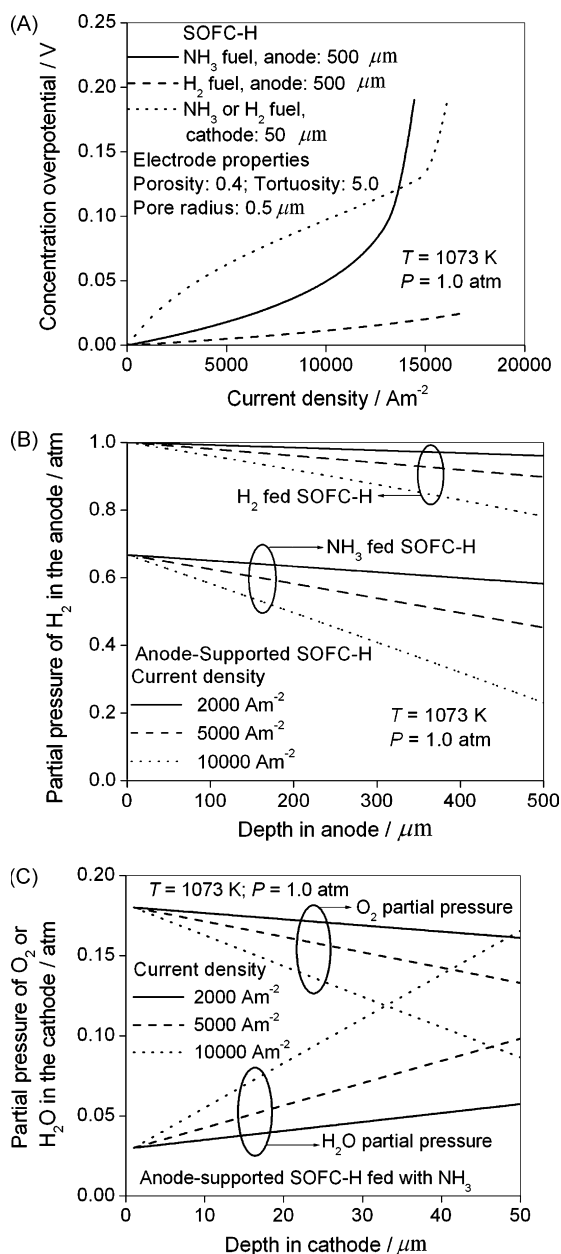
### 3.3. Concentration overpotentials in H<sub>2</sub>-fed and NH<sub>3</sub>-fed SOFC-H

The most important finding of this study is the significant difference in concentration overpotentials between the NH<sub>3</sub>-fed SOFC-H and the H<sub>2</sub>-fed SOFC-H as well as the H<sub>2</sub>-fed conventional SOFC-O. The anode-supported configuration is investigated because it is superior to electrolyte-supported and cathode-supported configurations. It is found that the anode concentration overpotential of the H<sub>2</sub>-fed SOFC-H is much lower than the NH<sub>3</sub>-fed SOFC-H (Fig. 5(a)). This is because the presence of N<sub>2</sub> in the anode of the NH<sub>3</sub> fuel cell slows down the transport of H<sub>2</sub> from the anode surface to the anode–electrolyte interface, resulting in a lower H<sub>2</sub> partial pressure in the anode and higher concentration overpotential (Fig. 5(a) and (b)). As a result, a limiting current density of about 15,000 A m<sup>-2</sup> is found. This means that the performance of the NH<sub>3</sub>-fed SOFC-H is limited by transport of H<sub>2</sub> to the reaction sites at a high current density. It also explains why both voltage and power density of the NH<sub>3</sub>-fed SOFC-H quickly drop to zero at a current density of around 15,000 A m<sup>-2</sup> (Fig. 3(b) and (c)). For comparison, H<sub>2</sub>, which presents as the single gas species in the anode of the H<sub>2</sub>-fed SOFC-H, can be transported efficiently from the anode surface to the anode–electrolyte interface, leading to a much higher H<sub>2</sub> partial pressure in the anode and thus lower concentration overpotentials (Fig. 5(a) and (b)).

Another important phenomenon observed from Fig. 5(a) is that the cathode concentration overpotential is significant despite of the thin cathode used (only 50 μm) in the anode-supported SOFC-H. The cathode concentration overpotential is comparable to the anode concentration overpotential in the NH<sub>3</sub>-fed SOFC-H but is much higher than the anode concentration overpotential in the H<sub>2</sub>-fed SOFC-H. Therefore, in the H<sub>2</sub>-fed SOFC-H, the cathode concentration overpotential is the limiting factor for the performance of the SOFC-H and this explains why the voltage and power density of the H<sub>2</sub>-fed SOFC-H suddenly drop to zero at a current density of about 16,000 A m<sup>-2</sup> (Fig. 3). In case NH<sub>3</sub> is used as a fuel in the SOFC-H, the cathode concentration overpotential is also as important as the anode concentration overpotential. In addition, the cathode concentration overpotential of the SOFC-H is also considerably higher than that of the conventional SOFC-O fed with H<sub>2</sub> [19]. This is because H<sub>2</sub>O is produced in the SOFC-H cathode, while it is produced in the SOFC-O anode. The presence of H<sub>2</sub>O in the cathode not only dilutes the concentration of O<sub>2</sub>, but also impedes the transport of O<sub>2</sub> from the cathode surface to the cathode–electrolyte interface, thus leading to a lower O<sub>2</sub> partial pressure and higher concentration overpotential of the SOFC-H (Fig. 5(a) and (c)). In addition, as the molar production rate of H<sub>2</sub>O is twice that of O<sub>2</sub>, a higher pressure at the cathode–electrolyte interface than the cathode surface is established. This pressure gradient could also impede the transport of O<sub>2</sub>.

Presently, most of the research works done on NH<sub>3</sub>-fed SOFC-H are focused on the development of new electrolyte materials with high proton conductivity but little attention has been paid on the microstructure of the electrodes. With the development of new materials and fabrication of thin electrolyte with a thick-





**Fig. 5.** Concentration loss and distribution of gas partial pressures in porous electrodes of the SOFC-H fed with NH<sub>3</sub> and H<sub>2</sub>: (a) concentration overpotentials, (b) partial pressure of H<sub>2</sub> in the anode, and (c) partial pressures of H<sub>2</sub>O and O<sub>2</sub> in the cathode.

ness of about 10  $\mu\text{m}$ , the ohmic overpotential will no longer be the dominating voltage loss, i.e. with thin electrolyte (10  $\mu\text{m}$ ), the ohmic overpotential is less than 0.1 V at a high current density of 10,000  $\text{A m}^{-2}$ . Therefore, the electrode concentration overpotentials should be seriously considered for development of high performance SOFC-H. Especially, the microstructure of the cathode needs to be optimized. In order to reduce the considerable concentration overpotential, large particles or pore size is needed, which, however, will reduce the length of triple phase boundary (TPB), where most the electrochemical reactions take place. On the other hand, in order to enhance the electrode's electrocatalytic activity, fine particles or small pores are needed at the electrode–electrolyte interface to ensure large TPB length. Therefore, the thin cathode of the SOFC-H needs to be carefully designed to minimize both activation and concentration overpotentials. A recent study has

demonstrated that functionally graded electrodes (FGE) could be effective to reduce the total overpotential of electrodes of the SOFC-O fed with H<sub>2</sub> [22]. Application of the FGE concept to the SOFC-H is expected to be fruitful.

#### 4. Conclusion

Electrochemical modeling analyses have been performed to study the electrical performance of SOFC-H fed with NH<sub>3</sub> and H<sub>2</sub>. It is found that the NH<sub>3</sub>-fed SOFC-H always has lower voltage and power density than the H<sub>2</sub>-fed SOFC-H. However, the differences in voltage and power density between the NH<sub>3</sub>-fed SOFC-H and H<sub>2</sub>-fed SOFC-H are insignificant for the electrolyte-supported configuration. These differences become larger for the anode-supported SOFC-H, especially when a thin film electrolyte is used.

It is found that the anode concentration overpotential of the NH<sub>3</sub>-fed SOFC-H is much higher than the H<sub>2</sub>-fed SOFC-H as the presence of N<sub>2</sub> dilutes the H<sub>2</sub> concentration and slows down the transport of H<sub>2</sub>. More importantly, the cathode concentration of the SOFC-H is found significant although the thickness of cathode is only 50  $\mu\text{m}$ . This is because the H<sub>2</sub>O formed in the cathode dilutes the O<sub>2</sub> concentration and impedes the diffusion of O<sub>2</sub> to the TPB. In case NH<sub>3</sub> is used as a fuel for the SOFC-H, the cathode concentration overpotential is comparable to the anode concentration overpotential. In case H<sub>2</sub> fuel is used, the cathode concentration overpotential becomes the limiting factor for the SOFC-H performance. In order to further improve the performance of the NH<sub>3</sub>-fed or a H<sub>2</sub>-fed SOFC-H, optimization of the electrode microstructure to minimize the activation and concentration overpotentials becomes extremely crucial.

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