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# The effect of electrolyte type on performance of solid oxide fuel cells running on hydrocarbon fuels

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## ABSTRACT

A two-dimensional model is developed to simulate the performance of methane fueled solid oxide fuel cells (SOFCs), focusing on the effect of electrolyte type on SOFC performance. The model considers the heat and mass transfer, direct internal reforming (DIR) reaction, water gas shift reaction (WGSR), and electrochemical reactions in SOFCs. The electrochemical oxidation of CO in oxygen ion-conducting SOFC (O-SOFC) is considered. The present study reveals that the performance of H-SOFC is lower than that of O-SOFC at a high temperature or at a low operating potential, as electrochemical oxidation of CO in O-SOFC contributes to power generation. This finding is contrary to our common understanding that proton conducting SOFC (H-SOFC) always performs better than O-SOFC. However, at a high operating potential of 0.8 V or at a lower temperature, H-SOFC does exhibit better performance than O-SOFC due to its higher Nernst potential and higher ionic conductivity of the electrolyte. This indicates that the proton conductors can be good choices for SOFCs at intermediate temperature, even with hydrocarbon fuels. The results provide better understanding on how the electrolyte type influences the performance of SOFCs running on hydrocarbon fuels.

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## 1. Introduction

Solid oxide fuel cells (SOFCs) are promising electrochemical energy conversion devices for clean power generation. One distinct feature of SOFCs is their fuel flexibility, as high operating temperature (i.e. 1073 K) enables internal reforming of hydrocarbon fuels or thermal cracking of ammonia in the porous anode of SOFC [1–3]. Thus, in principle all combustible fuels can be utilized in SOFCs for electricity generation, such as hydrogen, methane, methanol, ethanol, ammonia, dimethyl ether (DME) [4–7].

Conventionally SOFCs employ oxygen ion-conducting ceramics as electrolyte (here termed as O-SOFCs), such as yttria-stabilized zirconia (YSZ) [1]. In the recent years, proton

conducting materials (i.e. BaCeO<sub>3</sub> doped with Gd or Sm) have also been demonstrated as good electrolyte materials for SOFCs (here termed as H-SOFCs) [8]. The use of proton conducting electrolyte in H-SOFC changes the location of steam production from the anode to the cathode, allowing high fuel utilization. In addition, due to a higher hydrogen concentration in the anode, the Nernst potential of H-SOFC is higher than that of O-SOFC. Several thermodynamic analyses have shown that the maximum efficiency of H-SOFC is higher than that of O-SOFC with H<sub>2</sub> and hydrocarbon fuels [9–12]. To examine the actual performance of H-SOFC considering various overpotential losses, electrochemical models have been developed to compare H-SOFC with O-SOFC [13–15]. The electrochemical modeling reveals that H-SOFC has lower

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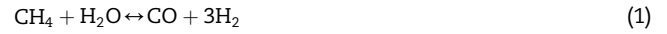
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anode concentration overpotential than O-SOFC but the cathode overpotential in H-SOFC is considerably increased as the steam impedes the transport of oxygen [15]. Moreover, it's found that the actual performance of H-SOFC may not be higher than O-SOFC if the ionic conductivity of the electrolyte in H-SOFC is not higher than that of O-SOFC [13,15]. Recently, Ishak et al. [16] performed thermodynamic and an electrochemical modeling analyses, adopting the same methodologies with references [12,17] but a higher proton conductivity for H-SOFC. It's found that the H-SOFC performance is higher than O-SOFC, due to a lower ohmic overpotential of the electrolyte and higher hydrogen concentration in H-SOFC anode [16]. As the typical ionic conductivity of H-SOFC electrolyte is higher than that of YSZ for O-SOFC, it is expected that the actual performance of H-SOFC should be higher than that of O-SOFC.

In the above-mentioned studies on H-SOFC and O-SOFC, only H<sub>2</sub> is considered as an electrochemically active fuel and the electrochemical oxidation of CO is completely neglected. However, experimental investigations have confirmed the electrochemical oxidation of CO in the anode of O-SOFCs, although its reaction kinetics is lower than that of H<sub>2</sub> electrochemical oxidation [18–21]. Electrochemical oxidation of CO in O-SOFC can increase the current density and thus contribute to power generation. However, H-SOFC does not support CO electrochemical oxidation. Thus, O-SOFC should have an essential advantage over H-SOFC with hydrocarbon fuels. Our recent thermodynamic analysis shows when CO electrochemical oxidation is considered, the maximum efficiency of O-SOFC is higher than that of H-SOFC [22]. As the thermodynamic analysis does not consider any overpotential loss, it is still unknown which type of SOFC has higher actual performance. To answer this question, a two-dimensional model is developed to compare the actual performance of H-SOFC and O-SOFC considering CO electrochemical oxidation in O-SOFC anode. All the complicated physical–chemical processes are considered, including heat and mass transfer, direct internal reforming (DIR) reaction, water gas shift reaction (WGSR), and electrochemical reactions.

## 2. Model development

The working principles and computational domains for CH<sub>4</sub> fed H-SOFC and O-SOFC are shown in Fig. 1(a) and (b), respectively. The computational domain includes the interconnector, fuel channel, porous anode, dense electrolyte, porous cathode and the air channel. The anode-support configuration is adopted as it can yield higher SOFC performance than cathode-support and electrolyte-support configurations [13,15]. The planar configuration is used as it is widely used in practice. However, it is understood that the button cell configuration should be used for detailed comparison with experimental data, as experiments are usually conducted with button cells. In operation, pre-reformed methane gas mixture is supplied to the anode channel while air is supplied to the cathode channel. In both H-SOFC and O-SOFC, DIR of methane and WGSR take place in the porous anode, represented by Eqs. (1) and (2), respectively.



### 2.1. O-SOFC

In O-SOFC, oxygen molecules diffuse from the cathode surface to the cathode–electrolyte interface and react with electrons to produce oxygen ions (Eq. (3)), which are subsequently transported to the anode side via the dense oxygen ion-conducting electrolyte. At the anode side, H<sub>2</sub> and CO molecules transport to the triple-phase boundary (TPB) at the anode–electrolyte interface, where they react with oxygen ions to produce electrons, H<sub>2</sub>O, and CO<sub>2</sub>, as shown in Eqs. (4) and (5).



The electrochemical oxidation of CH<sub>4</sub> is neglected due to its relatively low reaction kinetics [18]. In addition, reaction between CO<sub>2</sub> and CH<sub>4</sub> is not considered. Based on the working principles, a 2D thermo-electrochemical model is developed to simulate the coupled transport and reaction phenomena in O-SOFC. The 2D model consists of 3 sub-models: (1) an electrochemical model; (2) a chemical model; and (3) a computational fluid dynamics (CFD) model.

#### 2.1.1. Electrochemical model

The electrochemical model is used to calculate the local current density ( $J$ ) at given operating potentials ( $V$ ). The use of interconnector with high electrical conductivity along the entire flow channel leads to uniform operating potential along the main flow stream. Thus, the  $J$ – $V$  relationship can be established by solving the equations below [23].

$$V = E - \eta_{\text{act},a} - \eta_{\text{act},c} - \eta_{\text{ohmic}} \quad (6)$$

$$E_{\text{H}_2} = 1.253 - 0.00024516T + \frac{RT}{2F} \ln \left[ \frac{P_{\text{H}_2}^{\text{I}} (P_{\text{O}_2}^{\text{I}})^{0.5}}{P_{\text{H}_2\text{O}}^{\text{I}}} \right] \quad (7)$$

$$E_{\text{CO}} = 1.46713 - 0.0004527T + \frac{RT}{2F} \ln \left[ \frac{P_{\text{CO}}^{\text{I}} (P_{\text{O}_2}^{\text{I}})^{0.5}}{P_{\text{CO}_2}^{\text{I}}} \right] \quad (8)$$

where  $E$  is the equilibrium potential and the subscripts H<sub>2</sub> and CO represent the equilibrium potential associated with H<sub>2</sub> and CO fuels;  $T$  is temperature (K).  $R$  is the universal gas constant (8.3145 J mol<sup>-1</sup> K<sup>-1</sup>); and  $F$  is the Faraday constant (96,485 C mol<sup>-1</sup>).  $P^{\text{I}}$  used in Eqs. (7) and (8) refers to the partial pressure at the electrode–electrolyte interface. Thus, the concentration overpotentials at the electrodes are included in the Nernst potential ( $E$ ).  $\eta_{\text{ohmic}}$  is the ohmic overpotential and can be determined with the Ohm's law (Eq. (9)).

$$\eta_{\text{ohmic}} = JL \frac{1}{\sigma_{\text{ionic}}} \quad (9)$$

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