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Two-dimensional micro/macroscale model for intermediate-temperature solid oxide fuel cells considering the direct internal reforming of methane

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ABSTRACT

In this study, a two-dimensional micro/macroscale model is developed to simulate the operation of anode-supported, planar, intermediate-temperature solid oxide fuel cells (IT-SOFCs) fed with partially reformed methane fuel. The previous micro/macroscale model for hydrogen-fueled IT-SOFCs is extended to take into account the direct internal reforming (DIR) of methane inside the porous cermet anode and the multi-component mass transport and reforming reaction heat consumption. The intrinsic reaction kinetics for steam methane reforming (SMR) at the nickel catalyst surface is fully considered based on the micro/macroscale calculation framework under the assumption of fully-developed laminar channel flow. Using the developed micro/macroscale model, a detailed investigation of the methane-fueled IT-SOFC operation is conducted, followed by parametric studies on the effects of the inlet temperature, the co- or counter-flow configuration, the air flow rate, and the cell length on performance.

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Introduction

Solid oxide fuel cells (SOFCs) are promising energy conversion devices that generate electric power at higher efficiencies than conventional heat engines [1–3]. The operating temperature of SOFCs ranges from approximately 600 to 1000 °C, which provides several advantages, including the use of non-

precious metal catalyst to reduce manufacturing costs and the production of high-quality waste heat to use in hybrid or co-generation systems. Fuel flexibility is also among the main advantages of SOFCs. Accordingly, various hydrocarbon fuels, such as methane, methanol, ethanol, and gasoline, have been studied as an alternative fuel for SOFCs [4–6].

Methane is considered one of the promising alternative fuels for SOFCs due to its natural abundance among

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hydrocarbon fuels and its well-developed infrastructure (city gas) in many countries. In methane-fueled SOFCs, methane should first be reformed to hydrogen to participate in the electrochemical power generation. This reforming process can be performed at the external reformer before the fuel gas enters the SOFC or at the porous anode while the fuel gas flows through the channels of the SOFC. The direct internal reforming (DIR) of methane inside the SOFC has been extensively investigated to achieve a simpler and more cost-effective design of SOFC systems [7–9].

The DIR process in SOFCs is equivalent to steam methane reforming (SMR), which is a well-known industrial process for the large-scale production of hydrogen from methane [10–12]. In the SMR process, methane (CH_4) and water vapor (H_2O) react on nickel (Ni) catalyst surface (heterogeneous reaction), thereby producing hydrogen (H_2) and carbon dioxide (CO_2). The thick and porous anode structure in anode-supported SOFCs, composed of Ni and yttria-stabilized zirconia (YSZ) particles, provides the rich catalytic surface area required for methane reforming. The high operating temperature of the SOFCs and the electrochemical reaction heat facilitate the endothermic SMR process. In addition, the water produced from the electrochemical reactions further increases the amount of water vapor, which promotes the reforming reaction and also the water-gas shift (WGS) reaction that converts carbon monoxide (CO) and water vapor (H_2O) into hydrogen (H_2) and carbon dioxide (CO_2).

Although the DIR of methane is beneficial for the simple and cost-effective design of SOFCs, it may pose several problems, such as a larger temperature gradient and higher carbon deposition. In many SOFCs, the reforming reaction typically occurs in the porous anode within a short axial distance from the fuel inlet. A highly localized reaction rate leads to a large temperature gradient due to the strong endothermic nature of the SMR process. In addition, carbon deposition may occur when the steam-to-carbon (S/C) ratio of the fuel gas stream is not sufficiently high. The carbon deposition decreases the catalytic activity by covering the Ni surface area and increases the mass transport resistance by blocking the pore space. Such problems with DIR can be resolved by the control of the operating conditions (including the S/C ratio) and the development of advanced electrode structures and materials.

Extensive studies were conducted to investigate the operation of SOFCs with and without the internal SMR process inside the porous anode [7,8,13–20]. Different from the experimental approaches, numerical simulations provided detailed information regarding the temperature distribution, gas concentration, and reaction rate inside the SOFCs. In addition, parametric studies based on numerical simulations have clarified the effects of various operating conditions (such as the flow configuration, inlet temperature, and so on) on the SMR process inside the anode and the electrochemical performance of the SOFCs. However, most numerical studies have been based on the empirical or simplified current–voltage characteristics to model the electrochemical performance at the positive electrode/electrolyte/negative electrode (PEN) structure.

Microscale electrode models have been successful in determining the correlations between the microstructural and dimensional parameters of the porous electrodes and the

electrochemical performance of the SOFCs [21–28]. Important electrode parameters include the porosity, particle size (ionic and electronic), electronic phase fraction, and electrode thickness; these parameters influence the electrochemical reaction by changing the three-phase boundary length (TPBL), the charge transport by altering phase conductivity (ionic and electronic), and the mass transport by modifying tortuosity and pore size. These microscale models have been further extended to micro/macroscale models by including a macroscale analysis for fluid flow and mass and heat transport in the gas channels [29–32].

The DIR process is also dependent on the microstructural and dimensional parameters of the anode; smaller Ni particles and thicker anodes can provide larger Ni catalyst surface area for the SMR reactions. However, few micro/macroscale models have been developed for simulating methane-fueled SOFCs with DIR. A microscale model combined with a proper DIR process consideration can provide a more complete picture how microstructural, dimensional, and operational parameters impact the performance of DIR-SOFCs operated with methane fuel. Therefore, in this study, the two-dimensional micro/macroscale model for hydrogen-fueled intermediate-temperature SOFCs (IT-SOFCs) previously developed by Sohn et al. [31] is extended to include the SMR reactions inside the porous cermet anode. Based on the micro/macroscale calculation framework of [31], the new model fully considers the intrinsic kinetics of the SMR reactions at the Ni catalyst surface, as well as the accompanying multi-component mass transport and the reforming reaction heat consumption. Using the developed model, the detailed operation of methane-fueled SOFCs is simulated, and the effects of various operating conditions are investigated.

Theory and calculation

The two-dimensional micro/macroscale model for methane-fueled IT-SOFCs is developed by extending the previous model for hydrogen-fueled IT-SOFCs [31] to consider the DIR of methane. In these models, the electrochemical reactions and (charge and mass) transport processes in the porous electrodes are calculated according to the comprehensive microscale model [25,26], based on the statistical methods for electrode microstructure characterization [33–35], the continuum models for the electrode process [21–24], and the experimental data for patterned electrodes [36–38]. The comprehensive microscale model is combined with the macroscale model for mass and heat transport and fluid flow in the gas channels to build the micro/macroscale models, by assuming fully-developed axial velocity profiles in the gas channels. The overall numerical framework of the micro/macroscale model for hydrogen-fueled IT-SOFCs is presented in Ref. [31] and thus will not be repeated here. Instead, the extension of the previous model toward the model for methane-fueled IT-SOFCs is explained in detail.

Model description

An anode-supported, planar IT-SOFC operated with methane is illustrated in Fig. 1, where the use of standard materials is

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