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Full Length Article

Temperature and reactive current distributions in microtubular solid oxide electrolysis cells

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ARTICLE INFO

Article history:

Received 15 March 2016

Received in revised form

27 April 2016

Accepted 12 May 2016

Available online xxx

Keywords:

Solid oxide electrolysis cells

Micro-tubular

High temperature steam electrolysis

Temperature distributions

ABSTRACT

A two-dimensional tubular model of a cathode supported microtubular solid oxide electrolysis cell (SOEC) was developed in order to investigate the temperature distribution within the cell during high-temperature steam electrolysis. High-temperature steam electrolysis using SOECs allows for highly efficient hydrogen generation, because the heat from the overpotential can be recycled in the form of a heat source for the electrolysis reaction. However, to improve the durability and strength of the components, an understanding of the temperature distribution of the cell is essential, because heat absorption during electrolysis reaction leads to complex temperature distributions. The current density–voltage curves obtained using the above-mentioned model were in good agreement with the experimental ones. The calculated and measured temperature distributions indicated that the distribution in microtubular SOECs is a nonuniform one and suggested that the current-collecting positions strongly affect the temperature and reaction distributions under the conditions used herein.

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Introduction

In recent years, energy from natural energy sources such as solar and wind power has been added to the electric grid, as it is clean and renewable. However, both wind and solar power exhibit instability with respect to changes in the weather. As the proportion of these types of energies increases steadily, adequate controls will become necessary to stabilize the grid. Hydrogen generation systems that employ electrolysis

techniques are expected to be used as large-capacity power storage facilities with the ability to stabilize the electrical power supply. In particular, high-temperature steam electrolysis performed using solid oxide electrolysis cells (SOECs) can generate hydrogen with high efficiency [1–3], because the heat from the overpotential can be recycled in the form of a heat source for the electrolysis reaction. In addition, owing to the endothermic nature of the reaction, increases in the temperature of the SOEC are prevented. However,

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<http://dx.doi.org/10.1016/j.ijhydene.2016.05.161>

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understanding temperature distributions of the microtubular SOEC are essential to design the cell that have high durability and reliability while the measurements of electrolysis performances of SOECs were reported [4–6]. M. Laguna-Bercero et al. [4] measured electrochemical performance of SOECs with scandia stabilized zirconia. S. D. Ebbesen et al. [5] and C. Stoots et al. [6] measured the long term hydrogen production performance by the electrolysis using SOECs.

Although the temperature distributions in planar SOECs have been investigated previously [7–11], there have been few reports on the temperature distributions in microtubular SOECs. The temperature distributions in microtubular SOECs are probably more complex than those in planar SOECs, since nonuniform reaction distributions cannot be predicted easily, owing to the difficulties associated with current collecting, in determining the concentration and flow direction of the reaction gases, and with selecting the proper insulation method. Further, investigating these distributions by experimental methods alone is difficult, given the thinness of the microtubular SOEC components and their extremely high operating temperatures (~900–1200 K). Therefore, in order to understand these complicated phenomena, both experimental and numerical analyses are necessary.

A number of numerical models have been developed for solid oxide fuel cells (SOFCs), including a two-dimensional microporous model of a planar SOFC [8–14]. Because of the similarity of the phenomena that occur in SOFCs and SOECs, these reports serve as useful references for understanding the internal states of SOECs. However, the effects of the entropy changes are different for the reactions involved in electric power generation and electrolysis. In the case of electric power generation, an entropy change results in heat generation, whereas for electrolysis, it results in heat absorption. Therefore, the thermal-transport phenomena in SOECs will be more complex than those in SOFCs. As a result, SOFC models cannot be used directly to simulate SOECs.

Several models of planar SOECs have been described. Ni et al. [1,15] developed a one-dimensional SOEC model to investigate the effects of the reactant concentration and microporous design on the electrochemical performance of SOECs. However, this model did not account for the heat transport phenomena. Njdozefon et al. [16] determined the current–temperature characteristics of an SOEC experimentally, in addition to performing one-dimensional calculations. A comparison of the experimental and numerical results suggested that temperature considerations were important when determining the electrochemical performance. Two- and three-dimensional thermal models that take into account the temperature and reactive current distributions and the reactant concentrations have also been reported [7–11]. Herring et al. [7] calculated the temperature and current density profiles of a single planar SOEC using the three-dimensional FLUENT model and measured the potential–current characteristics as well as the local temperatures of a 10-cell planar solid oxide electrolysis stack. On the basis of the obtained computational and experimental results, they discussed the effects of heat transfer, reaction cooling/heating, and changes in the local gas composition. Ni [8] reported a two-dimensional numerical model for coelectrolysis performed using a planar SOEC and discussed the effects of the inlet gas

composition and inlet temperatures. Udagawa et al. [9,10] described steady and unsteady states one-dimensional numerical models of a cathode (nickel + yttria stabilized zirconia: Ni + YSZ) supported intermediate-temperature SOEC stack and calculated the electrochemical performance of the device at various current densities and temperatures. Hikosaka et al. [11] predicted the electrochemical performance of a planar SOEC using numerical modeling and reported the energy efficiency of hydrogen generation using a model SOEC system. In terms of electrochemical performance, the average current density–potential characteristics of isothermal and nonisothermal models are nearly the same under the assumed conditions. Further, the energy efficiency during electrolysis can be as high as 122% when the heat required is supplied from outside the system (for example, by sunlight) [11]. However, these planar SOEC models cannot be used directly to simulate tubular SOECs because the current collectors in these models are placed across the cells [7–11]. Although tubular SOECs exhibit high thermal stress resistance because of their cylindrical form, they also exhibit high electronic resistance along the axis direction because of asymmetrical current collection. The asymmetrical positions of the current collectors and the large electronic resistance along the axis direction cause the temperature and reactive current distributions to be nonuniform. In addition, in this study, we often found that the microtubular SOEC broke during high-temperature steam electrolysis and that the breaking point was located in the non-active area, as shown in Fig. 1(b). If the breakage was caused by a change in the temperature or a temperature gradient, a numerical analysis of the microtubular SOEC that included only the active area would be inadequate for evaluating the effect on mechanical strength.

In this study, a numerical model of a cathode supported microtubular SOEC was developed in order to elucidate the temperature distribution within these devices. The model takes into account heat, mass, and charge transport and the related chemical reactions. The calculated average current density–voltage (i – V) characteristics, average current density–change in temperature (i – ΔT) characteristics, and the temperature distributions at the surface of the SOEC were compared with the measured ones. The calculated i – V curves, i – ΔT curves, and temperature distributions were in good agreement with those obtained experimentally. The calculated temperature distribution indicated that the temperature distribution in microtubular SOECs is nonuniform and that the positions of the current collectors strongly affect the temperature and reaction distributions.

Numerical model

Fig. 1 shows a cathode supported microtubular SOEC. The microporous cathode consisted of a mixture of nickel and $(ZrO_2)_{0.92}(Y_2O_3)_{0.08}$ (YSZ); the electrolyte is YSZ. The microporous anode was made of a mixture of $La_{0.8}Sr_{0.2}MnO_3$ (LSM) and YSZ. The constitution and shape of the cell are shown in Fig. 1(a). The cell length was approximately 50 mm, with the outer diameter being approximately 2 mm.

The overpotential and temperature change characteristics of the microtubular SOEC were determined as functions of the

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