



Dynamic characteristics of a solid oxide fuel cell with direct internal reforming of methane



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ABSTRACT

An analysis of a solid oxide fuel cell (SOFC) working with direct internal reforming of methane when subjected to unsteady conditions is presented in this paper. A cell of planar type with the anode-supported structure and the co-flow configuration is investigated. Owing to the reforming and the water–gas shift reactions, it has been known that the steady operation of a cell working with natural gas is different from that with hydrogen. Moreover, since these chemical reactions are strongly temperature-dependent, the former is expected to become much more complex than the latter under unsteady states. It is necessary in the viewpoints of degradation and performance efficiency to understand the evolution of the thermal gradient/stress, the fuel concentration, etc. when the cell is subjected to changes such as load demand. In this work, a three-dimensional numerical model is employed and step changes of the output voltage are introduced to the cell. Results for the temporal profiles of the temperature, the current density, the activation overpotential and the gas concentration distributions in the cell are presented and discussed. It is found that the overshoot in the activation does not appear throughout the cell, but only in the exit region when the voltage jumps from 0.7 V to 0.6 V.

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1. Introduction

Solid oxide fuel cells (SOFCs) convert chemical energy into electrical energy via an electrochemical reaction at high temperatures (500–1000 °C) with high efficiency, low pollution and low noise. A typical cell consists of fuel and gas channels, porous anode and cathode electrodes, a dense electrolyte and electrical interconnects. SOFCs become more energy efficient when operating in a co-generation power system owing to the extra heat produced within the cells. Such a system may contain a heat exchanger, a reformer, a gas turbine, etc. in addition to an SOFC stack (see e.g. [1,2]); and any change in the operation of one of them would likely put the whole system into an unsteady state. Indeed, in industrial and/or residential applications, demand change may happen daily or even hourly. Therefore, it is essential, in the viewpoints of system control, performance optimization and degradation remediation, to understand the dynamic behaviors of SOFCs.

Experimental investigations of SOFC operation under unsteady conditions are especially difficult due to the fact that it is difficult to get access inside the cell/stack without affecting its performance.

In this context, a numerical approach can be more favorable. Indeed, there have been a considerable number of studies using numerical models over the last two decades. The models are in a wide range of detail level in terms of the number of dimensions considered: they can be lumped and/or cell-level [3,4], one-dimensional [5,6], two-dimensional [7,8], and three-dimensional [9–12]. The lumped models considering the entire stack as a “black box” can help in control of SOFCs operating dynamically together with other components in a system. Other models consider spatial variations of temperature, chemical species, etc. along the length (1D), the thickness (2D) and the width (3D) of the cell/stack. Using a 3D model, Achenbach [9] investigated the transient time of the cell voltage and the solid temperature of a planar cell when subjected to current jumps. The cell had a design of cross-flow configuration and operated with internal steam reforming of methane. Both ceramic and metallic types of interconnects were studied. The transient time of the cell voltage was found to be ~300 s and ~700 s for the ceramic and metallic interconnects, respectively. Moreover, as only the maximum and minimum solid temperatures were reported, it was ~300 s for the former and about 2.5 times larger for the latter for the cell made of the ceramic interconnects. In our previous work, the transient time was found to be in a range from 450 to 650 s for different variables such as the activation, the current density and the temperature when a planar unit cell

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Nomenclature

A_{ac}	active area per unit volume, m^{-1}
\bar{c}	constant-volume specific heat, J kg^{-1}
C	charge concentration, C m^{-3}
d	particle diameter, m
D	diffusion coefficient, $\text{m}^2 \text{s}^{-1}$
e	specific internal energy, $\text{J kg}^{-1} \text{K}^{-1}$
E	potential, V
F	Faraday constant, $96,487 \text{ C mol}^{-1}$
$F_{m,j}$	diffusive mass flux of species m , $\text{kg m}^{-2} \text{s}^{-1}$
$F_{h,j}$	diffusive energy flux component, $\text{J m}^{-2} \text{s}^{-1}$
i	current density, $\text{C m}^{-2} \text{s}^{-1}$
i_0	exchange current density, $\text{C m}^{-2} \text{s}^{-1}$
j	transfer current per unit volume, $\text{C m}^{-3} \text{s}^{-1}$
k	thermal conductivity, $\text{W m}^{-1} \text{K}^{-1}$
$k_{0,a}, k_{0,c}$	reaction rate coefficient, $\text{mole m}^{-3} \text{s}^{-1} \text{V}^{-1}$
K_i	permeability of porous media, $\text{kg m}^{-3} \text{s}^{-1}$
p	pressure, Pa
r	reaction rate, $\text{mole m}^{-3} \text{s}^{-1}$
R	universal constant of gas, $8,314 \text{ J mol}^{-1} \text{K}^{-1}$
R_e	electrochemical reaction rate, $\text{mole m}^{-3} \text{s}^{-1}$
Q_c	chemical heat source, $\text{J m}^{-3} \text{s}^{-1}$
Q_e	electrochemical heat source, $\text{J m}^{-3} \text{s}^{-1}$
Q_{Ohm}	Joule heating, $\text{J m}^{-3} \text{s}^{-1}$
s_i	momentum source component, $\text{kg m}^{-2} \text{s}^{-2}$
s_m	production/consumption rate of species m , $\text{kg m}^{-3} \text{s}^{-1}$
s_Σ	total mass source, $\text{kg m}^{-3} \text{s}^{-1}$
T	temperature, K

u_i	velocity component, m s^{-1}
U_i	superficial velocity component, m s^{-1}
x_i	Cartesian coordinate, m
Y_m	mass fraction of component m

Greek letters

β	symmetry factor, 0.5
ϵ	porosity
η	overpotential, V
μ	dynamic viscosity of gas, $\text{kg m}^{-1} \text{s}^{-1}$
Φ	potential, V
ρ	gas mixture density, kg m^{-3}
τ	tortuosity
τ_{ij}	stress tensor, Pa
σ	ionic/electric conductivity, $\Omega^{-1} \text{m}^{-1}$

Subscripts

m, n	species index
i, j, k	Cartesian directions

Abbreviations

ADL	anode diffusion layer
ACL	anode catalyst layer
CDL	cathode diffusion layer
CCL	cathode catalyst layer
SOFC	solid oxide fuel cell

was subjected to step changes in the operating voltage [12]. It is noteworthy that the transient time depends strongly on the materials, the operating parameters, the geometry and/or the dimensions of the cell/stack. It can range from 20 s for a micro-tubular SOFC [7] to about 17–18 min for a thirty-planar cell stack [11].

Owing to the high operating temperatures, SOFCs can be fed with hydrocarbons, methanol, bio-ethanol, biogas, etc. in addition to hydrogen (see e.g. [2,13,14]). The performance of SOFCs fed with these fuels are different from one another because of differences in their chemical and/or electrochemical reactivity. When working with hydrocarbon fuel such as methane or natural gas, an internal reforming process, with steam for instance, is required. This endothermic process consumes a large amount of heat within a small area near the fuel entrance due to its high reactivity, causing the cells to bear a great thermal gradient/stress. How it evolves during a transient state is an interesting question; and answering this is therefore one of the main objectives of the present work. In most of the aforementioned studies the fuel used was hydrogen (except [9]) and only cell-averaged quantities such as temperature were reported. Temporal lengthwise profiles of temperature, current density, activation overpotential, etc. were presented in our previous report [12].

In this paper, we aim to look at local evolution of these quantities and thus report their temporal profiles in all three spatial directions. The cross-sectional profiles would make it possible to analyze effects of the interconnect ribs on the transients. This is indispensably important as fuel depleted areas can be detected, hence preventing the cell/stack from failure. We use the same model as presented in [12], which was adopted from a version built for steady operations [15,16]; however, some special treatment in terms of simulation techniques would probably be required as now the system consists of chemical reactions besides other processes. Here we highlight some of the main features of the model (detailed

information about it should be referred to the mentioned references):

- It uses a single computational domain;
- Computational grid is generated in every component including the interconnect ribs and the thin electrolyte;
- Processes such as fluid flow, heat and mass transport, chemical and electrochemical reactions are taken into account.

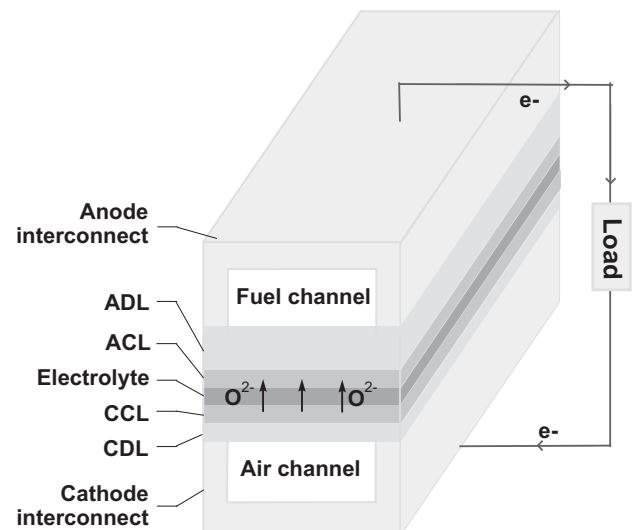


Fig. 1. Schematic diagram of a unit cell-modeling domain.

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