



## Key characteristics of a hydrocarbon-fueled solid oxide fuel cell examined by local thermodynamic states

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### ABSTRACT

Key features of hydrocarbon-fueled solid oxide fuel cell stack operation are elucidated by examining its local thermodynamic states with an aid of three-dimensional numerical simulations. A high-fidelity physical model, which resolves the coupling between thermo-chemical reactions and heat and mass transport, is developed and validated. To elucidate important reactions and transport phenomena, local thermodynamic state variables of hydrocarbon-fueled operation are compared with those estimated by assuming pure-hydrogen-supplied operation. Results show that thermochemical reactions proceed at high rates through the thick anode support layer. This induces complete methane conversion as soon as it is introduced to the anode and thermochemical reaction zones concentrated in the vicinity of the fuel inlet. In spite of the fast reaction processes, hydrocarbon-fueled operation has the same electrical current density profile as pure-hydrogen-supplied operation, resulting from changing its local thermodynamic states. Given that the presence of carbon substances and thermochemical reactions, in hydrocarbon-fueled operation, local chemical and electrical conditions are substantially different from those of pure-hydrogen-supplied operation. A lower hydrogen concentration induces a higher concentration overpotential and decreases a reversible electrochemical potential. A lower exchange current density is offset by increasing an activation overpotential at a given applied current. All these reduce the overall cell voltage, as compared to pure-hydrogen-supplied operation. Variation of transport properties such as diffusivities and viscosities influences heat and mass transport such that substantial stresses can be imposed on cell materials. In addition, thermal conditions result in lower incoming-gas heating and a larger heat transfer rate to a neighboring repeating unit. A larger temperature gradient near the fuel inlet may also impose stresses cell materials. A lower power output, attributed to the electrochemical losses in a form of activation and concentration overpotentials, and materials degradation can be accompanied in hydrocarbon-fueled stack operation.

### 1. Introduction

Solid oxide fuel cell (SOFC) has been studied for a variety of applications such as a vehicle range extender, residential and commercial building power supply, and a distributed (de-centralized) power plant [1,2]. For the past decades, SOFC has been extensively investigated as an advanced power generation technology given its high energy conversion efficiency, power density and fuel flexibility [3,4]. Such advantages make it promising to apply SOFC to mobile power units as well as stationary power systems. Through the Ene-farm project in Japan and Ene-field project in Europe, SOFC has been demonstrated for combined heat and power (CHP) system that not only produces

electrical power, but also recuperates exhaust heat for district heating [5,6]. The renewable portfolio standard (RPS) program in South Korea provides substantial incentives to SOFC technologies, which promotes its installation for distributed power supply [7]. Lately, its application to vehicles has been actively investigated to provide a cruising range similar to gasoline-powered cars [8]. All these applications use natural gas or liquid bio-fuel, attributed to the fact that SOFC can utilize directly hydrocarbon fuels without employing expensive reforming processes.

The fuel flexibility of SOFC is one of its key strengths that makes it more promising and enables easier deployment in the energy market. Its capability to utilize directly hydrocarbon fuels simplifies system

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Nomenclature	
$E$	activation energy [ $\text{J}\cdot\text{mol}^{-1}$ ]
$D_{ij}$	binary diffusivity between species $i$ and $j$ [ $\text{m}^2\cdot\text{s}^{-1}$ ]
$\dot{Q}$	charge-carrier source [ $\text{A}\cdot\text{m}^{-3}$ ]
$\sigma$	charged-species conductivity [ $\text{S}\cdot\text{m}^{-1}$ ]
$i$	current density [ $\text{A}\cdot\text{m}^{-2}$ ]
$\vec{F}_{Da}$	Darcy's friction force [ $\text{N}\cdot\text{m}^{-3}$ ]
$\rho$	density [ $\text{kg}\cdot\text{m}^{-3}$ ]
$\Sigma_i$	diffusion volume of species $i$ [ $\text{s}^{4.5}\cdot\text{K}^{2.625}\cdot\text{mol}^{0.75}\cdot\text{kg}^{-2.25}\cdot\text{m}^{-1.5}$ ]
$D_i$	diffusivity of species $i$ [ $\text{m}^2\cdot\text{s}^{-1}$ ]
$\mu_i$	dynamic viscosity of species $i$ [ $\text{kg}\cdot\text{m}^{-1}\cdot\text{s}^{-1}$ ]
$\phi$	electric potential [V]
$\Delta S^0$	entropy change of reactions [ $\text{J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1}$ ]
$F$	Faraday constant [ $\text{C}\cdot\text{mol}^{-1}$ ]
$R$	gas constant [ $\text{J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1}$ ]
$\dot{H}$	heat source [ $\text{W}\cdot\text{m}^{-3}$ ]
$h$	height [m]
$D_{Kn,i}$	Knudsen diffusivity of species $i$ [ $\text{m}^2\cdot\text{s}^{-1}$ ]
$L$	length [m]
$\omega_i$	mass fraction of species $i$ [-]
$\dot{R}_i$	mass production rate of species $i$ [ $\text{kg}\cdot\text{m}^{-3}\cdot\text{s}^{-1}$ ]
$\dot{W}$	mass source [ $\text{kg}\cdot\text{m}^{-3}\cdot\text{s}^{-1}$ ]
$D_{mix,i}$	mixture averaged diffusivity of species $i$ [ $\text{m}^2\cdot\text{s}^{-1}$ ]
$x_i$	molar fraction of species $i$ [-]
$M_i$	molecular weight of species $i$ [ $\text{kg}\cdot\text{mol}^{-1}$ ]
$\eta$	overpotential [V]
$\zeta$	permeability [ $\text{m}^2$ ]
$r_{pore}$	pore size of the porous media [m]
$\varepsilon$	porosity [-]
$A$	pre-exponential factor [ $\text{S}\cdot\text{K}\cdot\text{m}^{-1}$ ]
$S$	reaction site area per unit volume [ $\text{m}^{-1}$ ]
$C_{p,i}$	specific heat capacity of species $i$ [ $\text{J}\cdot\text{kg}^{-1}\cdot\text{K}^{-1}$ ]
$\lambda_{solid}$	thermal conductivity of solid materials [ $\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ ]
$\lambda_i$	thermal conductivity of species $i$ [ $\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ ]
$t$	thickness [m]
$\tau$	tortuosity [-]
$\vec{u}$	velocity [ $\text{m}\cdot\text{s}^{-1}$ ]
$\theta_{E.C.}$	volume fraction of electronic conductor in the composite [-]
$\theta_{P.F.}$	volume fraction of the pore-former in the anode support layers [-]
<b>Acronyms</b>	
AFL	anode functional layer
ASL	anode support layer
CCCL	cathode current collecting layer
CFL	cathode functional layer
CCL	current collecting layer
EL	electrolyte
CH	gas channel
IC	interconnects
<b>Subscripts</b>	
act	activation
a	anode
c	cathode
elec	electronic
ion	ionic
rev	reversible
<b>Superscripts</b>	
eff	effective
0	standard

configuration and enhances techno-economic competency [3,4,9,10]. Other fuel cell technologies such as proton exchange membrane, alkaline and phosphoric acid fuel cells require high-purity hydrogen as a fuel, which imposes large demand on external reforming and gas filtering. In contrast, SOFC can be fed directly with hydrocarbon fuels, decreasing such substantial parasitic losses. Given high operating temperature and catalysts embedded in the anode, hydrocarbon fuels can be internally reformed to produce hydrogen [11]. Internal production and supply of hydrogen, which is the main reactant for electrochemical reactions, facilitates the production of electrical power. In addition, ceramic materials used in the electrolyte and anode of SOFC provide higher tolerance to impurities that may come along with a hydrocarbon fuel stream [12]. Based on these inherent strengths, the direct use of hydrocarbon fuels in SOFC may lower its entry barriers into the energy market by utilizing current hydrocarbon infrastructure. This also reduces operation costs and enhances the cost competitiveness of SOFC [4,9]. Direct utilization of hydrocarbon fuels is essential to take advantage of SOFC's technical strengths and improve its economic competency.

Although it is highly beneficial to use hydrocarbon fuels in SOFC stacks, there exists a number of technical issues that need to be resolved prior to their market deployment. Hydrocarbon-fueled SOFC stacks have been shown to exhibit a lower power density than high-purity hydrogen-fueled operation [10]. In some cases, their performance was even largely degraded due to large thermo-mechanical stresses imposed on cell materials [10]. Note that the performance and reliability of SOFC stacks are predominantly determined by local conditions around/inside cells [13,14]. To overcome the technical issues of hydrocarbon-fueled SOFC stacks, it is critical to identify the local reacting

environment and primary sources for such change in performance and reliability. However, the coupling between thermochemistry, electrochemistry and heat and mass transport makes it significantly difficult to predict the internal reacting environment [3,9,10]. Thermochemical and electrochemical reactions proceed simultaneously and are inter-related through reactants and products. The presence of multiple species including carbon-containing substances in addition to hydrogen- and oxygen-containing species results in complex heat and mass transport processes. Such reaction and transport processes are interconnected through the local thermodynamic state, which makes the anode reacting environment substantially complex. Therefore, the local thermodynamic states inside hydrocarbon-fueled SOFC stacks should be resolved with sufficient details and elucidated their relationship with performance and reliability. Yet it is very difficult to measure experimentally the local thermodynamic state due to sealing condition and high temperature operation. In this regard, numerical simulations by using high-fidelity models need to be performed, which has not been accounted for in detail.

Previous studies have considered simplified geometry and reduced-order models and not elucidated the primary sources for different reacting environment of hydrocarbon-fueled SOFC stack, as summarized in Table 1. Evely and Yan et al. introduced two-dimensional models and investigated the effect of anode exhaust recycling and porosity variation on carbon deposition [12,15]. Their study focused primarily on the prediction of potential carbon deposition and suggested methodologies to overcome carbon coking. In spite of their contribution, their analysis did not examine the local thermodynamic state nor its coupling with thermochemical phenomena occurring inside the cell. Janardhanan et al. conducted numerical simulations employing

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