Electrochimica Acta 146 (2014) 151–163

Contents lists available at ScienceDirect

Electrochimica Acta

journal homepage: www.elsevier.com/locate/electacta

Electrochemical Simulation of Planar Solid Oxide Fuel Cells with Detailed Microstructural Modeling

A. Bertei [a,*], J. Mertens [b], C. Nicolella [a]

[a] *Department of Civil and Industrial Engineering, University of Pisa, Largo Lucio Lazzarino 2, 56126 Pisa, Italy*
[b] *Institute of Energy and Climate Research (IEK-9), Forschungszentrum Jülich, Ostring O10, 52425 Jülich, Germany*

ARTICLE INFO

Article history:
Received 6 December 2013
Received in revised form 9 July 2014
Accepted 29 August 2014
Available online 18 September 2014

Keywords:
Solid oxide fuel cell (SOFC)
Cell-level modeling
Numerical reconstruction
Electrode microstructure
Anode-supported Jülich cell.

ABSTRACT

A quasi-two-dimensional physically-based model for the description of transport and reaction in planar solid oxide fuel cells (SOFC) is presented in this study. Electrochemistry and transport phenomena in the cell are locally described in 2D using mass conservation equations and well-established global electro-kinetics, coupled with the 1D representation of gas channels in both co-flow and counter-flow configurations. The key feature of the model consists in the numerical reconstruction, through packing algorithms, of the three-dimensional microstructure of each porous layer for an accurate evaluation of the effective properties. Coupling of a detailed microstructural modeling into the cell-level electrochemical model allows the prediction of the polarization behavior from the knowledge of operating conditions and powder characteristics, thus eliminating the need for empirical correlations and adjusted parameters, which is typically the weak point of existing cell-level models. The framework is used for the simulation of a short stack of anode-supported cells with LSM-based cathode and 1.5 mm thick anode support, developed and tested by Forschungszentrum Jülich. The effective properties of each layer are calculated and compared with available experimental data. A good agreement is also reached when comparing simulated and experimental polarization curves under different operating conditions without fitting any parameter. Simulations show that at 800 °C the activation resistance in the cathode functional layer is the main contribution to the cell overpotential. In addition, the model suggests that gas concentration effects at the anode play an important role on the global electrochemical response. The study shows that quantitative predictions can be obtained using this integrated approach, making it an attractive tool to assist the SOFC development.

1. Introduction

Solid oxide fuel cells (SOFCs) are energy conversion devices which offer clear advantages if compared with conventional power systems, such as low emission of pollutants [1], high efficiency [2] and fuel flexibility [3], making them attractive for both stationary and mobile applications [4]. Recent advances in manufacturing techniques and improvements in cell lifetime have led this technology close to be launched on the market [5–7].

SOFC development is currently assisted by modeling and simulation techniques [8], which help researchers understand the complex interactions between electrochemical phenomena and material properties over different length scales [9]. In particular, many efforts have been done to model the electrochemical behavior of cells within a stack [10,11]. Cell-level models describe the chemical and electrochemical phenomena occurring within an SOFC in order to relate the global cell response (e.g., the cell current-voltage characteristic) to the microscopic processes occurring within the cell components, such as the transport of species within the electrodes and the local kinetic electrochemical phenomena. This allows researchers to interpret the experimental results and to predict the system performance.

Several cell-level models have been developed in the past. Aguiar et al. [12] proposed a dynamic model for anode-supported SOFCs, consisting of mass and energy balances applied to the fuel and air channels. Zhu et al. [11] and Janardhanan and Deutschmann [13] adopted a continuum approach based on local balance equations in the membrane-electrode assembly, focusing on the elementary kinetics of internal reforming. A similar approach was adopted by Bessler et al. [14], who presented a physically-based modeling framework for the description of kinetics and

---

Abbreviations: LSM, strontium-doped lanthanum manganite; SOFC, solid oxide fuel cell; TPB, three-phase boundary; YSZ, yttria-stabilized zirconia.

* Corresponding author. Tel.: +39-0502217814; fax: +39-0502217866.
E-mail addresses: antonio.bertei@for.unipi.it, berteiantonio@gmail.com (A. Bertei).

http://dx.doi.org/10.1016/j.electacta.2014.08.120

Notation

A_{des}	pre-exponential factor of hydrogen desorption kinetics (s·m^2·mol^{-1})
B^{la}	permeability in the layer *la* (*la* = *af*, *as*, *cf*, *cc*) (m^2)
d_p^{la}	mean particle size of phase *p* (*p* = *ela*, *elc*, *io*) in the layer *la* (*la* = *af*, *as*, *cf*, *cc*) (m)
d_{pore}^{la}	mean pore size in the layer *la* (*la* = *af*, *as*, *cf*, *cc*) (m)
$D_{K,s}^{eff,la}$	effective Knudsen diffusivity of species *s* (*s* = *H2*, *H2O*, *O2*, *N2*) in the layer *la* (*la* = *af*, *as*, *cf*, *cc*) (m^2·s^{-1})
$D_{s1/s2}$	binary diffusivity of the pair of species *s1/s2* (*s1/s2* = *H2/H2O*, *O2/N2*) (m^2·s^{-1})
$D_{s1/s2}^{eff,la}$	effective binary diffusivity of the pair of species *s1/s2* (*s1/s2* = *H2/H2O*, *O2/N2*) in the layer *la* (*la* = *af*, *as*, *cf*, *cc*) (m^2·s^{-1})
E°	standard electromotive force (V)
E_{des}	activation energy of hydrogen desorption kinetics (J·mol^{-1})
E_j^{act}	activation energy (*j* = *a*, *c*) (J·mol^{-1})
f_j	friction factor (*j* = *a*, *c*) (-)
F	Faraday constant (C·mol^{-1})
F_{air}	molar flow rate of air (mol·s^{-1})
F_{fuel}	molar flow rate of fuel (mol·s^{-1})
h_j	channel height (*j* = *a*, *c*) (m)
i_j	current density per unit of TPB length (*j* = *a*, *c*) (A·m^{-1})
i_{j0}	exchange current per unit of TPB length (*j* = *a*, *c*) (A·m^{-1})
i_{j0}^{ref}	exchange current per unit of TPB length at the reference temperature (*j* = *a*, *c*) (A·m^{-1})
I	current density per unit of cell area (A·m^{-2})
$\underline{J}_s$	molar flux of species *s* (*s* = *H2*, *H2O*, *O2*, *N2*, *e*, *O*) (mol·m^{-2}·s^{-1})
$\tilde{\underline{J}}_s$	mass flux of species *s* (*s* = *H2*, *H2O*, *O2*, *N2*, *e*, *O*) (kg·m^{-2}·s^{-1})
J_s^{cell}	molar flux of species *s* (*s* = *H2*, *H2O*, *O2*, *N2*) at the cell-channel interface (mol·m^{-2}·s^{-1})
L	cell length (m)
M_s	molecular weight of species *s* (*s* = *H2*, *H2O*, *O2*, *N2*) (kg·mol^{-1})
$\underline{n}$	normal unitary vector oriented from cell to channel (-)
OCV	open circuit voltage (V)
P_j	pressure (*j* = *a*, *c*) (Pa)
P_{H2}^{*}	reference pressure of hydrogen adsorption/desorption equilibrium (Pa)
P_{O2}^{ref}	reference oxygen partial pressure (Pa)
Pe_j	Peclet number (*j* = *a*, *c*) (-)
q_s	charge of species *s* (*s* = *e*, *O*) (-)
r_j	interconnect rib width (*j* = *a*, *c*) (m)
R	ideal gas constant (J·mol^{-1}·K^{-1})
Re_j	Reynolds number (*j* = *a*, *c*) (-)
t^{la}	thickness of the layer *la* (*la* = *af*, *as*, *cf*, *cc*, *ey*) (m)
T	operating temperature (K)
T_j^{ref}	reference temperature for reaction kinetics (*j* = *a*, *c*) (K)
v_j	mass-averaged gas velocity (*j* = *a*, *c*) (m·s^{-1})
V_{cell}	cell potential (V)
V_p	electric potential of phase *p* (*p* = *ela*, *elc*, *io*) (V)
w_j	channel width (*j* = *a*, *c*) (m)
x	coordinate along channel longitudinal direction (m)
y_s	molar fraction of gas species *s* (*s* = *H2*, *H2O*, *O2*, *N2*) (-)
$\tilde{y}_s$	mass fraction of gas species *s* (*s* = *H2*, *H2O*, *O2*, *N2*) (-)
z	coordinate along cell thickness direction (m)

Superscripts

af	anode functional layer
as	anode supporting layer
cc	cathode current collector
cf	cathode functional layer
ey	electrolyte

Subscripts

a	anodic
c	cathodic
e	electron
ela	electron-conducting phase in the anode (e.g., Ni)
elc	electron-conducting phase in the cathode (e.g., LSM)
H2	hydrogen
H2O	water
io	ion-conducting phase (e.g., YSZ)
IN	inlet conditions
N2	nitrogen
O	oxygen ions
O2	oxygen

Greek Letters

α_j	first transfer coefficient (*j* = *a*, *c*) (-)
β_j	second transfer coefficient (*j* = *a*, *c*) (-)
γ_0	sticking probability of hydrogen adsorption kinetics (-)
Γ	surface site density (mol·m^{-2})
δ_j	dimensionless factor of Taylor-Aris axial dispersion (*j* = *a*, *c*) (-)
$\Delta V_{eq,j}$	local equilibrium potential difference (*j* = *a*, *c*) (V)
ζ	reaction order of oxygen in cathode kinetics (-)
η_{cell}	cell overpotential (V)
η_j	activation overpotential (*j* = *a*, *c*) (V)
η_u	overpotential of compartment *u* (*u* = anode, cathode, electrolyte) (V)
θ^{la}	maximum angle of contact among the particles in the layer *la* (*la* = *af*, *as*, *cf*, *cc*) (°)
$\kappa_p^{eff,la}$	effective conductivity factor of phase *p* (*p* = *ela*, *elc*, *io*) in the layer *la* (*la* = *af*, *as*, *cf*, *cc*, *ey*) (-)
λ_{TPB}^{la}	TPB length per unit volume in the layer *la* (*la* = *af*, *as*, *cf*, *cc*) (m^{-2})
μ_j	gas dynamic viscosity (*j* = *a*, *c*) (kg·m^{-1}·s^{-1})
ρ_j	gas density (*j* = *a*, *c*) (kg·m^{-3})
σ_p	conductivity of phase *p* (*p* = *ela*, *elc*, *io*) (S·m^{-1})
τ^{la}	tortuosity factor in the layer *la* (*la* = *af*, *as*, *cf*, *cc*) (-)
$\tau_{w,j}$	shear-stress factor at the wall (*j* = *a*, *c*) (N·m^{-2})
υ_s	stoichiometric coefficient of species *s* (*s* = *H2*, *H2O*, *O2*, *N2*, *e*, *O*) (-)
ϕ^{la}	porosity of the layer *la* (*la* = *af*, *as*, *cf*, *cc*) (-)
ψ_{el}	solid volume fraction of electronic phase *el* (*el* = *ela*, *elc*) after sintering (-)