Journal of Power Sources 300 (2015) 365–375

Contents lists available at ScienceDirect

Journal of Power Sources

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

An analytical approach for solid oxide cell electrode geometric design

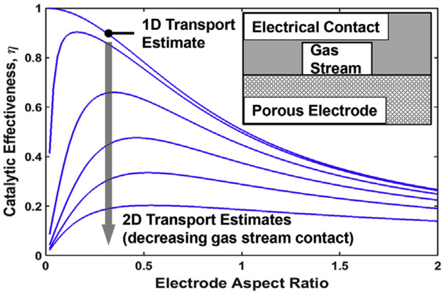
George J. Nelson*

Department of Mechanical and Aerospace Engineering, University of Alabama in Huntsville, 301Sparkman Drive, Huntsville, AL 35811, USA

HIGHLIGHTS

- Dimensionless metrics for solid oxide cell electrode design are presented.
- Geometry sensitive reactant depletion and catalytic effectiveness assessed.
- Analytical models verified with finite element models and published experiment.
- Demonstration that thinner electrodes do not exclusively benefit mass transport.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 13 July 2015
Received in revised form
2 September 2015
Accepted 23 September 2015
Available online 2 October 2015

Keywords:
Solid oxide fuel cell
Analytical model
Electrode design
Mass transport
Catalytic effectiveness
Reactant depletion

ABSTRACT

An analytical model for gas distributions in porous solid oxide cell electrodes is applied to develop dimensionless metrics that describe electrode performance. These metrics include two forms of a dimensionless reactant depletion current density and a geometry sensitive Damköhler number used to assess electrode catalytic effectiveness. The first dimensionless depletion current density defines when reducing electrode thickness no longer benefits mass transfer performance for a given cell geometry. The second dimensionless depletion current density provides a gage of deviation from the limiting current behavior predicted using button-cell experimental and modeling approaches. The Damköhler number and related catalytic effectiveness quantify two-dimensional transport effects under non-depleted operating conditions, providing a means of generalizing insights from reactant depletion behavior for typical cell operating conditions. A finite element solution for gas transport based on the dusty-gas model is used as a benchmark for the analytical model and dimensionless metrics. Estimates of concentration polarization based on analytical and numerical models compare well to published experimental data. Analytical performance predictions provide clear demonstration of the influence of two-dimensional electrode geometry on solid oxide cell performance. These results agree with finite element predictions and suggest that reduction of electrode thickness does not exclusively benefit cell performance.

1. Introduction

Solid oxide cells are energy conversion devices composed of porous composite electrodes that bound a solid ionic conducting electrolyte, as shown in Fig. 1. When operated as solid oxide fuel cells (SOFCs), the electrochemical oxidation of hydrogen and reduction of oxygen within these electrodes produces current. SOFCs directly convert chemical energy to electrical energy, alleviating many of the losses associated with thermal and mechanical energy conversion devices. This direct conversion can yield less

---

* Corresponding author.
E-mail address: george.nelson@uah.edu.

http://dx.doi.org/10.1016/j.jpowsour.2015.09.085
0378-7753/

Nomenclature

C_i	Reactant Concentration, mol m^{-3}
Da	Damköhler Number
D_i	Binary Diffusion Coefficient, m^2 s^{-1}
D_{ik}	Knudsen Diffusion Coefficient, m^2 s^{-1}
F	Faraday Constant, C mol^{-1}
I	Local Current Density, A m^{-2}
i_0	Exchange Current Density, A m^{-2}
L_{UC}	Unit Cell Total Width, m
L_{GS}	Gas Stream Contact Width, m
M	Molar Mass, g mol^{-1}
N_i	Molar Flux, mol m^{-2} s^{-1}
P	Total Pressure, Pa
R_u	Universal Gas Constant, J mol^{-1} K^{-1}
T	Temperature, K
t	Thickness, m
y_i	Mole fraction
z	Electrons Participating in Reaction
ε	Porosity
η	Catalytic Effectiveness
τ	Tortuosity
ϕ	Thiele Modulus

Superscripts

eff	Effective Diffusion Coefficient
0	Gas Stream Value

Subscripts

dep	Value at Reactant Depletion
elec	General Electrode Parameter
H_2	Hydrogen
H_2O	Steam
Lim	Value at Limiting Current
O_2	Oxygen

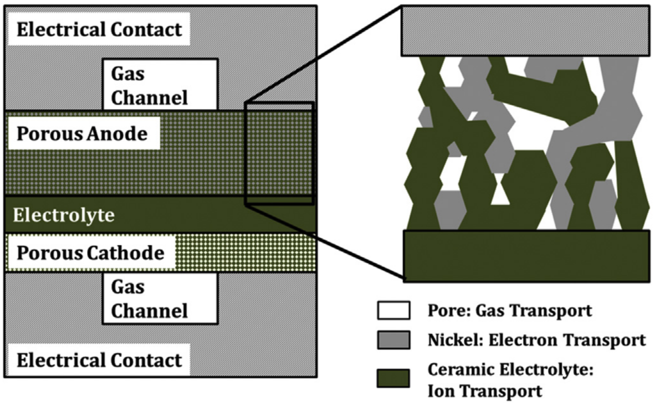


Fig. 1. A solid oxide cell consists of porous electrodes that bound an ion conducting ceramic electrolyte. The proposed research focuses on the porous SOE cathode (SOFC anode).

problematic, sequestration ready emissions streams with fewer components and reduced levels of criteria pollutants when compared to combustion based technologies [1]. Under an applied current these cells act as solid oxide electrolyzers (SOEs), which have been widely investigated for hydrogen production [1–5]. SOEs offer the opportunity for hydrogen production at higher operating temperatures, which result in enhanced reaction kinetics and lower operating voltages. Higher temperature operation results in a reduced Nernst potential, meaning a lower thermodynamic minimum voltage is required for SOE operation when compared to lower temperature electrolysis methods. Finally, higher SOFC and SOE operating temperatures permit the use of an inexpensive Ni catalyst that is tolerant of carbon monoxide. This CO tolerance simplifies the use of hydrocarbon fuels from both renewable and fossil sources. This tolerance also allows SOEs to reduce carbon dioxide [2,6–8], a capability that enables generation of syngas from steam and CO_2 for fuel production [2] and the potential for development of regenerative energy storage systems based on CO_2 storage [7,9]. Such regenerative systems could circumvent challenges presented by hydrogen storage and carbon sequestration.

The composite electrode is a critical component of the solid oxide cell. These electrodes, shown schematically in Fig. 1, combine a solid ion conductor, solid electron conductor, and an open pore phase to deliver charged species and reactant gases to reaction sites throughout the electrode. Achieving optimal performance in these composite solid oxide electrodes presents a multi-scale, multi-physics challenge. Transport of charged and neutral species (i.e., gaseous reactants like H_2, O_2, and H_2O) must be balanced at both the microstructural scale, nanometers to microns [10–16], and the device scale, microns to millimeters scale [17–21]. Across these scales variations in local reactant distributions can contribute to significant losses. At the microstructural scale, charge transport holds strong influence [10,11,13,14], while variation in gas concentration may occur [14]. At the device scale, geometry contributes to both ohmic [18,19,22] and concentration losses [17,21].

Accordingly, the modeling methods applied to solid oxide cell design and analysis span many levels of detail and must balance complexity and computational cost. This task proves difficult when translating microstructural or component level details to a cell or stack level. Finite element analysis (FEA) and models based on the Lattice Boltzmann Method (LBM) can capture details of mass and charge transport within the microstructure [11,14–16,23], but these approaches entail significant requirements for memory [11], computational time [11,14], and due diligence in mesh generation [23]. Detailed computational fluid dynamics (CFD) models also require large numbers of computational cells and related degrees of freedom to accurately capture multi-dimensional variations throughout the SOFC component layers [24]. Finite element models can present similar challenges when attempting to retain proper element aspect ratios in meshes that span multiple length scales within the component layers.

At the cell level, modeling efforts have attempted to include greater detail on transport phenomena within the electrode by developing general expressions for cell concentration and ohmic polarization [25–29]. These efforts have often relied upon a one-dimensional characterization of mass transport phenomena across the electrodes. Device level analyses seeking a more detailed description of transport phenomena at the continuum electrode level typically employ FEA and CFD to ascertain the multidimensional characteristics of SOFC transport phenomena [22,24,30–36]. Spatial variations in reactant composition and charge distributions within solid oxide cell electrodes may be readily estimated with analytical and numerical approaches. The impact of multi-dimensional geometry has been demonstrated as significant in several numerical and analytical studies. Ferguson et al. [31]