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Investigation of the electrochemical active thickness of solid oxide fuel cell anode

Keqing Zheng^a, Li Li^b, Meng Ni^{a,*}

^a Building Energy Research Group, Department of Building and Real Estate, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong, China

^b Ability R&D Energy Research Centre, School of Energy and Environment, City University of Hong Kong, Hong Kong, China

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ABSTRACT

Determination of the electrochemical active thickness (EAT) is of paramount importance for optimizing the solid oxide fuel cell (SOFC) electrode. However, very different EAT values are reported in the previous literatures. This paper aims to systematically study the EAT of SOFC anode numerically. An SOFC model coupling electrochemical reactions with transport of gas, electron and ion is developed. The microstructure features of the electrode are modeled based on the percolation theory and coordinate number theory. Parametric analysis is performed to examine the effects of various operating conditions and microstructures on EAT. Results indicate that EAT increases with decreasing exchange current density (or decreasing TPB length) and increasing effective ionic conductivity. In addition to the numerical simulations, theoretical analysis is conducted including various losses in the electrode, which clearly shows that the EAT highly depends on the ratio of concentration related activation loss $R_{act,con}$ to ohmic loss R_{ohmic} . The theoretical analysis explains very well the different EATs reported in the literature and is different from the common understanding that the EAT is controlled mainly by the ionic conductivity of electrode.

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Introduction

Solid oxide fuel cell (SOFC) has attracted increasing attention in recent years for its potential in clean and efficient power generation. SOFC electrode functions as a provider of the electrochemical reaction sites as well as the transport medium of ion, electron and gas. In operation, electrochemical reactions only occur in the triple phase boundaries (TPBs, as shown in Fig. 1) where reaction gas, ion-conducting phase and electron-conducting phase meet. However, even the TPBs in

SOFC electrode are percolated [1], the electrochemical reactions are believed to occur in a small depth from the electrolyte/electrode (E/E) interface, which is known as the electrochemical active thickness (EAT). Determination of the EAT is important in designing an SOFC electrode with optimum thickness [2]. If the electrode thickness is thinner than EAT, the active TPBs are reduced. If the electrode thickness is much larger than EAT, the concentration loss caused by gas transportation will also degrade SOFC performance. In addition, the accurate determination of the EAT is the prerequisite for optimizing the functionally graded electrode, in which the

* Corresponding author. Tel.: +852 2766 4152; fax: +852 2764 5131.

E-mail addresses: bsmengni@polyu.edu.hk, memni@graduate.hku.hk (M. Ni).
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Nomenclature	
j	electric current density, A/m^2
j_{TPB}	electric current generated in unit TPB length, A/m
j_{ref}	reference exchange current density, A/m
p	partial pressure, pa
p_{atm}	operating pressure, pa
r	particle radius, m
r_p	pore radius, m
n_{el}	particle number of electron conducting particles
x	gas molar fraction
$B_{A/C}$	anode/channel interface
$B_{C/C}$	cathode/channel interface
$B_{A/E}$	anode/electrolyte interface
$B_{C/E}$	cathode/electrolyte interface
$D_{i,k}^{eff}$	effective Knudsen diffusion coefficient of gas species i , m^2/s
$D_{i,l}^{eff}$	effective binary diffusion coefficient of gas species i and l , m^2/s
E_{eq}	equilibrium electric potential difference, V
E_{Nernst}	reversible electrode potential, V
F	Faraday constant, 96,485 C/mol
L_{el-io}	perimeter of contact area between electron and ionic particles, m
L^a	anode thickness
M	molecular mass, kg/mol
N	molar flux of gas species, $mol/(m^2 s)$
P_{erco}	percolation possibility
R_{ohmic}	ohmic loss, V
R_{act}	activation loss, V
R_{con}	concentration loss, V
$R_{act,con}$	concentration related activation loss, V
R	ideal gas constant, 8.314 J/(mol K)
T	operating temperature, K
Z_{el-io}	average ionic particle number around electron particle
V_{cell}	output voltage, V
Greek letters	
η_{local}	overpotential, V
ϕ	electric potential, V
σ	effective conductivity, S/m
σ^0	material intrinsic conductivity, S/m
λ_{TPB}	TPB length in unit volume (m/m^3)
ξ	tortuosity factor for gas diffusion
ν	diffusion volume (m^3/mol)
ψ	volume fraction
Subscripts	
el	electronic conducting phase
io	ionic conducting phase
inlet	inlet conditions
ref	reference conditions
local	local reaction sites
H_2	hydrogen
O_2	oxygen
H_2O	water
y	position y
Superscripts	
a	anode side
c	cathode side

microstructure and material composition are purposely varied in different layers according to their functions: for electrochemical reactions, gas transport, electron conduction or ion conduction.

However, the EAT reported in literatures significantly varies probably due to the different operating conditions, electrode microstructures and material properties used in their studies. Martin Andersson et al. [3] developed a CFD model including fully coupled heat, mass, momentum and charge transport and revealed that the reaction zone that contains 90% of the electrochemical reactions should be 2.4 μm in cathode and 6.2 μm in anode under operating temperature of

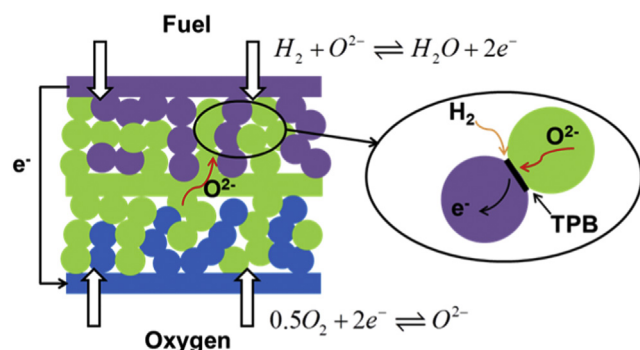


Fig. 1 – Working mechanism of SOFC.

1010 K. For comparison, it is reported by M. M. Hussain et al. [4] that the EAT in anode is about 20 μm when the operating temperature is 1073 K, current density is 0.5 A/m^2 and average particle radius is 0.1 μm . Furthermore, it is revealed in their study that the EAT is increased to about 60 μm when operating temperature is increased to 1273 K. Yoshinori Suzue et al. [5] and Naoki Shikazono et al. [6] solved the governing equations describing gas, ion and electron transport using Lattice Boltzmann Method (LBM) based on their reconstructed Ni/YSZ anode. The positive correlation between EAT (5 μm –20 μm) and operating temperature (873 K–1073 K) is also found in their study. However, the efficient thickness examined by Z.Y. Jiang et al. [7] increases from 36 μm to 126 μm with operating temperature decreases from 1073 K to 923 K, which is contradict to previous studies. It is commonly regarded that the narrow reaction zone is limited by the poor ionic conductivity of electrode, and thus high temperature should generate a large EAT due to the raised high ionic conductivity. The different operating temperature effect on EAT reported by Z.Y. Jiang et al. [7] implies a more complicated underlying relationship between operating parameters and EAT. Besides, it is reported by Naoki Shikazono et al. [6] that a thinner EAT is found with 10% humidified H_2 as fuel, compared with the EAT of about 10–15 μm when 1.2% humidified H_2 is used as fuel at 1273 K. This result also can't be explained by our common knowledge that the EAT is controlled by the electrode ionic conductivity.

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