

Article

The Geometry Effect of Cathode/Anode Areas Ratio on Electrochemical Performance of Button Fuel Cell Using Mixed Conducting Materials

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Abstract: Intermediate temperature (IT) fuel cells using mixed conducting materials have been reported by many researchers by adopting different compositions, microstructures, manufacture processes and testing conditions. Most i_{op} - V_{op} relationships of these button electrochemical devices are experimentally achieved based on anode or cathode surface area (i.e., $A_{an} \neq A_{ca}$). In this paper, a 3D multi-physics model for a typical IT solid oxide fuel cell (SOFC) that carefully considers detail electrochemical reaction, electric leakage, and e^- , ion and gas transporting coupling processes has been developed and verified to study the effect of A_{ca}/A_{an} on button cell i_{op} - V_{op} performance. The result shows that the over zone of the larger electrode can enhance charges and gas transport capacities within a limited scale of only 0.03 cm. The over electrode zone exceed this width would be inactive. Thus, the active zone of button fuel cell is restricted within the smaller electrode area $\min(A_{an}, A_{ca})$ due to the relative large disc radius and thin component layer. For a specified V_{op} , evaluating the responded i_{op} by dividing output current I_{op} with $\min(A_{an}, A_{ca})$ for a larger value is reasonable to present real performance in the current device scale of cm. However, while the geometry of button cells or other electrochemical devices approach the scale less than 100 μm , the effect of over electrode zone on electrochemical performance should not be ignored.

Keywords: electrode areas ratio effect; electrochemical performance; mixed conducting material; multi-physics numerical modeling

1. Introduction

In the past decade, low cost, clean and high efficiency energy conversation and storage devices, such as fuel cells [1], batteries [2] and super-capacitors [3], have been receiving more and more attentions. Solid oxide fuel cell (SOFC) has being recognized as a promising energy conservation device due to its efficiency [4] and capability to work with various fuels [5]. As high operating temperature might cause strict material compatibility constraints [6] and operational complexity [7], attentions have being devoted to the development of intermediate temperature (IT-) SOFC components (i.e., 350–650 °C) [8]. The key obstacles for reducing SOFC operation temperature are attributed to the insufficient activities of conventional cathode materials and low ionic conductivities of traditional electrolyte materials [9] (i.e., YSZ) in this temperature regime. Thus, mixed ion/ e^- conducting (MIEC) electrode materials [10] and alternative electrolyte materials [11] have received great attention for their potential applications in IT-SOFCs. Z. Shao et al. reported a mixed conducting $Ba_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O_{3-\delta}$ as a potential cathode material, which can conduct both electron and O^{2-} charges [10]. Then, an in-situ photoelectron spectroscopy method was proposed to investigate the electrochemically active region

within mixed conducting CeO_{2-x} electrode [12]. S. Wang et al. compared the performances of various LSCF-based cathodes and found that LSCF-SDC exhibited a larger activation overpotential than did the single-phase LSCF cathode [13]. More interestingly, the LSM-coated LSCF composite electrode was reported to exhibit a lower activation overpotential compared with that in a pure LSCF cathode [13]. Furthermore, proton conducting oxides [14], such as $\text{BaZr}_{0.7}\text{Pr}_{0.1}\text{Y}_{0.2}\text{O}_{3-d}$ [15] and $\text{BaZr}_{0.1}\text{Ce}_{0.7}\text{Y}_{0.2}\text{O}_{3-d}$ [16] were also greatly invented to be used in IT-SOFCs because of their low activation energy and high ionic conductivity around IT-range.

Generally, the electrochemical reaction processed within an IT-SOFC using mixed conducting materials or proton conducting oxides are very different from those using conventional composite electrodes [17]. Taking the cathode of LSCF-SDC/SDC/Ni-SDC IT-SOFC using mixed conducting materials as an example [13], the electrochemically active sites not only can be taken placed around the percolated three phase boundary sites (i.e., LSCF-SDC-pores and LSCF-dense electrolyte interfaces), but also can happen around the percolated double phase boundary sites (e.g., LSCF-pore) [18]. Up to now, many IT-SOFC button cells using the mixed conducting materials have been reported by many researchers by adopting different compositions (or materials), volume fractions, microstructure parameters, manufacture processes, operating conditions and different cell geometry sizes. It is interesting to note that different anode and cathode surface areas (i.e., different discs radii) were chosen during button cell fabricating and measuring. For a specified output voltage V_{op} , the corresponding output electric current density i_{op} was always evaluated by dividing output current I_{op} with the relative smaller surface area between anode A_{an} and cathode A_{ca} . This may lead to higher electrochemical performance results based on follow consideration. Generally, the performance of IT-SOFC button cell is a trade-off of electrochemical reacting, gas transporting, e^- and ionic conducting and their mutual coupling processes. The over zone of larger electrode can enhance charges and gases transport capacities within the button cells in a proper zone; and this may affect the $V_{\text{op}}-i_{\text{op}}$ performance measuring result. Thus, it is important to study the sensitivity of the SOFC button cell performance on different cathode and anode surface areas ratios.

3D multi-physics coupling numerical modeling is generally agreed to be an economic, valid and time saving approach for working detail investing [19], parameters-performance studying [20], geometric optimizing [21] and system operation optimizing [22]. In this paper, a 3D multi-physics model which carefully considers detail electrochemical reaction, electric leakage, and e^- , ion and gas transporting coupling processes within a typical IT-SOFC button cell is developed and verified. Then, the influences of different cathode/anode area ratios $A_{\text{ca}}/A_{\text{an}}$ on the button cell $V_{\text{op}}-i_{\text{op}}$ performances are carefully investigated, while different micro-structure parameters, electrode properties, component thicknesses and exchange current densities of reaction interfaces are varied within reasonable value ranges. The study results can help us achieve the valid affecting zone of the relative larger electrode; and assess the rationality that evaluating responded i_{op} by dividing I_{op} with $\min(A_{\text{an}}, A_{\text{ca}})$ for a larger value, while V_{op} is specified. The achieved conclusions would provide good references for understanding the geometric effects of cathode/anode cross sections relationship on electrochemical performance of IT-SOFC button cell and similar electrochemical devices.

2. Method and Theory

Taking a typical anode-supported LSCF-SDC/SDC/Ni-SDC IT-SOFC button cell in Figure 1a as an example, the relevant structure and geometry sizes of the distinct four different cell layers from the experiment report is illustrated. As shown in Figure 1b, the multi-physics working processes within these IT-SOFC button cells are complicated even in hydrogen fuel case. Oxygen within the air should be transported to the percolated LSCF-SDC-pores three phase boundaries (TPBs) or LSCF-pores double phase boundaries (DPBs) in the cathode side. ‘Percolated’ here is defined as a continuous connection through the entire electrode structure. At these places, O_2 will react with the electrons transported by the electronic conducting paths, such as percolated LSCF network and the external current circuit or dense electrolyte that presences electronic conducting capability (e.g., SDC and CGO). The produced

O^{2-} will be conducted to the percolated Ni-SDC-pore TPBs in anode side through O^{2-} conducting network which is constructed by both LSCF- and SDC-particles and dense electrolyte. These O^{2-} will react with the fuels diffused through porous anode. Most of the produced electrons will be circuited back to cathode reaction sites through the external current circuit. But part of the produced electrons will be conducted from anode to cathode side through dense electrolyte directly due to the presence of electronic conducting property of electrolyte material. These electric currents are considered as idle work and cause complex relationships among microstructure parameters, effective electrode properties and multi-physics calculating processes.

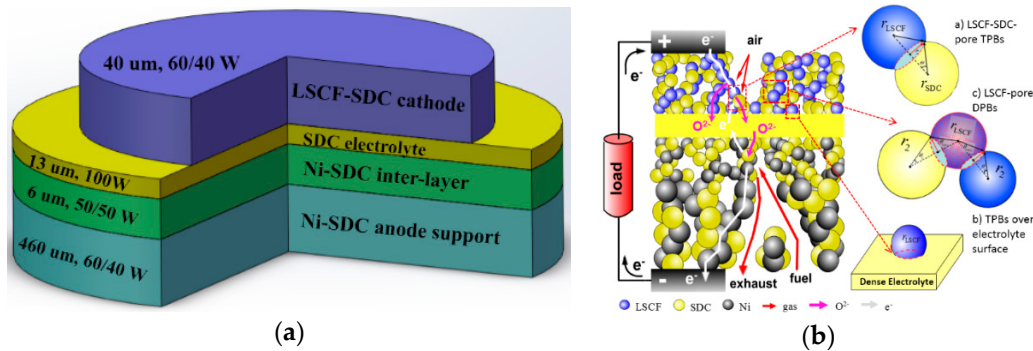


Figure 1. (a) The sketch figure of a typical LSCF-SDC/SDC/Ni-SDC IT-SOFC, (b) the corresponding multi-physics working processes within it.

As proposed in our previous paper [23], the characteristic properties of each SOFC component layer can be evaluated by the generalized percolation micro-model based on the thickness, composition, and microstructure parameters of each component layers. Taking the LSCF-SDC composite cathode as an example, the potential electrochemical active sites consists of the percolated LSCF-SDC-pores TPBs, percolated LSCF-dense electrolyte interfaces and percolated LSCF-pores DPB surface sites (illustrated in Figure 1b).

The percolated LSCF-SDC-pores TPBs per unit volume can be evaluated as

$$\lambda_{TPB, per}^V = \gamma_{LSCF, SDC} n_{LSCF}^V Z_{LSCF, SDC} P_{LSCF}^e P_{SDC}^{O^{2-}} \quad (1)$$

where subscript ‘per’ is used to represent ‘Percolated’. $\gamma_{LSCF, SDC} = \pi r_c^2 (r_c = \min(r_{LSCF}, r_{SDC}) \sin \theta)$ is the electrochemical reaction site per contact between LSCF- and SDC-particles (explained in Figure 2a), $n_k^V = (1 - \phi_g) \psi_k / (4\pi r_k^3 / 3)$ is number of k -particles per unit volume within electrode. P_{LSCF}^e and $P_{SDC}^{O^{2-}}$ are the probabilities of relevant particles belonging to the percolated electron and oxygen ion conducting paths, respectively. Both LSCF- and SDC-particles contribute to the O^{2-} conducting path, thus $P_{SDC}^{O^{2-}} = P_{LSCF}^{O^{2-}} = 1$. The probability of LSCF-particle belonging to percolated e^- conducting network can be estimated by Reference [16]

$$P_{LSCF}^e = 1 - \left(\frac{4.236 - Z_{LSCF, LSCF}}{2.472} \right)^{3.7}$$

$Z_{k, \ell}$ is the number of contacts between k -particle and all of its neighboring ℓ -particles

$$Z_{k, \ell} = 0.5(1 + r_k^2 / r_\ell^2) \bar{Z} \frac{\psi_\ell / r_\ell}{\sum_{k=1}^M \psi_k / r_k}$$

where ψ_k and r_k are the corresponding solid volume fraction and radius of k -particles. ϕ_g is the porosity of composite electrode.

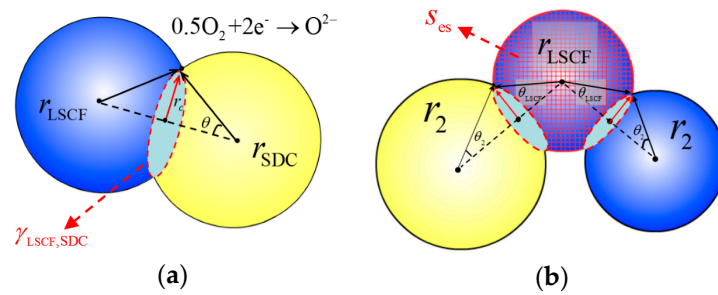


Figure 2. (a) Sketch of electrochemical reaction site per contact between LSCF- and SDC-particles $\gamma_{\text{LSCF,SDC}}$; (b) illustrated of the exposed surface area of per LSCF-particle s_{es} .

The percolated LSCF-pores DPB surface sites per unit volume can be evaluated basing on the cathode microstructure parameters by [23]

$$S_{\text{LSCF, per}}^V = n_{\text{LSCF}}^V s_{\text{es}} P_{\text{LSCF}}^e P_{\text{LSCF}}^{\text{O}^{2-}}, \quad (2)$$

$$s_{\text{es}} = 2\pi r_{\text{LSCF}}^2 [2 - (1 - \cos \theta_{\text{LSCF}}) Z_{\text{LSCF, LSCF}} - (1 - \cos \theta_{\text{LSCF}}) Z_{\text{LSCF, SDC}}]$$

as illustrated in Figure 2b, the exposed surface area of each LSCF-particle s_{es} should be estimated by subtracting the overlap parts of neighboring particles from spherical surface area.

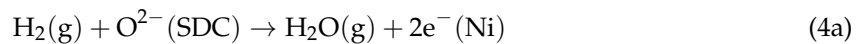
Similarly, the percolated LSCF-dense electrolyte interfaces per unit electrolyte surface area can be estimated by [23]

$$\lambda_{\text{TPB, per}}^S = \gamma_{\text{LSCF, ele}} n_{\text{LSCF}}^S P_{\text{LSCF}}^e \quad (3)$$

where $n_{\text{LSCF}}^S = (1 - \phi_g) \psi_{\text{LSCF}} / (2\pi r_{\text{LSCF}}^2 / 3)$ is LSCF-particles number per unit dense electrolyte surface. $\gamma_{\text{LSCF, ele}} = 2\pi r_{\text{LSCF}} \sin \theta$ is the electrochemical reaction site per connect between an LSCF-particle and a dense electrolyte.

More details about other effective electrode properties calculating, such as, the percolated Ni-SDC-pore TPBs, effective O^{2-} and e^- electric conductivities, hydraulic radius of the porous electrodes and so on could also been found in our previous paper on percolation theory for details [23]. Combing with the electrode microstructure parameters of LSCF-SDC/SDC/Ni-SDC IT-button cell in experiment process [13], the corresponding effective characteristic properties of each layers of the above button cell are estimated and provided in Supplementary Materials. Based on these properties, the multi-scale predictive model that comprehensive considers the special characteristics of the typical mixed conducting SOFCs is developed to study the geometric effects of cathode/anode cross sections relationship on electrochemical performance.

According to anodic e^- - O^{2-} charge transfer reaction, the electrochemical energy relationship at anode active sites, percolated Ni-SDC-pore TPBs (shown in Figure 1b), can be expressed as



$$\mu_{\text{H}_2} + \mu_{\text{O}^{2-}} - 2F\Phi_{\text{O}^{2-}} \geq \mu_{\text{H}_2\text{O}} - 2F\Phi_{\text{e}} \quad (4b)$$

where $\mu_{\alpha} = \mu_{\alpha}^{\text{st}} + RT \ln p_{\alpha}$ is chemical potential of reactant α at local reaction sites. μ_{α}^{st} is chemical potential at standard condition $p^{\text{st}} = 1$ atm. T and p_{α} are the local temperature and partial pressure of species α , respectively. F is Faraday constant. $\Phi_{\text{O}^{2-}}$ and Φ_{e} are the local electrical potentials of O^{2-} and e^- conducting phases, respectively.

'=' in Equation (4b) represents the energy equilibrium state at local place. In this case, the local electromotive force based on local working condition instead of the open circuit condition can be got as [18]

$$E_{\text{an}}^{\text{eq}} = \Phi_{\text{O}^{2-}}^{\text{eq}} - \Phi_{\text{e}}^{\text{eq}} = (\mu_{\text{H}_2} + \mu_{\text{O}^{2-}} - \mu_{\text{H}_2\text{O}}) / 2F$$

'>' in Equation (4b) is essential to process forward reaction with electric current produced. Thus, the activation overpotential is called as the ionic-electronic voltage difference shifted from the local electromotive force $\eta_{\text{act}}^{\text{an}} = E_{\text{an}}^{\text{eq}} - (\Phi_{\text{O}^{2-}} - \Phi_{\text{e}})$.

Similarly, the cathodic e^- - O^{2-} charge transfer reaction and electrochemical energy relationship in local active sites (i.e., percolated LSCF-pore DPBs or LSCF-SDC-pore TPBs in Figure 1b) are



$$0.5\mu_{\text{O}_2} - 2F\Phi_{\text{e}} \geq \mu_{\text{O}^{2-}} - 2F\Phi_{\text{O}^{2-}} \quad (5b)$$

The corresponding electromotive force of equilibrium state at local cathode active sites and the activation overpotential shifted from this $E_{\text{ca}}^{\text{eq}}$ are

$$\eta_{\text{act}}^{\text{ca}} = \frac{1}{4F}(\mu_{\text{O}_2} - 2\mu_{\text{O}^{2-}}) - (\Phi_{\text{e}} - \Phi_{\text{O}^{2-}}) = E_{\text{ca}}^{\text{eq}} - (\Phi_{\text{e}} - \Phi_{\text{O}^{2-}}) \quad (6)$$

Then, the relation between e^- - O^{2-} charge transfer rate per unit TPBs and the activation overpotential can be evaluated by empirical Butler-Volmer equation

$$j_{\text{TPB}} = j_{\text{TPB},0} \left[\exp\left(\frac{2\alpha_{\text{f}}F}{RT}\eta_{\text{act}}\right) - \exp\left(-\frac{2\beta_{\text{r}}F}{RT}\eta_{\text{act}}\right) \right] \quad (7)$$

α_{f} (or β_{r}) is forward (or reverse) reaction symmetric factor. Local exchange current per unit TPB length at anode and cathode sides can be respectively estimated by Reference [24]

$$j_{\text{TPB},0}^{\text{an}} = j_{\text{TPB},0,\text{ref}}^{\text{an}} \exp\left(-\frac{E_{\text{H}_2}}{R}\left(\frac{1}{T} - \frac{1}{T_{\text{ref}}}\right)\right) \left(\frac{p_{\text{H}_2}}{p_0}\right) \quad (8a)$$

$$j_{\text{TPB},0}^{\text{ca}} = j_{\text{TPB},0,\text{ref}}^{\text{ca}} \exp\left(-\frac{E_{\text{O}_2}}{R}\left(\frac{1}{T} - \frac{1}{T_{\text{ref}}}\right)\right) \left(\frac{p_{\text{O}_2}}{p_0^{\text{O}_2}}\right)^{0.25} \quad (8b)$$

where E_{H_2} and E_{O_2} are activation energies for H_2 oxidation and O_2 reduction reactions, respectively. $j_{\text{TPB},0,\text{ref}}$ is assigned empirically based on experiment at reference T_{ref} . p_{α}^0 is partial pressure of species α at open circuit state. Thus, the volumetric current sources for the transfer of e^- - O^{2-} electric charges around the TPBs are $i_{\text{e-O}^{2-},\text{TPB}}^{\text{V}} = j_{\text{TPB}}\lambda_{\text{TPB,per}}^{\text{V}}$ in A m^{-3} . The area metric current sources over electrode/electrolyte interfaces are $i_{\text{e-O}^{2-},\text{TPB}}^{\text{S}} = j_{\text{TPB}}\lambda_{\text{TPB,per}}^{\text{S}}$ in A m^{-2} .

Similarly, the e^- - O^{2-} charge transfer rate over per LSCF-pore DPB area can be evaluated as [18]

$$i_{\text{LSCF}} = i_{\text{LSCF},0} \left[\exp\left(\frac{2\alpha_{\text{LSCF}}F}{RT}\eta_{\text{act}}^{\text{ca}}\right) - \exp\left(-\frac{2\beta_{\text{LSCF}}F}{RT}\eta_{\text{act}}^{\text{ca}}\right) \right], [\text{A m}^{-2}] \quad (9)$$

where $i_{\text{LSCF},0} = i_{\text{LSCF},0,\text{ref}}(p_{\text{O}_2}/p_{\text{O}_2}^0)^{0.25} \exp(-E_{\text{O}_2}/(1/T - 1/T_{\text{ref}})/R)$. $i_{\text{LSCF},0,\text{ref}}$ is assigned empirically at reference temperature T_{ref} . And the volumetric current sources for the transfer of e^- - O^{2-} electric charges around DPBs are $i_{\text{e-O}^{2-},\text{LSCF}}^{\text{V,ca}} = i_{\text{LSCF}}S_{\text{LSCF,per}}^{\text{V}}$ in A m^{-3} .

The above items, expect $\mu_{\text{O}^{2-}}$, can be resolved by the local independent variables, such as, T , p_{α} , Φ_{e} and $\Phi_{\text{O}^{2-}}$. Generally, the constant potential shift does not alter e^- (or O^{2-}) electric potential profiles within the electronic (or ionic) conducting phase. To exclude the influence of $\mu_{\text{O}^{2-}}$ during calculating both η_{act} and charge transfer rate, local electric potentials Φ_{e} and $\Phi_{\text{O}^{2-}}$ were always shifted by different reference amounts, as reported by D. Jean et al. [25] and S. Liu et al. [26]. However, it is necessary to mention that only limited assumptions reported in the above literatures can be used, while the electronic leaking property in dense SDC electrolyte is considered. Because both Φ_{e} and $\Phi_{\text{O}^{2-}}$ are continuously distributed throughout the whole cell structure (anode, electrolyte and cathode).

While keeping Φ_e as it is, local $\Phi_{O^{2-}}$ are shifted by a reference amount as $\hat{\Phi}_{O^{2-}} = \Phi_{O^{2-}} + (\mu_{O_2}^{st} - 2\mu_{O^{2-}})/(4F)$. Then, the overpotential expresses should be adjusted accordingly

$$\eta_{act}^{an} = E^{st} + (\Phi_e - \hat{\Phi}_{O^{2-}}) - \frac{RT}{2F} \ln \frac{p_{H_2O}}{p_{H_2}} \quad (10a)$$

$$\eta_{act}^{cc} = \hat{\Phi}_{O^{2-}} - \Phi_e - \frac{RT}{4F} \ln \frac{1 \text{ atm}}{p_{O_2}} \quad (10b)$$

where $E^{st} = (\mu_{H_2}^{st} + 0.5\mu_{O_2}^{st} - \mu_{H_2O}^{st})/(2F)$ is the Nernst potential at standard state (1 atm).

Combing with the above e^- - O^{2-} charge transfer rates within the composite electrodes and electrode/dense electrolyte interfaces, multi-physics model can be completed by further coupling momentum, mass, electronic and ionic electric current conservation equations. The relationship among electric current densities, electric potentials and e^- - O^{2-} charge transfer rates can be solved by [18]

$$\nabla \cdot i_{O^{2-}} = \nabla \cdot (-\sigma_{O^{2-}}^{eff} \nabla \hat{\Phi}_{O^{2-}}) = \begin{cases} -(i_{e-O^{2-}}^{V,ca,TPB} + i_{e-O^{2-}}^{V,ca,LSCF}) & \text{in cathode} \\ 0 & \text{in dense electrolyte} \\ i_{e-O^{2-}}^{V,an} & \text{in anode} \end{cases} \quad (11a)$$

$$\nabla \cdot i_e = \nabla \cdot (-\sigma_e^{eff} \nabla \Phi_e) = \begin{cases} i_{e-O^{2-}}^{V,ca,TPB} + i_{e-O^{2-}}^{V,ca,LSCF} & \text{in cathode} \\ 0 & \text{in dense electrolyte} \\ -i_{e-O^{2-}}^{V,an} & \text{in anode} \end{cases} \quad (11b)$$

where $i_{O^{2-}}$ and i_e are the O^{2-} and e^- electric current densities within the button cell, respectively. $\sigma_{O^{2-}}^{eff}$ and σ_e^{eff} are the effective O^{2-} and e^- electric conductivities, respectively.

The dusty gas model is adopted to describe gas transport within porous anode and cathode layers [27]

$$\nabla \cdot N_\alpha = R_\alpha \quad (12a)$$

$$\frac{N_\alpha}{D_{\alpha K}^{eff}} + \frac{x_\alpha N_\beta - x_\beta N_\alpha}{D_{\alpha\beta}^{eff}} = -\frac{1}{RT} (p \nabla x_\alpha + x_\alpha \nabla p + \frac{x_\alpha B_0 p}{\mu_{mix} D_{\alpha K}^{eff}} \nabla p) \quad (12b)$$

N_α and x_α are molar flux and local molar fraction of species α , respectively. R_α is reaction rate of each species. It can be evaluated through the e^- - O^{2-} electric current transfer rates per unit electrode volume as

$$\left. \begin{aligned} R_{O_2} &= -(i_{e-O^{2-}}^{V,ca,LSCF} + i_{e-O^{2-}}^{V,ca,TPB})/(4F) \\ R_{N_2} &= 0 \end{aligned} \right\} \text{in cathode} \quad (13a)$$

$$\left. \begin{aligned} R_{H_2} &= -i_{e-O^{2-}}^{V,an,TPB}/(2F) \\ R_{H_2O} &= i_{e-O^{2-}}^{V,an,TPB}/(2F) \end{aligned} \right\} \text{in anode} \quad (13b)$$

The total gas pressure and permittivity within the porous structure can be evaluated by

$$p = \frac{c_{tot}}{RT}, B_0 = \frac{\phi_g^3 r_g^2}{8\tau^2} \quad (14)$$

where r_g is mean hydraulic pore radius of the specified porous electrode structure. τ is the corresponding tortuosity of the porous structure [28]. The effective dynamic viscosity of mixture gas μ_{mix} can be predicted by ideal gas mixing law [29]

$$\mu_{mix} = \sum_{\alpha=1}^n \frac{x_\alpha \mu_\alpha}{\sum_{\beta=1}^n x_\beta \Phi_{\alpha,\beta}}, \frac{\mu_\alpha}{\mu_\alpha^0} \approx \left(\frac{T}{T_0} \right)^{1.5} \frac{T_0 + S}{T + S} \quad (15a)$$

$$\Phi_{\alpha,\beta} = \frac{1}{\sqrt{8}} \left(1 + \frac{M_\alpha}{M_\beta} \right)^{-1/2} \left[1 + \left(\frac{\mu_\alpha}{\mu_\beta} \right)^{1/2} \left(\frac{M_\alpha}{M_\beta} \right)^{1/4} \right]^2 \quad (15b)$$

where M_α is the molar mass. μ_α is the dynamic viscosity of species α , which can be evaluated based on Sutherland's law based on the relevant parameters in Table 1.

Table 1. Gas composition and parameters for viscosity calculations by Sutherland's law.

Gas	$\nu_\alpha (\times 10^{-6} \text{ m}^3 \text{ mol}^{-1})$	$\mu_\alpha^0 (\times 10^{-6} \text{ kg m}^{-1} \text{ s}^{-1})$	$T_0 \text{ (K)}$	$S \text{ (K)}$
H ₂	6.12	8.411	273	97
vapor	13.1	11.2	350	1064
O ₂	16.3	19.19	273	139
N ₂	18.5	16.63	273	107

The effective Knudsen diffusion coefficient [30] of species α and effective binary diffusion coefficient [31] can be estimated by

$$D_{\alpha K}^{\text{eff}} = \frac{\phi_g}{\tau} \frac{2r_g}{3} \sqrt{\frac{8RT}{\pi M_\alpha}}, \quad D_{\alpha\beta}^{\text{eff}} = \frac{\phi_g}{\tau} \frac{3.24 \times 10^{-8} T^{1.75}}{p(\nu_\alpha^{1/3} + \nu_\beta^{1/3})} \left(\frac{1}{M_\alpha} + \frac{1}{M_\beta} \right)^{0.5} \quad (16)$$

where ν_α is diffusion volume of species α , which is collected in Table 1.

3. Result and Discussion

Figure 3 shows five calculated $i_{\text{op}}-V_{\text{op}}$ curves at different operation T for a LSCF-SDC/SDC/Ni-SDC button cell with the ratio of anode and cathode discs radii around $r_{\text{ca}}/r_{\text{an}} = 0.8 \text{ cm}/1 \text{ cm}$. In other words, the surface areas ratio of anode and cathode is $A_{\text{ca}}/A_{\text{an}} = 0.64$. The corresponding parameters are illustrated in Supplementary Materials. It is necessary to mention that the deviation of $i_{\text{op}}-V_{\text{op}}$ curves between the calculating and experiment results in high current density zone at 700 °C was considered to be an error caused by some unknown factors during the testing process based on follow considerations. (i) The sharp drop of $i_{\text{op}}-V_{\text{op}}$ curve at high current density zone is considered to be caused by concentration overpotential. However, the limited current density at 700 °C smaller than that at 600 °C is unreasonable. (ii) These deviations were happened around the up boundary operation zone (up operation temperature and current density zones). The deviation could be caused by abnormal factors. (iii) Good agreements between calculated and experiment results [13] at several other T can well illustrate that the modeling parameters can well represent the electrochemical properties of the button cell; and the cell-level multi-physics model can well describe the working details within it. It should be note that the feature of electronic leakage of dense electrolyte would lead to a sharp decrease of the open circuit voltage (shown in Figure 3).

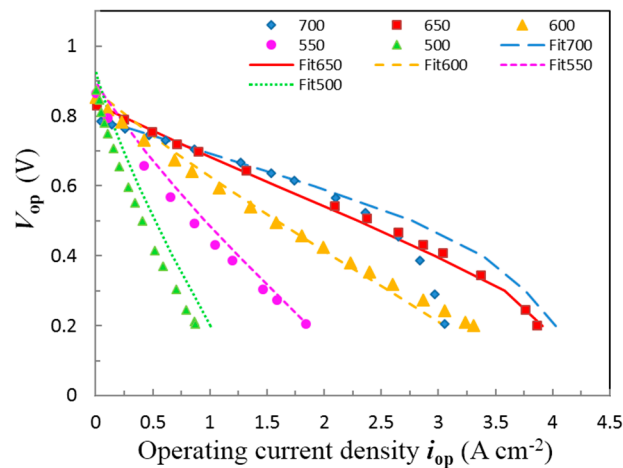


Figure 3. Comparison between the numerical modeling and experiment [13] results at various operating temperatures.

For a button cell, the support component layer is always fabricated with a relative larger surface area compared with the measured electrode [32] (i.e., the anode surface area in current button cell is 3.14 cm^2 and the corresponding surface area of measured cathode is only 2 cm^2). Obviously, for a specified output voltage V_{op} , the responded operating current density i_{op} can be evaluated by two ways, divided the output current I_{op} by A_{an} for a larger value or A_{ca} for a lower value. To improve the performance quality, most of the reported i_{op} - V_{op} curves of button cells were always obtained based on the relative smaller area between A_{an} and A_{ca} . Thus, it is important to evaluate the influence of the over zone from support layer on the experiment measuring and numerical calculating i_{op} - V_{op} performance results for IT-SOFC button cell using mixed conducting material.

Effect of different A_{ca}/A_{an} ratio at 700 and 600 °C: Figure 4 compares the I_{op} - V_{op} performances of LSCF-SDC/SDC/Ni-SDC IT-SOFC button cells with respectively cathode area 2 and 0.5 cm^2 (labeled as cells 1 and 2), while kept the surface area of support anode as 3.14 cm^2 . Generally, the experimentally measured i_{op} - V_{op} performances may be obtained by the following two steps. Firstly, the responded output currents I_{op} should be measured while the output voltages V_{op} are specified. Then, the corresponding output current densities i_{op} can be obtained through divided I_{op} by the electrode surface. Table 2 compares the i_{op} - V_{op} relations of cells 1 and 2 based on both anode and cathode cross section surfaces, respectively. Obviously, using the relative larger electrode surface (i.e., A_{ca} in current anode supported case) means larger i_{op} value. Using the relative lower electrode surface (i.e., A_{an}) means lower i_{op} value. It is interesting to find that there are very similar i_{op} - V_{op} relationships between cells 1 and 2, while estimated i_{op} based on the relevant smaller electrode surface area (i.e., A_{ca} in current case). There is no obvious difference between the performance results even at the maximum power density case. Taking $V_{op} = 0.5 \text{ V}$ and $T = 700 \text{ °C}$ as an example, the maximum power densities are 1.412 W cm^{-2} for cathode area 0.5 cm^2 and 1.397 W cm^{-2} for $A_{ca} = 2 \text{ cm}^2$ cases, respectively.

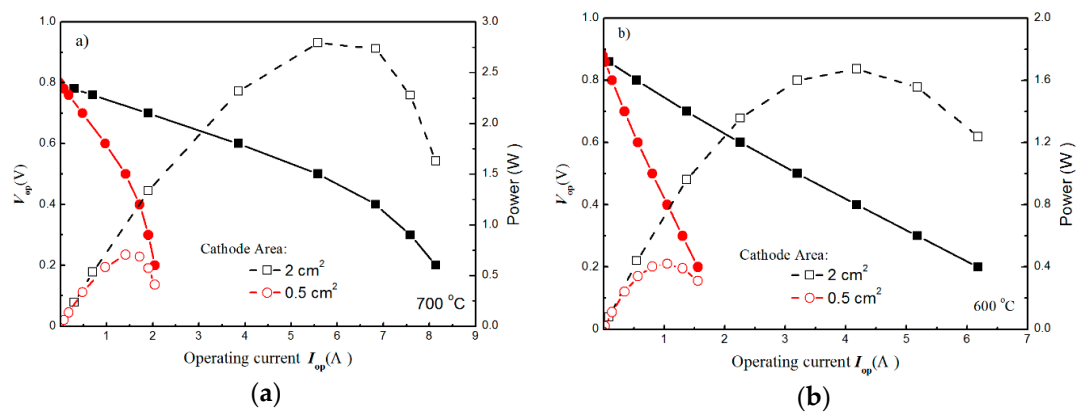


Figure 4. Effects of different cathode areas on cell, (a) I_{op} - V_{op} performances at 700 °C; (b) I_{op} - V_{op} performances at 600 °C.

Thus, it can be concluded that although the over zone of larger electrode is generally considered can enhance charges and gases transport capacities in a proper zone, the active zone of the button cell will be restricted at the electrode zone with relevant small area (i.e., cathode layer area in current anode support case), instead of the support layer surface areas (i.e., A_{an}). The influence of the over zone of anode surface area on i_{op} - V_{op} performance would be negligible. Taking the small area between anode and cathode surfaces to calculate i_{op} - V_{op} performance is more reasonable to indicate the electrochemical properties of the tested IT-SOFC button cell.

Table 2. For a specified V_{op} , comparing the responded i_{op} that are evaluated by divided I_{op} with A_{an} for larger value and A_{ca} for lower value, respectively.

V_{op}	I (A)		i_{op} Based A_{an} ($A\ cm^{-2}$)		i_{op} Based on A_{ca} ($A\ cm^{-2}$)	
	Cell 1	Cell 2	Cell 1	Cell 2	Cell 1	Cell 2
0.2	8.139	2.047	2.592	0.652	4.069	4.095
0.3	7.585	1.906	2.416	0.607	3.793	3.811
0.4	6.838	1.717	2.178	0.547	3.419	3.434
0.5	5.587	1.412	1.779	0.450	2.794	2.824
0.6	3.860	0.971	1.229	0.309	1.930	1.942
0.7	1.905	0.481	0.607	0.153	0.953	0.962
0.78	0.302	0.078	0.096	0.025	0.151	0.155

$A_{an} = 3.14\ cm^2$. $A_{ca} = 2$ and $0.5\ cm^2$ for cell 1 and 2, respectively.

Effects of Electrolyte thickness on cell performance: Generally, too thin electrolyte layer is considered as a key factor to weaken the influence of the over zone of larger electrode on i_{op} - V_{op} performance. Figure 5 further compares four different cell performances at 600 °C among the button cells with different combination of cathode surface areas (0.5 and $2\ cm^2$) and electrolyte thicknesses (i.e., 15 and 50 μm), while keeps the anode surface areas and other operating parameters values. Obviously, we can find that increase the dense electrolyte thickness in a limited value from 13 to 50 μm would not affect the above conclusions. Taking the small areas between anode and cathode surfaces to calculate i_{op} - V_{op} performance is more reasonable to indicate the electrochemical properties of the tested IT-SOFC button cell.

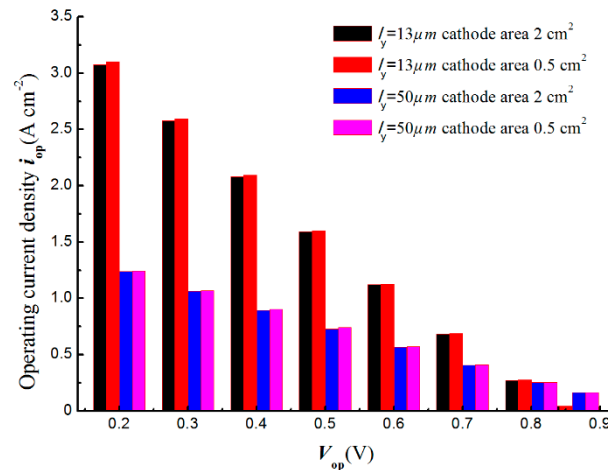


Figure 5. The effects of different A_{ca}/A_{an} on the button cell performances at 600 °C while increases the thickness of dense electrolyte from 15 to 50 μm .

Effects of exchange current density: The exchange current density $j_{TPB, 0, ref}^{an}$ based on TPBs is an important factor to character the electrochemical property of the button cells. A higher $j_{TPB, 0, ref}^{an}$ means that smaller activation overpotential is needed to convert same amount of charge between e^- and O^{2-} electric currents. As shown in Figure 6a, for a specified i_{op} , V_{op} increases with the increasing $j_{TPB, 0, ref}^{an}$. However, it is interesting to get that for all the three cases with $j_{TPB, 0, ref}^{an} = 8.0 \times 10^{-2}$, 8.0×10^{-3} and $8.0 \times 10^{-4} \text{ A m}^{-1}$, the maximum power density differences between $A_{ca} = 0.5$ and 2 cm^2 cases are less than 100 W m^{-2} , at 0.4 V and 600 °C. Therefore, for those button cells with exchange current densities within reasonable range, the effect of the over zone from larger electrode surface on button cell performance is insignificant.

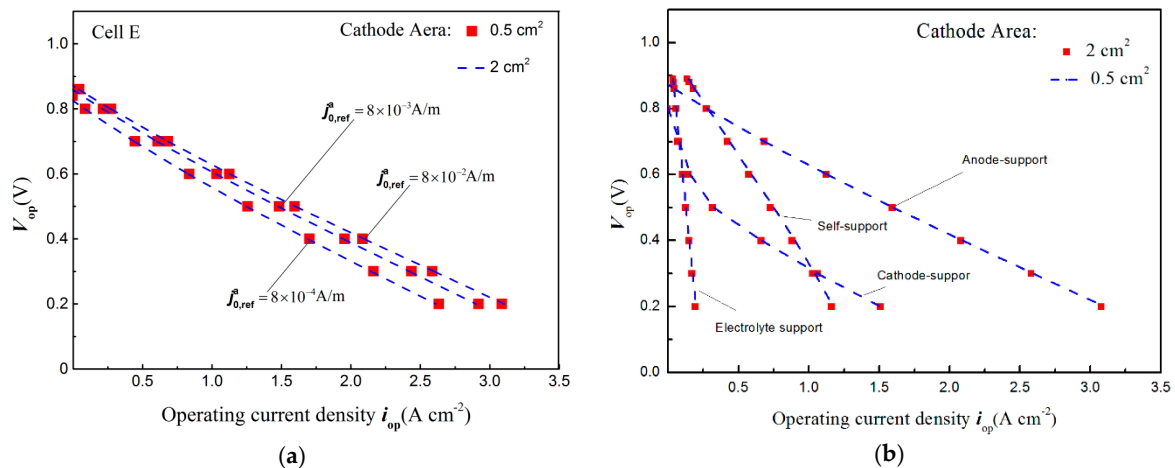


Figure 6. The sensitivity of button cell performances on different A_{ca}/A_{an} : (a) Under various exchange current densities, (b) while different component support cases are considered.

Effects of different component support cases: Figure 6b shows the sensitivities of button cell performances on different anode/cathode surface area ratios at 600 °C, while different component support cases are adopted. The geometry parameters of the anode, cathode, electrolyte and component-self-support button cells are respectively listed in Table 3. Although different component support cases would lead to very different button cell performances, it should be noted that the sensitivities of i_{op} - V_{op} performances on different A_{ca}/A_{an} are still insignificant while evaluating i_{op} based on the smaller electrode surface area between anode and cathode layers.

Table 3. Geometry parameters of the anode, cathode, electrolyte and component-self-support button cells in the unit of mm.

Item	Anode Support	Functional Layer	Dense Electrolyte	Cathode
Anode support	460	6	13	40
Cathode support	20	6	13	480
Electrolyte support	20	6	460	40
Self support	100	100	50	200

The real affection zone of the over electrode surface area: It is theoretically agreed that the over anode (or cathode) surface area due to $A_{ca}/A_{an} \neq 1$ will decrease the potential losses of the gas, electron and ion transports in the corresponding electrode. The above study results, however, show that the geometric effect of anode and cathode surface areas ratio on the IT-button cell i_{op} - V_{op} performance is quite limited, while the working parameters vary in a reasonable range. The active zone of the button cell is restricted within the smaller electrode area zone between anode and cathode (i.e., $\min(A_{ca}, A_{an})$). This should be caused by the geometric characteristics of button cell with relative thin component layer. Taking dense electrolyte layer as an example, the length-width ratio between thickness and radius of disc surface is $13 \mu\text{m}/1 \text{ cm} = 0.0013$. Thus, finding out the real influence width of this over electrode zone on the button cell performance would be very helpful to understand the working properties of IT-SOFCs.

Taking $T = 600^\circ\text{C}$ and $V_{op} = 0.4 \text{ V}$ as an example, the concentration distributions of H_2 and H_2O within porous composite anode, c_{H_2} and $c_{\text{H}_2\text{O}}$, are shown in Figure 7. c_{H_2} on anode side is $13.563 \text{ mol m}^{-3}$ initially and drops sharply along z -axis to 6.81 mol m^{-3} due to the oxidation reaction of H_2 . In contrast, the concentration of product vapor $c_{\text{H}_2\text{O}}$ on anode side increases along z axis from 0.42 to 12.4 mol m^{-3} . Obviously, c_{H_2} and $c_{\text{H}_2\text{O}}$ have opposite distribution characteristics. Theoretically, the over zone of the current anode surface (i.e., $A_{an} = 3.14 \text{ cm}^2$, $A_{ca} = 2 \text{ cm}^2$, and $A_{ca}/A_{an} = 0.64$ in current button cells) would enhance the hydrogen and vapor transports within porous anode in a proper width because of the enlarged cross section.

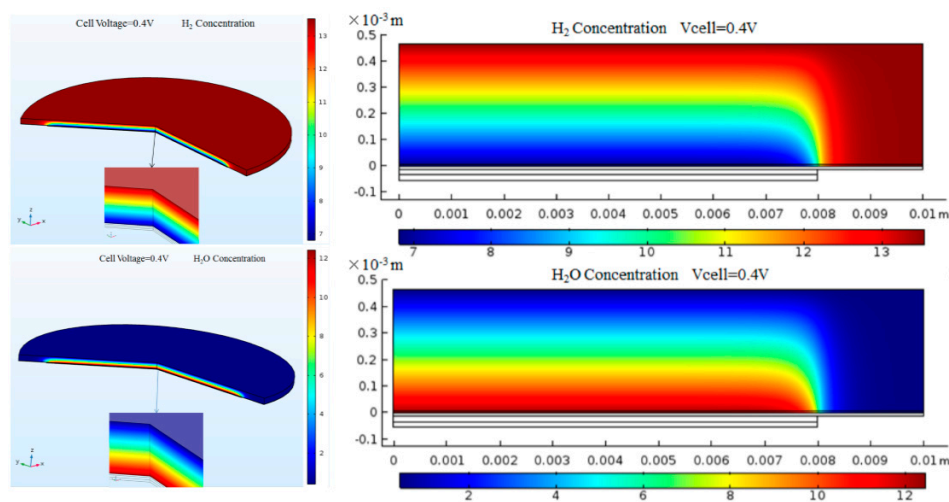
**Figure 7.** The concentration distributions of H_2 and H_2O within anode at $T = 600^\circ\text{C}$ and $V_{op} = 0.4 \text{ V}$ as an example.

Figure 8a further shows a c_{O_2} distribution within porous cathode. The O_2 concentration is consumed from 2.93 to 1.41 mol m^{-3} . As shown in Figure 8c, the over zone of the anode surface area can also enhance the conducting capacity of electronic current density in y direction due to the enlarged cross section. Combining Figures 7 and 8, we can find that the real effective width of the over electrode

zone in an IT-SOFC button cell is only in a scale of 0.03 cm. The over electrode zone exceed this width would be inactive. This can well explain that why the IT-SOFC button cells i_{op} - V_{op} performance is insensitive to the A_{ca}/A_{an} ratio; and a smaller A_{ca}/A_{an} may not greatly increase the measured i_{op} - V_{op} electrochemical quality. Because the button cell is fabricated in a radius scale of $r_{disc} = 1$ cm. These real effective width of the over electrode zone reference to the button cell disc overall radius is less than 5%.

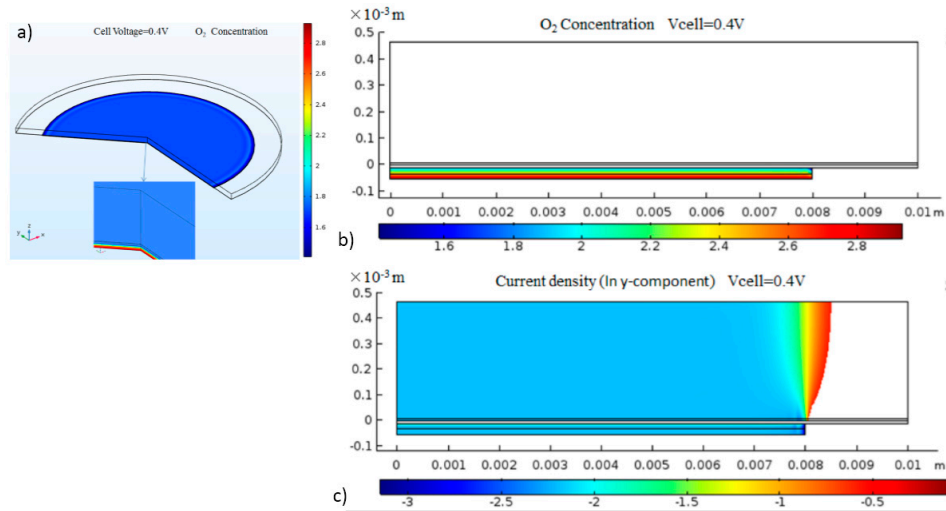


Figure 8. (a,b) c_{O_2} distribution within porous cathode at $T = 600$ °C and $V_{op} = 0.4$ V as an example; (c) i_e distribution throughout the whole button cell.

To further confirm this conclusion, a button cell in a smaller scale (i.e., $r_{disc} = 0.1$ cm, $A_{an} = 3.14 \times 10^{-2}$ cm²) is developed. Figure 9 compares the i_{op} - V_{op} performances between the buttons cells with different cathode/anode surface areas ratios (i.e., $A_{ca} = 2 \times 10^{-2}$ and 0.5×10^{-2} cm², $A_{ca}/A_{an} = 0.64$ and 0.16). The corresponding maximum power densities are 0.9749 and 0.8814 W cm⁻² for $A_{ca}/A_{an} = 0.64$ and 0.16 cases, respectively. Obviously, while the geometry of the button cells or other electrochemical devices approach the scale less than 100 μ m, the effect of the over electrode zone on the electrochemical performance should not be ignored. Taking the smaller electrode surface area to evaluate i_{op} for a specified V_{op} would cause an improper over evaluation of the electrochemical performance.

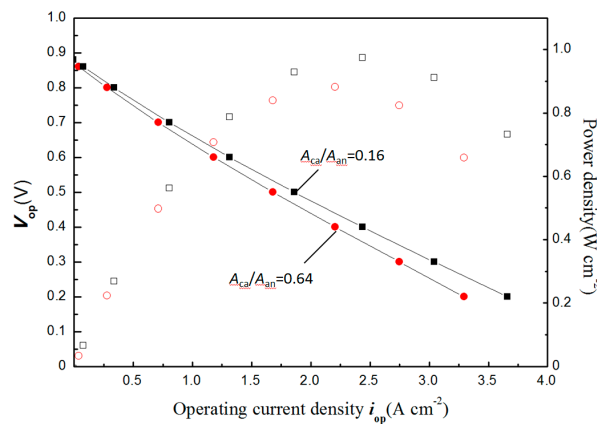


Figure 9. i_{op} - V_{op} performances between the buttons cells with different cathode/anode surface areas ratios (i.e., $A_{an} = 3.14 \times 10^{-2}$ cm², $A_{ca}/A_{an} = 0.64$ and 0.16).

4. Conclusions

The comprehensive multi-physics model of IT-SOFC button cells considers special features, such as using mixed conducting materials, electric leakage and complex multi-physics mutual coupling processes have been developed and verified. The geometry effect of different anode and cathode surface area ratios on i_{op} - V_{op} performance of IT-SOFC button cells are investigated and many conclusions are reached,

- (i). The over zone of the larger electrode can only enhance charges and gas transport capacities within a limited scale of only 0.03 cm, an over electrode zone exceeding this width would be inactive.
- (ii). The active zone of button cell is restricted within the smaller electrode area $\min(A_{an}, A_{ca})$ due to the relatively large disc radius in scale of cm and the thin component layer.
- (iii). For a specified V_{op} , evaluating the responded i_{op} by dividing output current I_{op} with $\min(A_{an}, A_{ca})$ for a larger value is reasonable for presenting the real performance in a current device scale.
- (iv). While the geometry of button cell or other electrochemical device approaches a scale of less than 100 μm , taking the smaller electrode surface area to evaluate i_{op} for a specified V_{op} would cause an improper over evaluating of the electrochemical performance.

Supplementary Materials: The following are available online at <http://www.mdpi.com/1996-1073/11/7/1875/s1>.

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Nomenclature

A_{ca}	the cross section area of the cathode layer, m^2
A_{an}	the cross section area of the anode layer, m^2
B_0	the flow permeability, m^2
c_α^0	the concentration of species α at the channel inlet, mol m^{-3}
DPBs	the LSCF-pore double phase boundaries
$D_{\alpha\beta}^{\text{eff}}$	the effective binary diffusivity, $\text{m}^2 \text{s}^{-1}$
$D_{\alpha K}^{\text{eff}}$	the effective Knudsen diffusivity of species α , $\text{m}^2 \text{s}^{-1}$
E^{eq}	the local equilibrium electric potentials difference at working state, V
E^{st}	the Nernst potential at the standard state, V
E_{H_2}	the activation energy for H_2 oxidation reaction, J
E_{O_2}	the activation energy for O_2 reduction reaction, J
F	the Faraday constant, C mol^{-1}
GDC	the $\text{Gd}_{0.1}\text{Ce}_{0.9}\text{O}_{1.95}$
j_{TPB}	the local e^- - O^{2-} charge transfer rate per unit TPB length, A m^{-1}
$j_{\text{TPB},0}$	the local exchange transfer current per unit TPB lengths, A m^{-1}
$j_{\text{TPB},0,\text{ref}}$	the value assigned empirically based on experiment at reference temperature
i_e	the local e^- electric current density, A m^{-2}
$i_{\text{O}^{2-}}$	the local O^{2-} electric current densities, A m^{-2}
i_{op}	the output current density, A m^{-2}
I_{op}	the output current, A
i_{LSCF}	the e^- - O^{2-} charge transfer rate per unit percolated DPB area, A m^{-2}
$i_{\text{LSCF},0}$	the local exchange transfer current per unit percolated DPB area, A m^{-2}
$i_{\text{e-O}^{2-},\text{TPB}}^{\text{V}}$	the e^- - O^{2-} charge transfer rate per unit volume based on percolated TPBs, A m^{-3}
$i_{\text{e-O}^{2-},\text{LSCF}}^{\text{V}}$	the e^- - O^{2-} charge transfer rate per unit volume based on percolated LSCF-pore DPBs, A m^{-3}
$i_{\text{e-O}^{2-},\text{TPB}}^{\text{S}}$	the e^- - O^{2-} charge transfer rate per unit dense electrolyte surface, A m^{-2}

LSCF	the $\text{La}_{0.6}\text{Sr}_{0.4}\text{Co}_{0.2}\text{Fe}_{0.8}\text{O}_{3-\delta}$
LSM	the $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$
M_α	the mole mass of species α , kg mol^{-3}
N_α	the molar flux of species α , $\text{mol m}^{-2} \text{s}^{-1}$
n_k^V	the number of k -particles per unit volume
n_k^S	the number of k -particles per unit dense electrolyte surface area
p_α	the partial pressure of gas species α at the local reaction sites, atm
p_α^0	the partial pressure of gas species α in the channel inlet, atm
$P_{\text{SDC}}^{\text{O}^{2-}}$	the probabilities of SDC-particles belonging to percolated O^{2-} conducting path
$P_{\text{LSCF}}^{\text{O}^{2-}}$	the probabilities of LSCF-particles belonging to percolated O^{2-} conducting path
P_{LSCF}^e	the probabilities of LSCF-particles belonging to percolated e^- conducting path
r_g	the mean hydraulic pore radius of porous electrode structure, m
r_k	the radius of k -particle, m
r_c	the neck radius between two connected particles, m
R	the universal gas constant, $\text{J mol}^{-1} \text{K}^{-1}$
R_α	the sources/leak of species α , $\text{mol m}^{-3} \text{s}^{-1}$
SDC	the $\text{Sm}_{0.2}\text{Ce}_{0.8}\text{O}_{2-\delta}$
$S_{\text{LSCF, per}}^V$	the percolated LSCF-pore DPBs per unit volume, m^{-1}
s_{es}	the exposed surface area of each LSCF-particle, m^2
T	the operating temperature, K
TPBs	the three phase boundary sites
V_{op}	the output voltage at working state, V
v_α	the diffusion volume for species α , $\text{m}^3 \text{mol}^{-1}$
x_α	the molar fraction of species α
YSZ	the yttrium-stabilized zirconia
$Z_{k,l}$	the average number of contacts between k - and all of its neighboring l -particles
$\bar{Z}$	the average coordination number of all particles

Greek letters

α_f, β_r	the forward and reverse reaction symmetric factors
$\gamma_{\text{LSCF, SDC}}$	the 1D circular length per contact between LSCF- and SDC-particles, m
$\gamma_{\text{LSCF, ele}}$	the 1D circular length per contact between LSCF-particle and the dense electrolyte, m
$\lambda_{\text{TPB, per}}^V$	the percolated TPB length per unit volume, m^{-2}
$\lambda_{\text{TPB, per}}^S$	the percolated TPB length per dense electrolyte surface area, m^{-1}
ϕ_g	the porosity of porous structure
Φ_e	the local e^- electric potential, V
$\Phi_{\text{O}^{2-}}$	the local O^{2-} electric potential, V
$\hat{\Phi}_{\text{O}^{2-}}$	the shift of $\Phi_{\text{O}^{2-}}$ by a reference amount, V
ψ_k	the solid volume fraction of k -particles
η_{act}	the local activation overpotential, V
θ	the smaller contact angle between two particles
τ	the tortuosity of gas transport path within the porous electrode
σ_e^{eff}	the effective electronic conductivity, S m^{-1}
$\sigma_{\text{O}^{2-}}^{\text{eff}}$	the effective O^{2-} ionic conductivity, S m^{-1}
μ_{mix}	the viscosity of gas mixture, $\text{kg m}^{-1} \text{s}^{-1}$
μ_α	the mole chemical potential of reactant α , J mol^{-1}

Superscripts and subscripts

an	anode
act	activation
ca	cathode
eq	equilibrium
st	standard condition (1 atm)
ref	reference value

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