



Short communication

Interfacial strain effect on gas transport in nanostructured electrodes of solid oxide fuel cells

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HIGHLIGHTS

- Evaluate correlation between interfacial strain and concentration polarization.
- Shows that compressive strain is unfavorable for reducing concentration polarization.
- Designs interfaces to improve performance of fuel cells.

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ABSTRACT

Most efforts regarding strain effect at the interfaces between electrolytes and electrodes are mainly focused on enhancing the ionic conductivity in electrolytes. However, fundamental insights into the strain effect on gas transport properties in electrodes of fuel cells are still lacking. In this report, quantitative analysis is performed to evaluate the correlation between interfacial strain and the important fuel cells parameters, including limiting current density and concentration polarization. We demonstrate that the strain effect plays an important role in the performance of solid oxide fuel cells with nanostructured electrodes. Our studies provide a powerful platform for reducing concentration polarization by engineering quantitatively the interfacial strain, and facilitating the development of high-efficiency nanostructured fuel cells.

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1. Introduction

Solid oxide fuel cells (SOFCs) have received extensive attention due to the high efficiency, low pollution emission, fuel flexibility and scalable modularity. However, their high operating temperature (typically over 800 K) leads to the high cost and short service time, which impede the commercialization of SOFCs [1,2]. Many efforts have been focused on decreasing the operating temperature to a more operational level. In particular, utilizing existing materials to realize nanoscale electrodes/electrolytes has been the primary trajectory [3,4]. Compared to bulk materials, nanostructured electrolytes and electrodes have exhibited promising application advantages in energy storage systems due to their improved conversion efficiency. Most nanoscale electrode/electrolyte thin films with high transport properties have been

fabricated by advanced deposition techniques [5,6]. While the electrode/electrolyte materials are made into nanoscale thin films, substrates are needed to support such thin films. Thus, nanoscale heterostructures are formed between electrode/electrolyte thin films and substrates. Interestingly, the strain effect involving to the structure deviation on the atomistic scale at the dissimilar interface has shown extraordinary capability of improving the performance of SOFCs [7–12]. Barriocanal et al. observed that a 7% tensile strain of 1 nm YSZ in YSZ/SrTiO₃ system could result in an ionic conductivity enhancement on 8 orders of magnitude at 573 K [13]. Kushima et al. reported a ~4 orders of magnitude enhancement in ionic conductivity induced by the 4% tensile strain in YSZ at 400 K [14]. Souza et al. also found that a 4% tensile strain could enhance the in-plane ionic conductivity in CeO₂ by ~4 orders of magnitude at 500 K [15]. Furthermore, in addition to the electrolyte heterostructures, the interfacial strain has also been found to be formed between the electrodes and electrolytes due to their different thermal expansion coefficients and lattice mismatch [16–19]. Nevertheless, most efforts were merely focused on the

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tensile strain effect on enhancing the ionic conductivity in electrolytes, the discussion of compressive strain effect on gas transport in porous electrodes of fuel cells is still lacking. In practice, the slow gas diffusivity in porous electrodes could significantly limit the energy conversion efficiency of fuel cells due to the much energy loss [20]. In addition, microscopic structure deviations in electrodes induced by interfacial strain can influence the accurate gas diffusivity measurement with our previous proposed electrochemical devices, leading to the inaccurate evaluations of limiting current density and concentration polarization [21–23]. Therefore, a systematic analysis of the quantitative correlation between interfacial strain effect and gas transport properties is desirable. In this report, we investigate the strain effect at the interface between nanoscale electrolytes and electrodes on gas diffusivity efficiency in porous electrodes of SOFCs. Two important fuel cell parameters—limiting current density and concentration polarization are employed to characterize the properties of SOFCs. Our work provides a quantitative basis for evaluating and engineering the variation of limiting current density and concentration polarization in fuel cells induced by the interfacial strain.

2. Theoretical analysis

As the thicknesses of electrodes/electrolytes are decreased to nanoscale, more strain energy at the interfaces between electrodes and electrolytes will be released by the structure deviation [24,25]. Since the adjacent electrode and electrolyte materials have different thermal expansion coefficient and lattice mismatch, strain is formed at the dissimilar interfaces. As shown in Fig. 1, for porous electrode, the pore volume (V) and size (a_0) can be altered by the strain (ε) at the interface between the nanoscale electrodes and electrolytes,

$$V = \frac{\delta(a_0^2 + a_0a_1 + a_1^2)}{3} + a_0^2(L - \delta) \quad (1)$$

$$\varepsilon = \frac{a_1 - a_0}{a_0} \quad (2)$$

$$\delta = \frac{1}{4} \sqrt{\frac{2}{3} \frac{1 - \nu}{1 + \nu}} \cdot l \quad (3)$$

where a_1 is the side length of strained pore, L is the electrode thickness, and δ is the width of the strained interface region [25], which depends on the Poisson ratio (ν) and diameter (l) of the crystallites in electrodes, as Eq. (1) shows. In general, δ is ~10 nm [26]. The meanings of all the symbols in the report are shown in Table 1. The electrode volume percent of porosity (V_V) is expressed as Eq. (4). Combining Eqs. (1), (2) and (4), leads to Eq. (5),

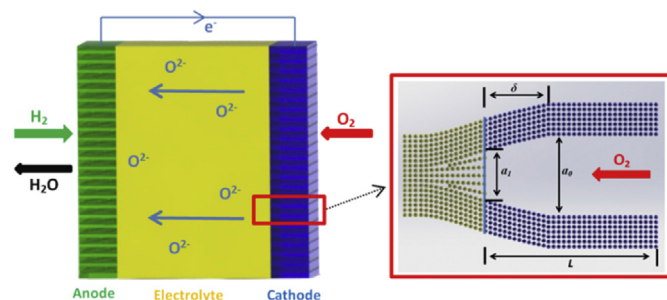


Fig. 1. Schematic of a strain model at the interface between electrodes and electrolytes.

$$V_V = \frac{V}{V_T} \quad (4)$$

$$V_V = \frac{a_0^2 \left[L + \delta \left(\frac{\varepsilon^2}{3} + \varepsilon \right) \right]}{V_T} \quad (5)$$

where V_T is the total volume of electrode.

Since the pore size of electrode is nanoscale, the binary diffusivity bulk and Knudsen diffusivity coexist in electrodes [27], and both of them are dependent of V_V , as expressed in Eqs. (6)–(9),

$$D_{H_2,k}^{eff} = \frac{dV_V}{3\tau} \sqrt{\frac{8RT}{\pi M_{H_2}}} \quad (6)$$

$$D_{O_2,k}^{eff} = \frac{dV_V}{3\tau} \sqrt{\frac{8RT}{\pi M_{O_2}}} \quad (7)$$

$$D_{H_2-H_2O}^{eff} = \frac{0.00186T^{\frac{3}{2}}V_V}{p\sigma_{H_2-H_2O}^2\Omega_1\tau} \left(\frac{1}{M_{H_2}} + \frac{1}{M_{H_2O}} \right)^{\frac{1}{2}} \quad (8)$$

$$D_{O_2-N_2}^{eff} = \frac{0.00186T^{\frac{3}{2}}V_V}{p\sigma_{O_2-N_2}^2\Omega_2\tau} \left(\frac{1}{M_{O_2}} + \frac{1}{M_{N_2}} \right)^{\frac{1}{2}} \quad (9)$$

$$d = \frac{a_0(2L + \varepsilon\delta)}{2L} \quad (10)$$

where $D_{H_2,k}^{eff}$ ($D_{O_2,k}^{eff}$) and $D_{H_2-H_2O}^{eff}$ ($D_{O_2-N_2}^{eff}$) is effective Knudsen diffusivity of H_2 (O_2) and effective binary diffusivity of H_2 and O_2 (O_2 and N_2), respectively, d is the mean diameter of pores that is dependent of strain, as described in Eq. (10), τ is the tortuosity factor, R is the ideal gas constant, T is the operating temperature, p is the total pressure (1 atm), $\sigma_{H_2-H_2O}$ ($\sigma_{O_2-N_2}$) is the average collision diameter set at 2.734 Å (3.6325 Å), Ω_1 (Ω_2) is the collision integral set at 0.8998 (0.8568) [27,28], M_{H_2} (M_{O_2}) and M_{H_2O} (M_{N_2}) denote the molar weights of H_2 (O_2) and H_2O (N_2), respectively. The diffusivities of H_2 (D_{H_2}) in anode and O_2 (D_{O_2}) in cathode are expressed through Eqs. (11) and (12), respectively [20,29].

$$\frac{1}{D_{H_2}} = \frac{1}{D_{H_2,k}^{eff}} + \frac{1}{D_{H_2-H_2O}^{eff}} \quad (11)$$

$$\frac{1}{D_{O_2}} = \frac{1}{D_{O_2,k}^{eff}} + \frac{1}{D_{O_2-N_2}^{eff}} \quad (12)$$

Substituting Eqs. 5–10 in Eqs. (11) and (12) gives:

$$D_{H_2} = \frac{V_V}{\frac{A_1}{2L+\varepsilon\delta} + B_1} = \frac{a_0^2 \left[L + \delta \left(\frac{\varepsilon^2}{3} + \varepsilon \right) \right]}{V_T \left(\frac{A_1}{2L+\varepsilon\delta} + B_1 \right)} \quad (13)$$

$$D_{O_2} = \frac{V_V}{\frac{A_2}{2L+\varepsilon\delta} + B_2} = \frac{a_0^2 \left[L + \delta \left(\frac{\varepsilon^2}{3} + \varepsilon \right) \right]}{V_T \left(\frac{A_2}{2L+\varepsilon\delta} + B_2 \right)} \quad (14)$$

where $A_1 = 6\tau L/a_0 \sqrt{\pi M_{H_2}/8RT}$ ($A_2 = 6\tau L/a_0 \sqrt{\pi M_{O_2}/8RT}$) and

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