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ABSTRACT

The technique allowing for direct gas diffusivity measurement in fuel cells has been advanced rapidly in recent years. Such development has largely improved the accuracy in the evaluation of fuel cell limiting current density (LCD) and overall concentration polarization (CP). However, electrolyte discs were employed in all the previous gas diffusivity electrochemical devices, which inevitably induced gas leak in the gas diffusivity measurement. In this report, quantitative analysis is performed to evaluate the correlation between gas leak and the important fuel cell parameters including electrode diffusivity, LCD and CP. To avoid gas leak, electrochemical devices without any electrolyte discs is then proposed. Our quantitative study and the proposed devices facilitate the accurate analysis on the electrochemical performance of a fuel cell system and the development of highly-efficient fuel cells.

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1. Introduction

Research in the fuel cell field has been mainly focused on the search for new functional materials and the improvement in existing energy materials. For instance, Genorio et al. reported the synthesis and analysis of chemically modified platinum for the anode application in proton exchange membrane fuel cell (PMEFC) [\[1\]](#page--1-0). Liu et al. investigated the catalytic properties of nanostructured $Ba_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O_{3-\delta}$ for the cathode application in solid oxide fuel cells (SOFCs) [\[2\].](#page--1-0) Dieterle et al. developed nanoscale $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and analyzed its promising application potential to be employed as cathode material in fuel cells <a>[3]. Meanwhile, Périllat-Merceroz et al. improved the structural and electrical properties of SOFC anodes by

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using lamellar titanates [\[4\].](#page--1-0) Park et al. doped strontium in the cathode material $Pr_{1-x}Sr_{x}CoO_{3-\delta}$, and the resulted cathode exhibited excellent electrical properties [\[5\]](#page--1-0). These investigations facilitate the development of highly efficient fuel cells with low cost. In addition to materials research, another important direction, the development of efficient techniques for the electrochemical performance evaluation of fuel cells, has also been advanced rapidly in recent years $[6-8]$ $[6-8]$ $[6-8]$. In particular, the authors have designed and developed several electrochemical devices for the direct gas diffusivity measurement in fuel cells. For instance, single-sensor, doublesensor and multi-sensor devices have been designed for the anode/cathode diffusivity measurement in fuel cells $[9-13]$ $[9-13]$. Recently, the authors developed an electrochemical device for the three-dimensional (3D) diffusivity measurement in fuel cells [\[14\]](#page--1-0). Through the device, the diffusivities of an electrode in three electrode directions can be measured simultaneously. Such devices largely enhance our capability of rationally evaluating the important fuel cell parameters including gas diffusivity, LCD and CP, all of which had typically been evaluated via the calculation of multiple additional parameters based on the famous Butler-Volmer equation $[9]$. However, the additional gas leak induced by the use of electrolyte discs was ignored in all the previous diffusivity measurement and this inevitably caused an inaccuracy in the diffusivity measurement and the subsequent evaluations of LCD and CP. In this report, the quantitative correlation between pressure uncertainty and the evaluation errors of fuel cell parameters is investigated. In addition, new devices with optimized gas tightness are designed to ensure efficient gas diffusivity measurement and accurate LCD and CP evaluation.

2. Theoretical analysis

The correlation between the gas diffusivity measurement error (ΔD) and the pressure uncertainty induced by gas leak is described in Eqs. (1) and (2),

$$
\Delta D_{\text{H}_2-\text{H}_2\text{O}}^{\text{eff}} = \frac{\text{RTl}_a \text{i}}{4F (p_{\text{H}_2}^{\text{o}} - \Delta p_{\text{H}_2}^{\text{i}})}
$$
(1)

$$
\Delta D_{\text{O}_2-\text{N}_2}^{\text{eff}} = \frac{\text{RTl}_{\text{c}}\text{i}}{8\text{F}(\Delta p_{\text{O}_2}^{\text{i}} - p_{\text{O}_2}^{\text{o}})}
$$
(2)

where R is the gas constant, F is the Faraday constant, T is the measurement temperature, l_a (l_c) is the anode (cathode) thickness, i is the current density, $p_{\text{H}_2}^{\text{o}}(p_{\text{O}_2}^{\text{o}})$ is the H₂ (O₂) pressure out of the measurement device and $\Delta p_{\rm H_{2}}^{\rm i}(\Delta p_{\rm O_{2}}^{\rm i})$ is the H_2 (O₂) pressure deviation induced by the gas leak [\[15\].](#page--1-0)

3. Results and discussion

According to Eqs. (1) and (2), the plot of diffusivity measurement error versus pressure uncertainty is shown in Fig. 1. For both anodes and cathodes, the measurement error of gas diffusivity increases with increasing pressure uncertainty induced by gas leak. For the four considered working temperatures, 650 °C, 700 °C, 750 °C and 800 °C, the diffusivity measurement error first increases slowly with increasing pressure uncertainty and then increases abruptly as the $\Delta p/p$ approaches 0.7. This suggests that the diffusivity measurement error can be significantly large as serious pressure uncertainty is caused by gas leak during gas diffusivity measurement. The temperature dependence of this correlation between diffusivity measurement error and pressure uncertainty appears to be random; for instance, for 800 °C the anode gas diffusivity measurement error is the largest among the four considered temperatures while for 800 \degree C the cathode gas diffusivity measurement error is the smallest among the four temperatures. Therefore, reducing gas leak is necessary for the gas diffusivity measurement in the full range of the considered fuel cell operating temperatures.

To analyze the evaluation error of LCD, the evaluation error as a function of pressure uncertainty is plotted according to Eqs. (3) and (4) , as shown in [Fig. 2.](#page--1-0)

$$
\Delta i_{\rm a} = \frac{4F p_{\rm H_2}^{\rm o} \Delta D_{\rm H_2-H_2O}^{\rm eff}}{RTl_{\rm a}}
$$
\n(3)

$$
\Delta i_{c} = \frac{8F p_{O_2}^{o} \Delta D_{O_2 - N_2}^{eff}}{RTl_{c}} \left(\frac{p_{t}}{p_{t} - p_{O_2}^{o}} \right)
$$
(4)

Fig. $1 -$ (a) Plots of anode gas diffusivity measurement error versus pressure uncertainty induced by gas leak (b) plots of cathode gas diffusivity measurement error versus pressure uncertainty induced by gas leak. Anode thickness is 750 μ m and cathode thickness is 200 μ m.

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