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Precise measurement of effective oxygen diffusivity for microporous media containing moisture by review of galvanic cell oxygen absorber configuration

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ABSTRACT

The performance of polymer electrolyte fuel cells is influenced by moisture control in their gas diffusion layer (GDL). Therefore, to achieve suitable control, it is necessary to clarify the mass transfer characteristics within a GDL by high precision measurement of oxygen diffusivity. We have previously proposed that measurement of the effective oxygen diffusivity in a GDL containing moisture can be achieved using a galvanic cell oxygen absorber and demonstrated this to be an effective technique for the measurement of microporous media. However, the diffusion resistance of a single dry GDL is low, so that the margin of error in the oxygen diffusivity measurement is high. In this study, high precision measurement of the oxygen diffusivity in a GDL was developed further by analysis of the major error factors and modification of the measurement apparatus configuration. The results indicate a reduction in the maximum measurement error from 50% to 20% for a dry GDL with minimal diffusion resistance.

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1. Introduction and summary of relevant former studies

The objective of this study is to improve the precision of effective oxygen diffusivity measurements for microporous media containing moisture, such as the gas diffusion layer (GDL) of a polymer electrolyte fuel cell (PEFC), using a galvanic cell oxygen absorber.

PEFCs are expected to be used as lightweight and compact power sources for vehicles, due to their high power densities and superior start-up characteristics at low operating temperatures. PEFCs have also become commercially available as domestic cogeneration systems. However, a reduction of the system cost and higher output power density are still required. Degradation of performance due to the amount of moisture is an especially important problem. For example, water or condensation from humidified vapor accumulates in the gas diffusion layer (GDL) and gas channels of the separator, which hinders diffusion of the reactant gas under conditions of high humidity and high current density. In contrast, proton conductivity in the polymer electrolyte film decreases when the cell becomes dry under low humidity conditions. Therefore, it is important to control the liquid water in PEFCs and to clarify the oxygen diffusion characteristics in a GDL containing moisture at the cathode side, where the influence of generated water is most significant. Thus, it is necessary to measure the effective oxygen diffusion coefficient for the GDL with a high degree of accuracy.

Various methods for measuring the effective diffusion coefficient in porous media have been previously reported. For example, the diaphragm method was used by Henry et al. [1] and Masamune and Smith [2]. This method is used to determine the effective diffusivity by directly measuring the gas permeability through a porous medium. Diluted gases with different concentrations are passed through the channels that sandwich the porous medium, and the difference in concentration between the upper and lower flows is measured. However, this method is complex and requires a considerable amount of time to obtain highly accurate measurements. A reaction–diffusion system was used by Wakao and Funaki [3], and a separation membrane method was employed by Hamai and Mitani [4] and Gibilaro and Waldram [5]. The reaction and adsorption methods were used to measure the rate of reaction or rate of adsorption of a gas on the surface of a porous medium. In addition, mass transport in a porous material was modeled on the basis of certain assumptions to determine the relationship between the mass transfer rate and the effective diffusivity. The effective diffusivity is quantified from the change in the measured carrier gas concentration with elapsed time. Accordingly, the

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Nomenclature

A	area of carbon electrode film [m^2]
C	mass transfer conductance [m^3/s]
D	oxygen diffusivity [m^2/s]
E	output voltage [mV]
F	Faraday constant [s A/mol]
I	output current [A]
J	mass flux of oxygen [kg/(m^2 s)]
P	pressure [atm]
R	mass transfer resistance [s/m^3]
S	average saturation in GDL [%]

Greek symbols

ρ	density [kg/m^3]
ω	mass fraction of oxygen [–]

Subscripts

ave	average
C	carbon electrode film
eff	effective
exp	experimental
Gas	gas
GDL	gas diffusion layer
Glv	galvanic cell
num	numerical
O_2	oxygen
1	effect of liquid surface difference
2	effect of distance between films

accuracy of the diffusion coefficient is strongly dependent not only on the measurement accuracy, but also on the simulation model employed. Further, an oxygen sensor with an yttria-stabilized zirconia (YSZ) electrolyte was employed by Mezedur et al. [6] to measure gas diffusivity in porous electrode catalysts based on oxygen ion transfer in the YSZ electrolyte. The effective oxygen diffusivity was determined by simultaneously measuring the permeating oxygen flux in the porous medium and the oxygen concentration at its surface. However, this sensor is generally used at temperatures above 300 °C, and it is difficult to simply and precisely measure gas diffusivity under wet conditions where liquid water is present in the micropores.

Recently, the effective diffusivity of the GDL in PEFCs was experimentally measured. Kramer et al. [7] and Flückiger et al. [8] obtained the effective diffusivity using electrochemical diffusimetry. Electrochemical impedance spectroscopy was applied to measure the effective ionic conductivity of an electrolyte-soaked GDL, and the effective diffusivity, which was expressed as the ratio of the bulk diffusion coefficient to the diffusion coefficient for the GDL, was determined by taking advantage of the analogy between Fick's law and Ohm's law. The effective diffusivity in a dry GDL was measured by Baker et al. [9] and that in a wet GDL by Hwang and Weber [10] using the limiting current density technique. Hwang and Weber [10] measured the effective diffusivity of hydrogen through argon in a binary mixture with a sample containing moisture. Zamel et al. [11] and Chan et al. [12] measured the effective diffusivity in a dry GDL using a modified Loschmidt cell and applying the one-dimensional Fick's law. However, there have been few studies that have measured the effective diffusivity in a sample containing moisture.

Through research on water management in PEFCs, the authors have developed a method to measure the oxygen diffusivity of microporous media containing moisture [13,14]. Utaka et al. [13] evaluated the gas diffusion characteristics of a dry GDL using a method that employed a galvanic cell oxygen absorber, where multi-layered GDLs with a larger diffusion resistance were used to obtain the precise oxygen diffusivity. The galvanic cell oxygen absorber was uniquely designed by upgrading a galvanic cell oxygen sensor that is practically used to detect the oxygen concentration and measure a comparatively large oxygen flux. However, diffusivity measurement of a single GDL containing moisture is required under conditions similar to those during practical operation. The galvanic cell oxygen absorber with peripheral equipment and the measurement method were further improved to evaluate the oxygen diffusion characteristics in a single GDL with/without the presence of moisture [14]. This method was demonstrated to be effective for measuring a single GDL containing moisture and the measurement error factors were also examined. However, further improvement of the diffusivity measurement precision is necessary, especially for investigating dry single GDLs that have the lowest diffusion resistance to moisture saturation. It is important to consider the effect of the concentration distributions in the vicinity of both GDL surfaces due to a reduction of mass transfer resistance in the GDL during a single GDL measurement. However, it is difficult to correctly measure the concentration distributions at both surfaces of the sample. Therefore, a special method was required for deriving the diffusion coefficient with sufficient accuracy. The authors have previously developed a comparison method [14], where the apparent mass transfer conductance of a standard test sample with a known diffusion coefficient is measured consecutively under the same experimental conditions with and without the sample. Using the standard sample measurement, the effective oxygen diffusion coefficient for the test sample was determined by adopting the bulk characteristics of oxygen reaction in the galvanic cell oxygen absorber as a variation of the effective diffusion resistance. Furthermore, this method was applied to measure the diffusivity in a specially devised GDL containing moisture [15]. In such an evaluation, error analysis indicated that stability of the output characteristics of the galvanic cell oxygen absorber was the most important issue for achieving high precision measurement of the oxygen diffusivity with the comparison method. Heretofore, the measurement precision with this method was guaranteed by using only results obtained when the change in the output of the galvanic cell oxygen absorber under the same conditions was low [14]. However, there was still a limitation on the measurement accuracy. The measurement results for the effective diffusivity in a dry GDL had a spread of ±50% around the average value, while there a lower data spread for the diffusivity in a moist GDL with larger diffusion resistance.

In this study, those factors that should be improved for output stability of the galvanic cell oxygen absorber are examined and the diffusivity measurement accuracy is improved by upgrading the configuration of the galvanic cell oxygen absorber measurement apparatus.

2. Galvanic cell oxygen absorber and effective oxygen diffusivity measurement

2.1. Fundamentals of galvanic cell oxygen absorber apparatus

Fig. 1 shows a schematic diagram of the experimental galvanic cell oxygen absorber apparatus used to measure the effective

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