Water Transport Analysis in a Polymer Electrolyte Electrolysis Cell Comprised of Gas/Liquid Separating Interdigitated Flow Fields

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Abstract

A novel interdigitated flow field design for polymer electrolyte electrolysis cells (proton exchange membrane water electrolysis cells) composed of oxygen exhaust channels apart from liquid water feed channels has been developed for ground and space applications because the design is advantageous in terms of oxygen/water separation without buoyancy, and dispenses with water circulators for bubble removal in a cell and external separators by natural or centrifugal buoyancy. Finite element modeling of water transport in the polymer electrolyte (proton exchange) membrane in a cell with the interdigitated flow fields is conducted. Current-voltage (I-V) measurement of the cell is also performed for comparison with numerical modeling. Deviation of the experimental I-V characteristics from those of the numerical model indicates a possible water transport path in the in-plane direction of hydrophobic microporous layers (MPLs) coated on gas diffusion layers installed between the anode catalyst layers (CLs) and oxygen flow channels in the cell. Analysis of the deviation associated with the limitation of water transport also suggests fractional bubble coverage of produced oxygen gas at the CLs. The hydrophobic MPL acts to separate oxygen gas and pressurized liquid water due to the capillary pressure, while it determines the limitation of water transport to the CLs with the oxygen bubble coverage.

Keywords— Water electrolysis, Gas/liquid separation, Interdigitated flow field, Microporous layer
Intr
oduction

Water electrolysis has been studied for hydrogen production using renewable energy to reduce environmental carbon dioxide emissions. Polymer electrolyte electrolysis cells (PEECs) (proton exchange membrane water electrolysis cells (PEMWEs)) have been promising for water electrolysis in terms of corrosion, energy efficiency, their size, and the purity of the hydrogen gas produced.1–3

In conventional water electrolysis, oxygen is produced in liquid water supplied to the anode as bubbles containing water vapor while hydrogen produced conveys a large amount of electroosmotic drag water transported with protons from the anode to the cathode through the membrane. The water can be removed by condensation and gravitational force. However, to use hydrogen for energy resources, the condensation and removal of water involve energy loss, and a large volume of energy-consuming facilities to remove water such as chillers is required.

In addition to ground applications, space applications of water electrolysis including closed-loop air revitalization have been developed for space missions4–8 such as the international space station (ISS) and manned Moon/Mars exploration projects, in which the water management, however, becomes more complicated. The primary product required from water electrolysis is oxygen in this case. The oxygen is released to the cabin; thus, there is a requirement to minimize contamination with water. Cathode-feed water electrolysis for closed environments under microgravity conditions has thus been developed, by which relatively dried oxygen can be produced at the anode side using water diffused through the membrane from the cathode side.9

We have so far developed a novel flow channel design with an interdigitated configuration composed of hydrogen/oxygen exhaust flow channels that are apart from the liquid water feed flow channels, as shown in Fig. 1.10–12 The anode and cathode CLs and flow channels oppose each other with the PEM between. Pressurized water is fed perpendicularly to the anode side of the polymer electrolyte (proton exchange) membrane (PEM) through the flow channels, humidifying the PEM and being followed by injection into the adjacent anode catalyst layers (CLs) in contact with microporous layer (MPL)-coated gas diffusion layers (GDLs) connected to the anode gas (oxygen) flow channels. Pressurized water can
provide an advantage to reduce the humidity of the product oxygen for practical use under atmospheric pressure by producing pressurized oxygen.

The proposed cell is feasible to separate and transport hydrogen and oxygen gases produced to the outside of the cell without gravity-assisted gas/liquid separation (i.e. buoyancy) due to the hydrophobicity of the MPLs, which have strong liquid water support force with capillary pressure,\textsuperscript{10,13,14} so that water circulators for bubble removal and conveyance in a cell by drag force with liquid water flow\textsuperscript{15} used for conventional water electrolyzers can be dispensed with. External separators with natural or centrifugal buoyancy can also be omitted. This cell can thus be applied to hydrogen and oxygen production in the microgravity environments of space missions.

In the present study, for fundamental information to develop optimized designs of the cell components, numerical modeling is conducted for the water transport in a PEM assembled with interdigitated flow fields, being coupled with electrochemical reactions, with the aid of measurement of current-voltage (I-V) characteristics. Thereby, water transport through the MPL is indicated.

1 Experimental

Electrochemical characterization was conducted with an in-house PEEC of bipolar plates (BPPs) made of titanium with the interdigitated oxygen gas/liquid water and hydrogen gas flow fields illustrated in Fig. 1.

A ca. 50 $\mu$m thick Nafion 212 membrane was sandwiched between the anode and cathode gas diffusion electrodes (GDEs), as reported elsewhere,\textsuperscript{11} where an anode CL of IrO$_2$ paste ink (Chemix Co. Ltd.) was sprayed onto the MPL side of an MPL-coated GDL (SIGRACET® 29BC, SGL Carbon Inc.). The amount of IrO$_2$ loaded on the anode was 1.5 mg cm$^{-2}$. A cathode CL was also attached onto the MPL side of the GDL (SIGRACET® 29BC, SGL Carbon Inc.) by a transfer method to prepare the GDE.\textsuperscript{11} A loaded amount of Pt on the cathode was 1.0 mg cm$^{-2}$. A cross-sectional drawing of the cell configuration is shown in Fig. 2.
The geometrical active area of the anode and cathode CLs was 15.5 cm$^2$ each. They were both composed of five CLs with a width of 5 mm and opposed each other with the PEM between. To avoid the excess leakage of the water injected into the anode water flow channels to the O$_2$ gas flow channels, the edge of the anode GDE was sealed with silicone putty (Cemedine Co. Ltd., 8051 N) diluted in hexane.

The cell was characterized by I-V measurements using a potentiostat/galvanostat (Hokuto Denko Co., HA-151B) under potentiostatic mode (voltage control) with the four-terminal method. The temperature of the cell was maintained with an oven for the I-V measurements. Water was injected into the anode flow channels at 50 kPaG by pressurized N$_2$ gas introduced into a water reservoir. The gases produced were exhausted through the outlet ports of the cell without controlled backpressure.

2 Numerical Model

The COMSOL Multiphysics (4.3a, 5.4) packages were used for finite element modeling of the coupling processes involved in the electrochemical energy conversion.\textsuperscript{16, 17} The mass and charge balances were applied on a three-dimensional domain of the PEM shown in Fig. 1, with a focus on water transport in the membrane as a starting point.

To reduce the computational cost, a quarter of the cell was employed on the basis of geometrical symmetry. The complicated heat balance of the system was also omitted by assuming isothermal operation at an experimental cell temperature. The model assumes a system of steady-state for the mass transfer of dissolved water in the membrane.

2.1 Mass Balance

Water transport in the membrane is modeled with Fick’s law, where the water flux by diffusion is considered with the following water diffusion coefficients (m$^2$s$^{-1}$) at a cell temperature, $T$.\textsuperscript{18, 19}
\[ D_{\text{H}_2\text{O}}^{\text{PEM}} = 3.1 \times 10^{-7} \lambda (\exp(0.28\lambda) - 1) \exp \left( \frac{-2436}{T} \right) (0 < \lambda < 3) \]  

(1)

\[ D_{\text{H}_2\text{O}}^{\text{PEM}} = 4.17 \times 10^{-8} \lambda (161 \exp(\lambda) + 1) \exp \left( \frac{-2436}{T} \right) (3 \leq \lambda < 17) \]  

(2)

where water uptake,

\[ \lambda = \frac{\text{EW}}{\rho_{\text{PEM}} C_{\text{PEM} \text{H}_2\text{O}}} \]  

(3)

is related to the water concentration in a PEM, \( C_{\text{PEM} \text{H}_2\text{O}} \), with the equivalent weight of a PEM, EW, of 1100 g eq\(^{-1}\) and a dry density, \( \rho_{\text{PEM}} \), of 1970 kg m\(^{-3}\) for Nafion 212.

Water flux with electroosmosis, \( j_{\text{H}_2\text{O}}^{\text{os}} \), associated with the proton electromigration under an electrical potential gradient \( \nabla \phi \) in the membrane is also included in the model:\(^{19,20}\)

\[ j_{\text{H}_2\text{O}}^{\text{os}} = \frac{n_d \lambda \sigma (\lambda = 16)}{16 F} \nabla \phi \]  

(4)

The electroosmotic drag coefficient, \( n_d \), of 2.5 at \( \lambda = 16 \) for a fully hydrated PEM in contact with liquid water is employed, although there has been variation among reports.\(^{21}\)

For each species, the following expression is solved for the PEM depicted in Fig. 1.

\[ \nabla \cdot j_{\text{H}_2\text{O}} = S_{\text{H}_2\text{O}} \]  

(5)

where \( j_{\text{H}_2\text{O}} \), and \( S_{\text{H}_2\text{O}} \) denote the summation of the diffusive and electroosmotic mass flux vectors of water and the source term, respectively.

A boundary condition was employed for the PEM surface in contact with the water channels, where a water concentration of \( 2.8 \times 10^4 \) mol cm\(^{-3}\), which corresponds to a \( \lambda \) of 16 in Eq. 3, is assumed to be established in the fully hydrated membrane in contact with liquid water.

We also model with a constant water concentration of the fully hydrated membrane over its entire region to reproduce a case without a limitation of water transport, assuming a free liquid water transport.
pathway among the CLs and water flow channels along the membrane surface.

### 2.2 Charge Balance

The ionic charge transfer with protons in the PEM is modeled with Ohm’s law employing the following proton conductivity \((\text{Sm}^{-1})\)\(^{22,23}\) in accordance with its temperature and water uptake dependence.\(^{24}\)

\[
\sigma(\lambda, T) = \exp \left[ 1268 \left( \frac{1}{303} - \frac{1}{T} \right) \right] (0.514\lambda - 0.326)
\]  

(6)

The porous anode and cathode CLs are simplified as two-dimensional layers on the PEM, where the electrochemical charge transfer is represented by the common kinetic equation as follows, which is reduced to the Butler-Volmer equation for the constant concentrations of the reactant and product species assumed below.

\[
i = i_0 \left\{ \frac{c_i^e}{c_i^{\text{in}}} \exp \left( \frac{\beta nF\eta}{RT} \right) - \frac{c_j^e}{c_j^{\text{in}}} \exp \left( -\frac{(1 - \beta) nF\eta}{RT} \right) \right\}
\]

(7)

where \(i\) (A cm\(^{-2}\)), \(i_0\) (A cm\(^{-2}\)), \(n\), \(\beta\), and \(\eta\) (V) are the current density, the exchange current density, the number of electrons, the symmetry factor, and the mixed (activation and concentration) overpotential,\(^{16,25,26}\) respectively.

\(c_i^{\text{in}}\) (mol m\(^{-3}\)) and \(c_i^e\) (mol m\(^{-3}\)) denote the concentration of the reactant species \(i\) at the CL surface for the initial state (equilibrium potential) and electrolysis, respectively, while \(c_i^{\text{in}}\) and \(c_j^e\) represent the concentration of the product species \(j\) for the initial state (equilibrium potential) and electrolysis, respectively.

At the anode, \(c_i^{\text{in}} = 2.8 \times 10^4\) mol cm\(^{-3}\) for the fully hydrated PEM in contact with liquid water, while \(c_j^e/c_j^{\text{in}} = 1\) with the assumption of a constant oxygen partial pressure. At the cathode, \(c_i^e/c_i^{\text{in}} = 1\) for constant proton concentration in the PEM and \(c_j^e/c_j^{\text{in}} = 1\) with the assumption of a constant hydrogen partial pressure.

At the anode, the oxygen bubbles produced would partially cover the CLs and limit the liquid water.
supply, reducing the active area. Hence, as an approximation on the basis of earlier studies,23,27 the fractional coverage with oxygen bubbles is included in the exchange current density as a total volumetric fraction of the gas phase at an anode MPL, $\epsilon_{MPL} s_{G_{MPL}}$, where gas saturation, $s_{G_{MPL}}$, is defined as the volumetric ratio of the gas phase in the pore volume of the anode MPL with a porosity of $\epsilon_{MPL}$ adjacent to the anode CL in the form of liquid saturation, $1 - s_{G_{MPL}}$:

$$i_0 = \epsilon_{MPL}(1 - s_{G_{MPL}}) \gamma_M \exp \left[ \frac{E_{A,a}}{R} \left( \frac{1}{T_{\text{ref}} - 1} \right) \right] i_{0,\text{An,Ir}}^{\text{ref}}$$

with $\gamma_M$, roughness factor, $E_{A,a}$ (kJ mol$^{-1}$), anode activation energy, $T_{\text{ref}}$, reference temperature, and $i_{0,\text{An,Ir}}^{\text{ref}}$, anode exchange current density at the reference temperature. We assume $s_{G_{MPL}} = 0$ in the present model, while $s_{G_{MPL}}$ is estimated from comparison between experimental and numerical I-V characteristics.

Unlike the anode, $s_{G_{MPL}}$ can be ignored in the cathode because the protons are reactant species conducted from the PEM, and the hydrogen gas produced does not limit the reactant supply. Hence

$$i_0 = \gamma_M \exp \left[ \frac{E_{A,c}}{R} \left( \frac{1}{T_{\text{ref}} - 1} \right) \right] i_{0,\text{Ca,Pt}}^{\text{ref}}$$

where $E_{A,c}$ (kJ mol$^{-1}$) is the cathode activation energy, and $i_{0,\text{Ca,Pt}}^{\text{ref}}$ is the cathode exchange current density at the reference temperature.

The concentration of dissolved water in the PEM is introduced into the Butler-Volmer equation for the mass balance coupled with the charge balance, so that the model clarifies the impact of the concentration gradient in the PEM that develops among the CLs and the water inlet boundaries at the PEM surface in contact with the water flow channels on the kinetics of the oxygen evolution reaction. Table 1 summarizes the parameters for the activation overpotential and exchange current density.27

The overpotential in the Butler-Volmer equation is described as

$$\eta = \phi_a - \phi_l - \phi_{eq}$$
where $\phi_s (V)$ and $\phi_l (V)$ are the potentials of the CLs and the PEM. The cell voltage is defined as the difference of $\phi_s$ between the anode and cathode. Meanwhile, $\phi_{eq} (V)$ represents equilibrium potentials for the anode, $\phi_{eq,a}$, and cathode, $\phi_{eq,c}$, holds the following Nernst equation with hydrogen and oxygen partial pressures, $p_{H_2}$ and $p_{O_2}$, respectively,

$$\phi_{eq,a} - \phi_{eq,c} = \phi^{0,\text{liq}}(T) + \frac{RT}{2F} \ln \frac{p_{H_2}p_{O_2}^{0.5}}{a_{H_2}^{\text{liq}}a_{O_2}^{0.5}}.$$  \hspace{1cm} (11)

where $p_{H_2}/p^0 = 1$ and $p_{O_2}/p^0 = 1$ for a pressure, $p^0$, of chosen standard states (10^5 Pa or 1 atm, in general) with liquid water activity of $a_{H_2O}^{\text{liq}} = 1$ equilibrated with saturated vapor at $T$, being associated with a standard (formal) redox potential chosen for the standard states with liquid water, $\phi^{0,\text{liq}}(T)$.\textsuperscript{28,29} The dependency of the Gibbs free energy change on the pressurization of the liquid water is neglected.

3 **Results and discussion**

3.1 **Water electrolysis test**

Potentiostatic (voltage-controlled) I-V curves for the cell are shown in Fig. 3. The onset of electrolysis appears around 1.2 V, where the cell voltages are approaching equilibrium potentials with evolved oxygen and hydrogen. With elevation of the temperature, the onset voltage dropped possibly due to decreased ohmic and activation overpotentials, and the Gibbs free energy change.

The I-V characteristics exhibit steeper increases in the cell voltage for the larger current densities in the large current density regions, which can be ascribed to the limitation of water transport to the anode CLs.

3.2 **Numerical Analysis**

Figure 4 presents I-V characteristics from the finite element modeling for the water transport in the PEM and the fully hydrated membrane with the experimental I-V curves. The full hydration model assumes a
free liquid water transport pathway among the anode CLs and water flow channels along the membrane surface.

The three-dimensional numerical model employing water transport in the membrane gives extremely smaller currents than the experimental currents at the same cell voltages. The numerical model exhibits significant current restriction to the edge part of the CLs facing the water inlet boundary at the anode PEM surface in Fig. 5(a), which is associated with water depletion at the anode CLs in Fig. 5(b). Thus the water transport in the in-plane direction through the membrane does not play a main role in the experimental I-V characteristics due to the small cross-section of the thin membrane.

On the other hand, the I-V characteristics in Fig. 4 for the numerical model of the fully hydrated membrane at the constant water concentration show larger currents without a water transport limitation than the experimental I-V characteristics at the same cell voltages. Water transport pathways other than the membrane are thus indicated in the experimental cell, which involves water transport limitation to some extent. The pressurized water was possibly injected into the anode CLs through the gasket surface and incomplete sealant between the water flow channels and GDLs adjacent to the oxygen flow channels as well as the hydrophobic MPL in contact with the anode CLs shown in Fig. 2. This results in intrusion against the capillary pressures of the MPL pores, although appreciable water leakage to the oxygen flow channel was not observed. Once the water penetrated the gasket surface and MPLs, a much smaller pressure gradient may have been required for water transport until the capillary pressure at the interface between accumulated oxygen bubble and liquid water developed in the MPL pores.

The deviation between the experimental I-V characteristics and those of the fully hydrated membrane model, in which $s_{G}\text{MPL}$ is assumed to be 0, suggests the limitation of water transport at the anode CLs due to coverage with oxygen bubbles. Thus, $s_{G}\text{MPL}$, which represents the fractional coverage at the CLs, was estimated from the current ratios above the thermoneutral voltage using Eq. 9 at $\epsilon_{\text{MPL}}$ of ca. 0.7 for the GDL, SIGRACET® 29BC (SGL Carbon Inc.) as depicted in Fig. 6.

According to earlier reports of the liquid water saturation in an MPL of around 0.5 at a liquid water
pressure of 50 kPa against gas phase, $s_{MPL}^G$ of 0.5 to 0.6 at the smallest current densities seems reasonable. An increase in the current leads to an increase in the anode gas saturation, that is, fractional oxygen bubble coverage at the anode CLs with a decrease in the liquid water saturation, which results in the further limitation of the water transport to the CLs. Elevation of the temperature appears to suppress the water transport limitation with the gas saturation, as indicated by neutron imaging analysis for a porous transport layer of sintered titanium powder by Lee et al.\textsuperscript{31} The membrane hydration with water vapor\textsuperscript{32} evaporated from the intruded liquid water in the anode GDLs will be evaluated for a part of the anode CLs uncovered with liquid water in our future work.

The design of the MPL in terms of the hydrophobicity and pore structure for control of the gas saturation and pressure control of the fed water against the capillary pressure are thus key challenges to lower the electrolysis voltage (overpotential) while maintaining oxygen gas/liquid water separation, which would be particularly advantageous in microgravity environment without buoyancy.

4 Conclusions

Water transport in a PEM of a PEFC with interdigitated flow fields that separates oxygen gas exhaust and liquid water supply for ground and space applications was numerically modeled to analyze the experimental I-V characteristics of the cell. As a result, a possible water transport pathway from the liquid water channels to the anode CLs through the gasket part and the MPL was suggested. The hydrophobic MPL provides the gas/liquid separation device with pressurized water feed, whereas temperature-dependent limitation of the water transport to the CLs results possibly by an increase in the fractional oxygen bubble coverage at the CLs. Optimized hydrophobicity and the porous structure of the MPL, as well as optimized pressure control of the fed water against the capillary pressure, can be used to control the fractional oxygen bubble coverage to lower the electrolysis voltage while maintaining oxygen gas/liquid water separation in an environment without buoyancy.
Acknowledgements

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Table 1. Parameters for the kinetic model

| Symbol  | Value       | Unit                    | Comments and Reference                                      |
|---------|-------------|-------------------------|-------------------------------------------------------------|
| $R$     | 8.314       | J K$^{-1}$ mol$^{-1}$   | Universal gas constant                                      |
| $n\beta$ | 1           | -                       | (Charge) transfer coefficient term                           |
| $F$     | 96485       | C mol$^{-1}$            | Faraday constant                                            |
| $\gamma_M$ | 150         | -                       | Roughness factor                                            |
| $E_{A,a}$ | 76          | kJ mol$^{-1}$           | Anodic activation energy                                    |
| $E_{A,c}$ | 18          | kJ mol$^{-1}$           | Cathodic activation energy                                  |
| $T_{\text{ref}}$ | 353         | K                       | Reference temperature used for exchange current density      |
| $i_{\text{ref},\text{An},\text{Ir}}$ | $1 \times 10^{-7}$ | A cm$^{-2}$ | Anode exchange current density at 353 K                      |
| $i_{\text{ref},\text{Ca},\text{Pt}}$ | $1 \times 10^{-3}$ | A cm$^{-2}$ | Cathode exchange current density at 353 K                    |
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Current collector

Pressurized water

50 μm

250 μm

2 mm

2 mm

Current collector

Water

Current collector

Gasket

MPL

GDL

H₂ gas

2 mm

2 mm

Catalyst layer (5 mm width)

Pressurized water

O₂ gas

Sealant

250 μm

2 mm

2 mm

Current collector

Current collector

MPL

GDL
Voltage-controlled

Voltage (V)

Current density (A cm\(^{-2}\))

25°C (RT)
40°C
60°C
80°C
(a) Anode catalyst layer

(b) Water inlet boundary
