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Effect of the hydrophilic and hydrophobic characteristics of the gas diffusion medium on polymer electrolyte fuel cell performance under non-humidification condition

Heesung Park *

Research and Development Division, Hyundai-Kia Motor Company, South Korea

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ABSTRACT

Water is a significant component of polymer electrolyte fuel cells, affecting the proton conductivity in the membrane electrolyte. Therefore, polymer electrolyte fuel cells are generally operated with a humidifier to maintain a high relative humidity of the supplied gases; however, the humidifier contributes additional weight and cost. Although many studies have attempted to develop polymer electrolyte fuel cells without a humidifier, the studies have been mainly focused on the self-humidified membrane electrolyte and catalyst layer. In this paper, the author investigates the effect of polytetrafluoroethylene coated gas diffusion medium on the water content in the membrane electrolyte. The water condensation on the surfaces of the gas diffusion medium is visualised when the polymer electrolyte fuel cell is operated under non-humidification conditions. Numerical simulation suggests that the optimum water saturation is between 0.1 and 0.3 at the gas diffusion medium to hydrate the membrane electrolyte sufficiently without significantly blocking the diffused species under non-humidification conditions.

1. Introduction

Increasing pollution and diminishing fossil fuel resources have led to a movement toward more environmental and efficient power sources [1]. The new technologies of wind, solar, geothermal, hydro, and tidal energy have drawn attention to scientific and engineering societies [2]. Among the new energy technologies, the fuel cells have been widely employed for the portable electrics power supply [3], residential cogeneration system [4], grid power system [2,5], and combined heat, power, and hydrogen production system [1]. Besides, polymer electrolyte fuel cell (PEFC) is a potential alternative technology for internal combustion engines in automotive applications for emission-free sustainable mobility with high power density [6,7]. In PEFCs, the electrical current generated from the electrochemical reactions between hydrogen and oxygen in the catalyst layers provides power to the application systems. To this end, the PEFCs are operated under high electric current density, and the product water tends to be a liquid phase, which is prone to obstructing the reactant gases from being transported to the catalyst layers. Meanwhile, the hydration of the membrane electrolyte is required to increase the proton conductivity [8,9]. To keep the polymer electrolyte hydrated, effective humidity control methods for the reactant gases have been proposed [10–13]. For example, hydrophilic constituents in the microporous gas diffusion layer were suggested to enhance water diffusion and provide preferential pathways for the liquid water [12]. On the other hand, high temperature PEFC was studied with the benefits of simpler heat and humidity managements [14]. Although external humidifiers are generally used to supply adequate humidity in PEFCs, they have critical drawbacks, such as additional cost, weight, and parasitic power loss. It is highly desirable to develop the PEFCs without an external humidifier. In this regard, numerous studies have been conducted to allow PEFCs to operate under low humidification or self-humidification. The focuses of previous studies can be categorised into the recirculation of exhaust reactants, self-humidified membrane electrolytes, and gas diffusion media (GDMs). Yang et al. [15] investigated the effect of the circulation flow rate and exhaust amount of reactant on the performance of PEFCs without humidification. Wan et al. [16] proposed the use of a condenser to recover enough water to ensure the adequate humidity of the supplied gases. The PEFC performance was estimated to be only 6.75% lower with the use of recirculated exhaust air than with a humidifier [17]. Although recirculation is effective without discharging the product water, it still needs additional power to recirculate. Other studies have focused on self-humidifying membranes that could retain the product water

* Address: 104, Mabuk-Dong, Giheung-Gu, Yon-In City 446912, South Korea. Tel.: +82 10 2690 3461.
E-mail addresses: heesung.park@hyundai.com, heros93@gmail.com

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Nomenclature

a	water activity
A	Tafel slop
C_r	condensation rate, s^{-1}
C_i	molar concentration of species i, kmol m^{-3}
D_c	confined distance, m
D_i	diffusivity of species i, m^2 s^{-1}
F	Faraday constant, 96,485 C mol^{-1}
I	electric current density, A m^{-2}
I^{tr}	volumetric transfer current, A m^{-3}
j	exchange current density, A m^{-2}
P	pressure, Pa
P_c	capillary pressure, Pa
R	gas constant, 8.314 J mol^{-1} K^{-1}
R_{ohm}	ohmic resistivity, Ω m
s	liquid water saturation
S	source term
T	temperature, K

Greek symbols

γ	surface tension, J m^{-2}
ε	porosity
η	overpotential, V
λ	membrane water content
μ	dynamic viscosity, kg s^{-1} m^{-1}
ρ	density, kg m^{-3}
φ	electric potential, V

Superscripts and subscripts

an	anode
ca	cathode
eff	effective value
l	liquid water
mem	membrane
ref	reference value
sat	saturation
sol	solid

to maintain the humidity at the membrane electrolyte. The hygroscopic materials were added to the membrane electrolyte [18–21] or catalyst layer [22–24], and yet the membrane electrolytes can be only achieved under restricted operating conditions.

In terms of reactant gas diffusion, a large number of studies have been conducted because the GDM plays a significant role in hydrogen and air transport in addition to water management [25]. The presence of liquid water in GDM reduces the gas diffusion and results in losses [26]. In this regard, the liquid water in PEFC has been visualised by using optical microscopy [27–31], neutron radiography [26,32–38], and X-ray imaging [39–41]. Fishman and et al. [27] studied the liquid–solid–gas interfacial phenomena on the variable GDMs by using optical microscopy. They showed that the contact diameter pinning behaviour during evaporation is correlated to the material topography. Owejan et al. [32] investigated the liquid water distribution in operating fuel cells by using the neutron radiography method. They suggested that flooding within the electrode layer or at the electrode-diffusion media interface is the primary cause of the significant voltage loss. Park et al. [33] visualised the liquid water distribution in PEFC and showed that the PEFC performance was strongly affected by the presence and accumulation of liquid water, especially at high electrical current densities. Hartnig et al. [39] investigated the evolution of liquid water and its transport in GDM by using synchrotron X-ray radiography. They explained that the observed liquid water in GDM was condensed on the hydrophilic spots. Although these visualisation techniques have provided detailed insight in GDM with regard to the nature of water transport inside PEFCs, however, most of the previous studies have focused on the high external humidification conditions.

Under non-humidification conditions, the Kitahara research team have studied the pore size and polytetrafluoroethylene (PTFE) content of GDM [42] and suggested a hydrophilic and hydrophobic double microporous layer [43] to effectively retain the humidity at the catalyst layer. Chen and Chang [13] proposed a composite carbon black material to manufacture GDMs with microporous layers that can effectively preserve high humidity level in the reaction sites. Nonetheless, only a few studies have investigated the characteristics of GDMs in detail for operation under non-humidification conditions. The mechanisms behind the improvement of the hydration of the membrane electrolyte or deterioration of the hydrogen and air transport to the catalyst layers by the hydrophilic and hydrophobic characteristics of GDMs have yet to be explained.

In the presence of water, the hydrophilic and hydrophobic characteristics of the GDM can significantly affect PEFC performance. Berning et al. [44] reported the irreducible saturation that accounts for discontinued liquid phase regions [45] or a fraction of hydrophilic pores that ranged from 22% [46] to 85% [47] in GDMs. The initial liquid water saturation by nucleation within GDM was assumed to numerically evaluate the overall water saturation level in the GDM [48]. The numerical simulation proposed a pore network pattern arising from the water condensation at nucleation site within bulk of GDM. Although the model did not predict the liquid water nucleation, the study provided insight in terms of water saturation caused by nucleation within GDM. On the other hand, the existence of heterogeneous surfaces due to defects or cracks in hydrophobic carbon could induce water condensation [49]. With these, it is highly plausible to cause water condensation at the surfaces of GDM which is composed of carbon fibres [50]. Moreover, in ambient conditions with a relative humidity of 40%, heterogeneous water condensation can be initiated where the confined distance is approximately 1 nm [51]. In general, PEFCs are operated with activation process to optimise the electrode structure and improve the utilisation of catalyst in addition to hydrate the membrane [52]. During the activation procedure, the generated water from the electrochemical reaction can be condensed at the GDM surfaces where defects and cracks inevitably present in the nano/micrometre scale.

In this paper, water condensation was visualised in the experiment under non-humidification condition after finishing the activation process which was defined to apply a constant voltage of 0.6 V for 3 h. The experimental observations revealed that the volume fraction of liquid water within GDM were 0.15 and 0.62 depending on the PTFE content. It was found that the liquid water distribution was strongly dependent on the hydrophilicity of GDM rather than the gas flow especially on the non-humidification condition. Numerical simulations are useful to investigate the PEFC performances that are linked with many physical and electrochemical parameters [53]. By conducting numerical simulations with considering heterogeneous water condensation in the PEFC, it was suggested that the optimal water saturation was between