



# Effect of Catalyst Layer Ionomer Content on Performance of Intermediate Temperature Proton Exchange Membrane Fuel Cells (IT-PEMFCs) under Reduced Humidity Conditions



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## ABSTRACT

For intermediate-temperature polymer electrolyte membrane fuel cells (IT-PEMFCs), the effects of the cathode gas flow rate and the ionomer content are experimentally examined at 120 °C under conditions of low relative humidity (RH). The IT-PEMFC operation at low RH should be beneficial in developing compact systems with smaller humidifiers. First, analysis of the effect of gas flow rate at various current densities confirms that drying of the membrane electrode assembly (MEA) is an important factor in IT-PEMFC operation, whereas cathode flooding becomes significant in regions of high current density with low flow rates. Then, MEAs with various contents of Aquivion<sup>TM</sup> ionomer are fabricated, and the combined effect of drying and flooding is further investigated by IT-PEMFC tests at various RH conditions and current densities. The optimum ionomer content increases with decreasing current density at 20% RH or below, indicating that MEA drying becomes dominant over cathode flooding.

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## 1. Introduction

Polymer electrolyte membrane fuel cells (PEMFCs) have been extensively studied as efficient energy conversion devices utilizing gaseous hydrogen and oxygen to generate electricity [1–3]. Since the electrochemical conversion of chemical energy to electric energy provides significantly higher thermodynamic efficiency (>40%) than conventional internal combustion engines (20–30%), various types of PEMFCs have been investigated, including those operated at low (~80 °C), intermediate (100–120 °C), and high

temperatures (up to 200 °C) for practical PEMFC deployment [2]. Among the different PEMFCs studied, intermediate-temperature PEMFCs (IT-PEMFCs) [1] have received much attention due to their superior heat/water management, CO tolerance, and electrode reaction kinetics [2,3] than the low temperature (LT-) PEMFCs. However, the reliability and performance of IT-PEMFCs should be further enhanced to be utilized commercially as portable and stationary power sources.

An IT-PEMFC employs a membrane electrode assembly (MEA), composed of anode and cathode catalyst layers, a polymer membrane electrolyte, and gas diffusion layers to conduct the electrochemical redox reactions with appropriate electronic and ionic conductivities and molecular mass transport. Several polymer membrane electrolytes have been reported, e.g. short-side-chain perfluorinated sulfonic acid (SSC PFSA) [4–6], sulfonated hydrocarbon polymer [7–10], polymer/inorganic composites [11–14], etc. The PFSA polymers with long side chains, such as Nafion<sup>®</sup>, are conventionally used in LT-PEMFCs (60–80 °C) [15], but they do not have stable function in IT-PEMFCs due to poor thermal stability at a glass transition temperature <110 °C [16].

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The catalyst layer, which consists of the metal catalysts and the ionic conductors, is a key component of PEMFC MEAs. Since the hydrogen oxidation reaction (HOR) and the oxygen reduction reaction (ORR) essentially occur in the catalyst layer, involving mass transport of reactants and products, the catalyst layer structure and properties should be optimized to achieve high PEMFC performance and stability. Therefore, the catalyst layer structure in LT-PEMFCs have been intensively researched for decades, focusing on various variables, including ionomer content [17–22], catalyst loading [23], catalyst layer structure [21,24–26] and thickness of catalyst layer [27]. Especially, the optimum ionomer loading and the resultant microstructure should be determined in terms of the preparation methods, catalyst, and ionomer materials under desired cell operating conditions. Generally, proton conductivity in catalyst layers increases with increasing ionomer loading; however, water removal rate decreases simultaneously, leading to significant flooding. As the proton conduction and water removal characteristics are strongly dependent on the operating conditions, such as RH and current density, the optimum ionomer loading should be discussed for specific operating conditions. For example, Kim et al. reported that the optimum ionomer content of catalyst-coated-membrane-type MEAs was 35 wt.% at low current density ( $0.4 \text{ A cm}^{-2}$ ) and low RH (40%), but decreased to 20 wt.% when the concentration overpotential became more dominant on increasing the current density to  $1.2 \text{ A cm}^{-2}$  (larger water production) or the RH to 87% (slower water removal) [22]. Similar trend was also reported for LT-PEMFCs prepared by the decal method [19].

The development and optimization of catalyst layers in IT-PEMFCs have received less attention; only a few studies have been reported, including studies on the effect of additives like ammonium carbonate [28] and PEG [29]. The effects of ionomer contents on the performance of IT-PEMFCs were investigated at

35% [30] and 100% RH [31]. In general, higher cell performances have been achieved at higher RH [31,32], but a large humidifier is required to maintain the RH. Since larger amount of water is required due to the higher saturated vapor pressure at  $120^\circ\text{C}$ , operation at lower RH would be desirable for IT-PEMFC; therefore, MEA optimization is required for such conditions. The high vapor pressure at  $120^\circ\text{C}$  also largely reduces the oxygen partial pressure; therefore, in IT-PEMFC, the cathode feeds are usually pressurized (0.5–2.0 bar) [4,29,31,33,34] to provide sufficient oxygen supply, while some studies employ atmospheric pressure [28,30,32]. Applying back pressure should also be beneficial in decreasing water removal in electrodes.

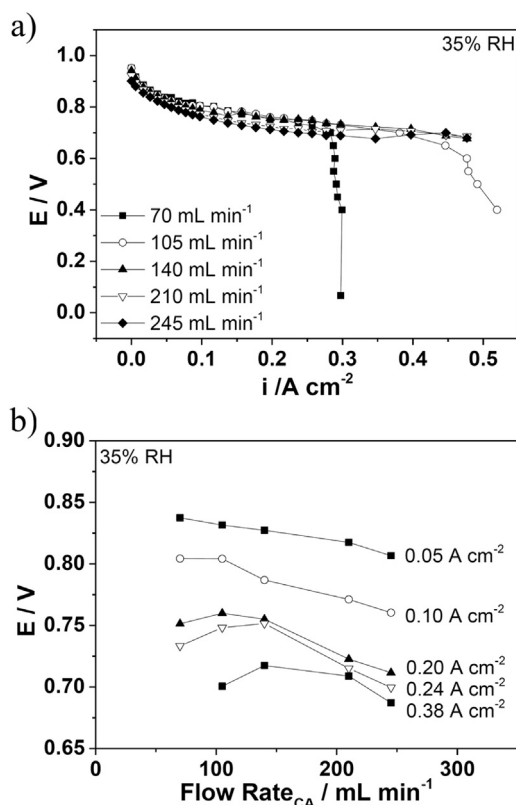
Since the ionic conductivity and mass transport of the catalyst layer are strongly dependent on the ionomer films layered on the catalyst particles, the efficient operation of IT-PEMFC under extremely low humidification conditions would be significantly affected by the properties of the ionomer in the catalyst layer. Briefly, the amount of ionomer present in the catalyst layer should produce sufficient ionic conductivity to minimize the ohmic drop under dried conditions, whereas mass transport can be hindered if excess ionomer and water are present in the electrodes. Especially under dehydrated operating conditions, the higher ionomer content can be advantageous because it can retain water molecules, resulting in better performance with increased ionic conductivity. Hydration of the MEA is further affected by other electrode properties, e.g. porosity, hydrophobicity, thickness, etc. and by operation conditions, e.g. RH, applied current, reactant flow rates, temperature, etc. [35,36]. Therefore, effective catalyst layers with optimum ionomer content should be designed to obtain highest IT-PEMFC performance under specific operating conditions. However, there has been no report on the effects of the Aquivion<sup>TM</sup> ionomer content in the catalyst layer on IT-PEMFC performance under low-humidity operating conditions (<35% RH).

Herein, we aim to construct effective catalyst layers of IT-PEMFC for the low-humidity operating conditions, and ionic conductivity and mass transport of the fuel cell electrodes were examined with various ionomer content in the catalyst layer. We investigated the dependence of single cell performance on Aquivion<sup>TM</sup> ionomer contents of the catalyst layer under reduced RH conditions. The effects of RHs (10–35%) and inlet gas flow rates ( $75\text{--}250 \text{ mL min}^{-1}$ ) on the IT-PEMFC performance were studied by conducting electrochemical analyses at  $120^\circ\text{C}$ , and the optimum ionomer content of IT-PEMFC was suggested in terms of the ohmic resistance and mass transport in the catalyst layers. The obtained results provide valuable insights on the operation of IT-PEMFCs under low-humidity conditions (<35% RH) and optimization of its catalyst layer to achieve high efficiency of the PEMFCs.

## 2. Experimental

### 2.1. Preparation of membrane electrode assembly

Commercial and homemade MEAs were used to study the effects of gas flow rate and cathode ionomer content, respectively. Catalyst inks were prepared by mixing a carbon-supported platinum catalyst (Pt 45.7 wt.%, Tanaka K. K.), isopropyl alcohol (A.C.S grade, Burdick & Jackson) and Aquivion<sup>TM</sup> ionomer solution (D83-06A, Solvay Solexis) to serve as electrodes of the homemade MEAs. The cathode ionomer contents were 20, 30, and 40 wt.% of the total solid weight, while the anode ionomer content was fixed at 30 wt.%. The total solid amount was 0.5 wt.% of the catalyst ink solutions for both electrodes. Then, the MEAs were fabricated by spraying the catalyst inks directly onto the 30  $\mu\text{m}$  thickness of Aquivion<sup>TM</sup> membranes (E87-03S, Solvay Solexis) using an automated system (Gunman, Jeewon Hi-tech). Pt loading was fixed at  $0.4 \text{ mg cm}^{-2}$  for both the electrode catalyst layers with an



**Fig. 1.** a) *i*-*V* curves with increasing cathode flow rate and b) performances at corresponding current densities under 35% RH.

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