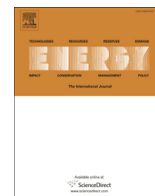




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Effect of LiCl content on pore structure of catalyst layer and cell performance in high temperature polymer electrolyte membrane fuel cell

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ABSTRACT

GDEs (gas diffusion electrodes) fabricated with catalyst layer, supporting layer and GDL (gas diffusion layer) are prepared for high temperature polymer electrolyte membrane fuel cell. A supporting layer consisting of different amount of PTFE (polytetrafluorethylene)-bonded carbon black from 0.0 g to 0.2 g is coated onto GDL to prevent Pt loss into GDL and to enhance the intimate contact between catalyst layer and GDL. Various catalyst layers are prepared by different LiCl content in PBI (polybenzimidazole) solution and coated directly on the top of optimized supporting layer. After preparing GDE, LiCl is leached out of the catalyst layer and the pore structure of GDE left behind the leaching is investigated with LiCl content. The N_2 adsorption isotherm indicates that leaching of LiCl from catalyst layer leaves the pores behind and the BET surface area is increased with LiCl content. It is seen that the LiCl content significantly affects the dispersion of Pt nanoparticles in PBI: that is, too low LiCl content leads to non-uniform dispersion of Pt nanoparticles, resulting in poor cell performance. An appropriate amount of LiCl would lead to better cell performance.

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1. Introduction

PEMFC (polymer electrolyte membrane fuel cell) has attracted an intensive attention from a variety of areas such as automotive, portable and stationary applications due to high power density, fast start-up, simplicity of operation and high energy conversion efficiency [1–4]. However, the most widely used Nafion membrane in low temperature PEMFC exhibits some inherent problems such as high cost, high fuel crossover, low conductivity at low humidity, difficult water and heat management, intolerance to impurities in feed stream such as CO and sluggish electrochemical kinetics at cathode. In order to overcome the current problems involved in low temperature PEMFC, a high temperature PEMFC (HT-PEMFC) to be operated above at 100 °C has been focused. As one of efforts to develop high temperature PEMFC, several cost effective and thermally stable membranes have been developed. Among several candidates for HT-PEMFCs [5,6], poly[2,2-(*m*-phenylene)-5,5-benzimidazole] named as PBI (polybenzimidazole) is considered

to be the best due to its excellent thermal, oxidative, chemical and hydrolytic stability under operation conditions, especially at high temperature [7]. PBI solution is typically prepared by dissolving in *N,N*-dimethylacetamide (DMAc) which has been known as the best solvent. Kojima et al. [8,9], however, reported the molecular aggregation of PBI in DMAc and formic acid. Lin et al. [10] also confirmed that the aggregation of PBI would be attributed to the interactions of –NH– (proton donor) and –N= (proton acceptor) groups within polymer. They suggest that as a stabilizer, LiCl is supposed to prevent the aggregation through the interaction of –NH– and –N=: that is, Li^+ may interact with –N= while Cl^- may react with –NH–, thus preventing aggregation of PBI molecules. In addition to good dispersion of PBI without aggregation, the pore structure and uniform dispersion of Pt nanoparticles in catalyst layer is also critical in the aspect of mass transport of fuel to catalyst active sites, proton transfer from catalytic active sites to membrane, removal of water vapor produced at cathode and cell performance. Fischer et al. [11] reported the addition of $LiCO_3$, ammonium carbonate and ammonium oxalate as a pore forming additive to the electrode in low temperature PEMFC where Nafion was used as a binder. They claimed that leachable filler, $LiCO_3$ created the overall porosity of 65%. Although several researches [12–17] reported that

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the addition of LiCl as a stabilizer was highly effective for homogeneous PBI dispersion in casting the membrane, no other study for LiCl as a pore forming additive in catalyst layer has been reported.

Another important factor is GDL (gas diffusion layer). The GDL requires fulfilling several aspects such as sufficient mass transport of reactant molecules (i.e. H_2 and O_2) to catalyst active sites and efficient removal of water vapor in case of HT-PEMFC produced at cathode as well as excellent electrical conductivity. It has been pointed out, however, that Pt loss into GDL and poor interfacial contact between catalyst layer and GDL lead to the deterioration of cell performance. Seland et al. [18] and Kongstein et al. [19] reported that the support layer deposited on GDL could enhance the cell performance. They claimed several advantages of carbon support layer coated on GDL: those are, i) prevention of catalyst loss due to its penetration into GDL, ii) better interfacial contact between catalyst layer and GDL and iii) provision of pathway for reactants and iv) provision of smooth surface for catalyst deposit. More recently, Pan et al. [20] also reported various porogens such as ammonium acetate and zinc oxide as well as ammonium carbonate and ammonium oxalate as pore forming additives in HT-PEMFC. They added porogens to GDL and supporting layer and found that the overall porosity of the electrode was ranged from 38% to 59%. It has been well known that like a catalyst layer, the backing layer, GDL should also provide a reasonable porosity to facilitate the mass transfer of reactants and removal of water vapor produced at cathode. As mentioned above, however, a direct deposit of catalyst ink on GDL results in Pt loss due to its penetration into GDL and poor interfacial contact due to irregular surface of GDL. In order to solve these problems, a supporting layer is desirable. However, the thickness of supporting layer becomes critical because the mass transfer resistance is significantly affected by its thickness. As of now, LiCl as pore forming additive as well as a stabilizer for catalyst layer in HT-PEMFC and the optimization of supporting layer have not been reported.

In this study, therefore, we optimize supporting layers and investigate the effect of LiCl addition to the catalyst layer on Pt dispersion and pore structure of catalyst layer. In addition, various MEAs are fabricated using the optimized supporting layer, and the cell performance is evaluated and discussed.

2. Experimental

2.1. Preparation of GDL

The carbon fiber paper, Toray TGP-H-060 (Toray Co.), was employed as the GDL. For wet-proofing, the carbon paper was immersed in 20 wt% PTFE (polytetrafluorethylene) dispersion (Sigma–Aldrich, USA, 60 wt% in H_2O) for about 10 min. After drying in air overnight at room temperature, the carbon paper was sintered in air at $360^\circ C$ for 15 min. The wet proofing by PTFE prevents the carbon paper from soaking and carbon particle loss into carbon paper by penetration.

2.2. Preparation of SL (supporting layer)

It is known that carbon supporting layer plays important roles such as prevention of Pt loss into GDL, intimate contact between catalyst layer and GDL for effective electrical conduction, good mass transfer of fuel and facilitation of water removal produced at cathode and smoothing the GDL surface for uniform deposit of Pt nanoparticles [17]. A supporting layer of PTFE-bonded carbon black was prepared as described elsewhere [20]. 18 wt% poly(vinyl butyral) (PVB, Sigma–Aldrich, USA) in xylene (Samchun, Korea) as a solvent was prepared, to which the PTFE dispersion with a plasticizer (Santicizer[®] 160) was then added under stirring. Carbon black (Vulcun XC) was finally added one and a half times in a weight as much as PTFE content and mixed thoroughly. The different amount of slurry for

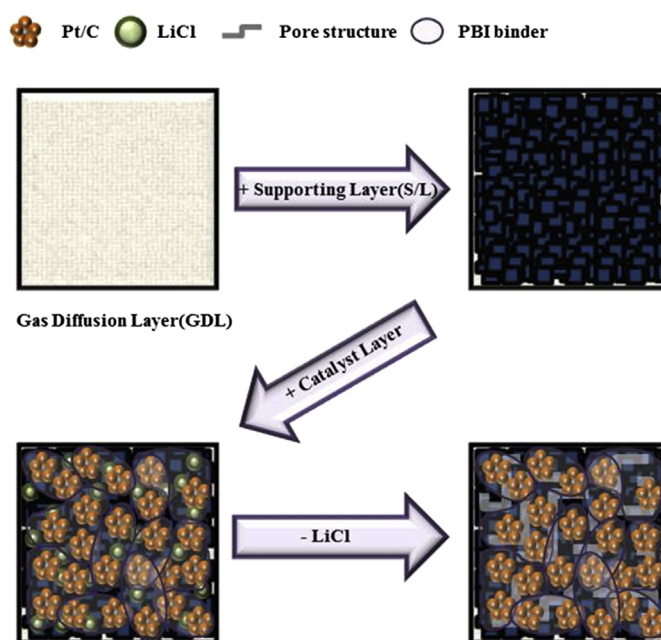


Fig. 1. Scheme of GDE fabrication.

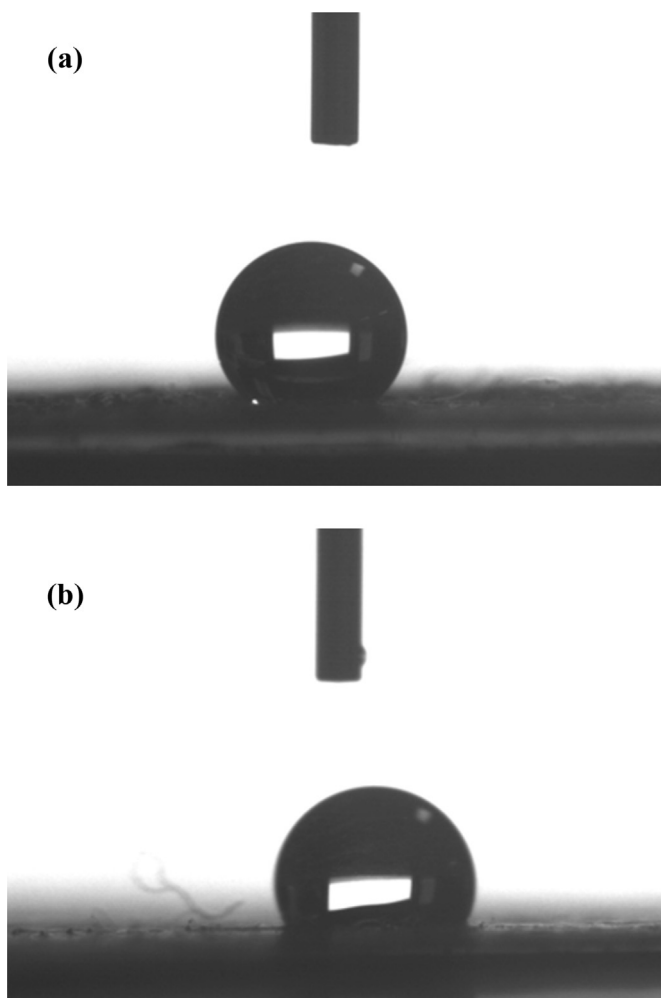


Fig. 2. Pictures of water droplets on (a) PTFE-treated GDL and (b) Untreated GDL for contact angle.

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