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Effects of Nafion impregnation using inkjet printing for membrane electrode assemblies in polymer electrolyte membrane fuel cells

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ABSTRACT

We present a method of using inkjet printing to deposit Nafion ionomer as the transport media onto catalyst layer made into membrane electrode assemblies (MEAs) for polymer electrolyte fuel cells (PEM-FCs). This method provides a more suitable mode of controlling the solution deposition than the existing deposition methods such as spray painting. The cyclic voltammetry results also show that the inkjet printing method has better performance than spray painting by improving catalyst efficiency. Using the appropriate Nafion loading of 0.64 mg cm⁻² by inkjet printing, we have demonstrated that this technique can be used to improve the performance of the MEA for PEMFCs.

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1. Introduction

Polymer electrolyte membrane fuel cells (PEMFCs) have received much attention because of their unique properties and their ability to create an efficient and clean source of energy [1–3]. The heart of a PEMFC is the membrane electrode assembly (MEA), which comprises of a proton exchange membrane, catalyst layers (CLs), and gas diffusion layers (GDLs). MEA requires catalyst as an active material of electrodes, for which it is important to increase the reaction sites in the catalytic layer to improve the electrode performance. Preparation of more efficient and cheaper electrodes has become an important research and development direction of PEMFCs in recent years [4,5].

It is known that catalyst particle as an effective reactive site connects with Nafion solution as a proton conductor to ensure high catalyst utilization and cell performance in Fig. 1. This fact suggests that proton conductivity through ionomer in catalyst layers is an important factor affecting MEA performance. The catalyst layer is impregnated with a dilute solution of electrolyte, thereby extending the three-phase boundary region. Aiming to improve three-phase boundary region of catalytic layer and catalyst utilization, several studies have specifically examined novel methodologies for MEA development [6,7]. An adequate dispersion

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http://dx.doi.org/10.1016/j.electacta.2014.02.133 0013-4686/© 2014 Elsevier Ltd. All rights reserved. of Nafion ionomer must be present within catalyst layer to ensure efficient proton conduction. Numerous studies have demonstrated that the ionomer network formed in catalyst layer can optimize the electrode structure and can improve catalyst utilization in PEMFCs.

Many deposition methods (such as spraying [8,9], screenprinting [10,11], electro-deposition [12,13], and sputtering [14,15]) have been developed recently to disperse the Pt content close to the electro-membrane interface, where electrochemical reactions take place. Inkjet printing techniques show great potential for increasing Pt efficiency by reducing amount of ionomer and by controlling Pt content. Inkjet printing cannot be regarded as a conventional process for MEA fabrication. Recently however, great interest has arisen with respect to inkjet printing technology for manufacturing catalyst layers onto the MEA [16-18]. Taylor et al. demonstrated a method of inkjet printing to deposit catalyst materials onto GDLs that are made into membrane electrode assemblies for PEMFCs [18]. He reported that inkjet printing had been demonstrated as a catalyst application method for PEMFCs and that high-precision of inkjet printing allows for controlled catalyst deposition, especially for ultra-low Pt loadings.

Therefore, inkjet printing technology was used here to prepare a solution that should be impregnated in catalytic layer to increase Pt utilization for proton exchange membrane fuel cells. However, Nafion solution as the transport media in the solutions and within catalyst layer generally exists as aggregated states, thereby leading to an insufficient Nafion utilization. To provide close contact between Nafion membrane and Pt catalyst, a Nafion solution is







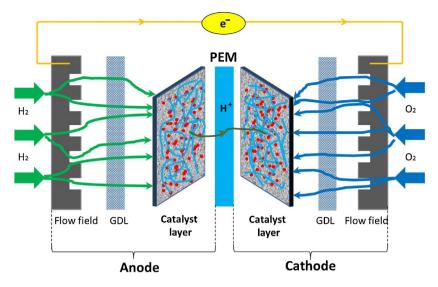


Fig. 1. Schematic diagram of microstructure in the catalyst layer for single typical proton exchange membrane fuel cell.

usually added to catalyst electrode. The appropriate loading of Nafion necessary for the best performing MEAs was investigated previously for electrodes prepared using ink processes [19–22]. A previous study by Kim et al. analyzed the effects of ionomer contents on the proton exchange membrane fuel cell performance of MEA fabricated using a catalyst-coated membrane spraying method in partially humidified atmospheric air and hydrogen [23]. That report described that the best MEA performance was obtained by an MEA containing 30 wt% ionomer in the cathode and 25 wt% ionomer in the anode.

Inkjet printing can be regarded as an efficient method for catalyst deposition due to its excellent control of Pt loading which results in better utilization of Pt than that by conventional catalyst deposition methods such as screen printing and spray painting. However, the effect of Nafion loading in catalyst layer by inkjet printing is not discussed.

This report describes demonstration of a new method for depositing Nafion ionomer as the transport media onto catalyst layer to increase the contact surface area using inkjet printing. The objective of this work is to study the effect of Nafion loading by assessing the performance of fuel cell using inkjet printing and spray painting method. The results showed that this method consumes a markedly lower amount of Nafion ionomer as the primary liquid media, which can be favorable for the fabrication of high-performance PEMFCs.

2. Experimental

2.1. Preparation of Nafion membranes

Nafion 117 membrane as the proton exchange membranes was first pre-treated step-by step with H_2O_2 and H_2SO_4 solutions to remove the remaining organic and inorganic contaminants [24,25]. The pre-treatment of the Nafion membrane was accomplished successively by treating the membrane in boiling 3% (v/v) H_2O_2 solution, distilled water at 100 °C, boiling 1 mol l^{-1} H_2SO_4 solution and then in distilled water at 100 °C again, for 30 min each step. The pretreated membranes were kept in water before the MEA fabrication.

2.2. Formation process of inkjet printing technology

Fig. 2 shows the schematic illustration of the formation and evolution process of Nafion droplets on the hydrophobic substrate

using inkjet printing. The typical inkjet printing process is based on a drop-on-demand printing method. It is necessary to understand the formation process of the printed ink droplet for effective design and fabrication of catalyst layer for PEMFCs. The printed dot is produced from the nozzles of the inkjet printer, which ejects ink onto the substrate. The nozzles are mounted about 1 mm over the substrate, and ink ejection velocities are in the range of 5 to 10 meters per second. Drops are formed by propagating a pressure pulse in a chamber behind the printing nozzle. The liquid drop hits a substrate, and this process is controlled by a number of physical processes and can be driven by inertial forces, capillary forces, and gravitational forces. Generally, the impact behavior of a drop can be divided into impact driven and capillary driven [26,27]. Therefore, the vertical speed and droplet gravity force play important roles in impregnating Nafion ink into the catalyst layer to improve the performance of the MEA. Furthermore, the droplets fabricated by inkjet printing have better orientation character than spray painting which enhances the impregnation of the Nafion ionomer in the catalyst layer. Subsequent phase change will transform the liquid into solid after solvent evaporation [27]. Fig. 2 also shows a clear

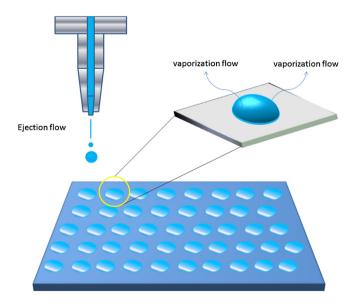


Fig. 2. Schematic illustration of the formation and evolution process using inkjet printing.

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