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Development of a novel decal transfer process for fabrication of high-performance and reliable membrane electrode assemblies for PEMFCs

Hong Je Cho, Hyunsook Jang, Seokhee Lim, EunAe Cho, Tae-Hoon Lim, In-Hwan Oh, Hyoung-Juhn Kim, Jong Hyun Jang*

Fuel Cell Research Center, Korea Institute of Science and Technology, 39-1 Hawolgok-dong, Seongbuk-gu, Seoul 136-791, Republic of Korea

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ABSTRACT

To improve the performance of a polymer electrolyte membrane fuel cell (PEMFC), various membrane electrode assemblies (MEAs) were fabricated by the decal process. When peeling the decal films away from a Nafion membrane, a novel liquid nitrogen (LN_2) freezing method was employed. The results of a Fourier Transform Infrared (FTIR) analysis of the Nafion membranes demonstrate that this proposed method has no impact on the molecular structure of the Nafion polymer. In addition, the method makes it possible to achieve complete decal transferring under a wide range of hot-pressing pressures and temperatures: 9.8–15.7 MPa and 100–140 °C, respectively. Another approach to optimize the decal technique is to dry catalyst layers under vacuum. Catalyst layers dried under vacuum show better cell performances than atmospherically dried ones. Vacuum drying significantly facilitates the formation of small pores within Pt/C agglomerates on catalyst layers. Third, the use of Additive-A as a commercial dispersant in the catalyst ink has been investigated. From rheological characterizations, including thixotropy and catalyst ink viscosity, it is obvious that the additive plays an important role in elevating the dispersion stability of the ink. In addition, surface images of the catalyst layers revealed that the dispersing agent reduces cracks or fractures within the layers. Although adding Additive-A did not have an effect on the single-cell performance, the MEAs with the dispersant are expected to have better results for a long-term performance test of a single cell.

1. Introduction

Polymer electrolyte membrane fuel cells (PEMFCs) are attractive because of their low pollution and high energy density at low temperatures (50–90 °C). However, to successfully commercialize a PEMFC, mass production of the membrane electrode assembly (MEA), which consists of a polymer electrolyte membrane with catalyst layers on both sides and gas diffusion layers (GDLs), is necessary. It is generally accepted that the MEA is the heart of PEMFCs and that it determines their performance. The MEA performance strongly depends on how the MEA is fabricated [1–4].

To date, several MEA fabrication techniques have been developed, such as decal, catalyst-coated membrane (CCM) and catalyst-coated gas diffusion layer (CCG) methods [5–11]. Among these MEA fabrication methods, the decal process is

* *Corresponding author*. Tel.: +82 2 958 5279; fax: +82 2 958 5199.
E-mail address: eacho@kist.re.kr (E.A. Cho).
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considered the most suitable method for mass producing MEAs. In this procedure, as shown in Fig. 1, the catalyst ink is first prepared with a Pt catalyst, an ionomer, and certain solvents. It is then coated over the decal substrate, such as Teflon film, by using a doctor blade in general. After spreading the catalyst ink on the substrate, the remaining solvent has to be evaporated to form an effective three-phase boundary where the catalyst, the reactants and the ionomer meet for electrochemical reactions to occur. In order to join the anode, the cathode and the membrane together, a hot-pressing process is subsequently carried out, and then the decal substrates are peeled away from both electrodes. In terms of the decal process, many researchers have been working to improve the performance of PEMFCs. However, several problems remain. First, the decal method causes uneven or incomplete transfer of the Pt catalyst from the decal substrate to the membrane [5]. In addition, we must find the optimum conditions for hot-pressing parameters such as the pressure and the temperature [1,5,11,15–19]. Second, in high-current regions, the microstructures of catalyst layers should be designed to eliminate the product water easily [10]. Third, the catalyst ink has to be prepared by dispersing appropriate amounts of catalyst and ionomer solution in a mixture of solvent, boosting Pt utilization and proton conduction [11].

In this work, we examined the three important factors mentioned above by optimizing a decal technique. To address the first problem, during the peeling-off process in Fig. 1, a liquid nitrogen (LN_2) treatment was employed to achieve complete decal transfer of the catalyst layers onto the membranes under a wide range of hot-pressing pressures and temperatures [12]. In addition, to address the second research topic associated with mass transfer problems at a high-current density, we applied a vacuum drying method when drying the catalyst layers on the decal films [13]. We compared the cell performances of MEAs fabricated by two different drying methods, atmospheric and vacuum drying. Even though extensive studies have investigated decal processes, the effects of atmospheric and vacuum drying conditions on PEMFC performance have not yet been elucidated. Third, we chose to create a well-dispersed catalyst ink by adding a commercial dispersing agent into the catalyst ink [14]. The effect of the dispersant on the ink was examined through various experiments. We present here the results of these three categories of experiments that were focused on optimizing the decal process.

2. Experimental

2.1. Materials

As an ionomer solution, 20 wt% Nafion solution (EW = 1100, DuPont Co.) was adopted. The catalyst inks were prepared by mixing a carbon-supported Pt catalyst (Tanaka K. K., Pt 45.6 wt %), isopropyl alcohol (J.T. Baker, HPLC grade), deionized water (resistivity of 18.2 MΩ cm, Millipore, USA) and the Nafion solution. Additive-A, a commercial dispersant from BYK Additives & Instruments, was used for assessing the effect of a dispersing agent on the ink. A small amount of the additive was mixed with the catalyst ink. In this study, the Nafion ionomer content was adjusted to 30 wt% of the total solid content. In addition, Nafion 112 (thickness ~50 μm, DuPont Co.) was used as the polymer electrolyte membrane. As a gas diffusion layer, Sigracet 10BC was purchased from SGL Carbon, Inc.

2.2. MEA preparation by decal process

The catalyst ink was stirred for 30 min with a magnetic bar. Next, a homogenizer (ULTRA-TURAX, Youngjin Corp.) was employed to ensure that all of the Pt particles came into contact with the other components uniformly. Table 1 shows

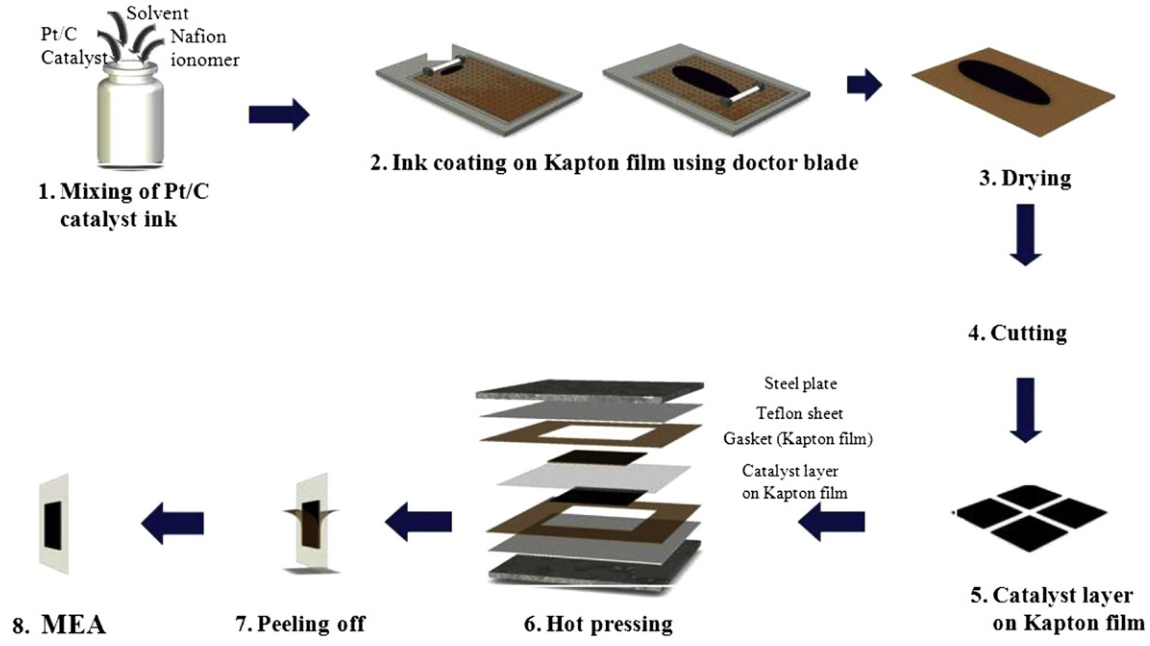

Fig. 1 – A schematic procedure for the MEA fabrication by the decal process.