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In-plane gas permeability and thought-plane resistivity of the gas diffusion layer influenced by homogenization technique and its effect on the proton exchange membrane fuel cell cathode performance

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ABSTRACT

In this study, in-plane gas permeability and through-plane resistivity of gas diffusion layer (GDL) made by different microporous layers (MPLs) were characterized by a new combined measurement setup under different compression ratios. Applied MPL ink-homogenizing techniques were pulse sonication, continuous sonication, bath ultrasonic and mechanical stirring. Results showed that 20% GDL compression produced optimized in-plane gas permeability and through plane resistivity, which is suitable for application in proton exchange membrane fuel cell (PEMFC). However, bath ultrasonic technique is more desirable technique for MPL ink homogenization in term of obtained gas permeability and total resistivity values; 0.45 ($\times 10^{-12}$ m^2) and 2.63 (mΩ cm^2) respectively.

Morphological investigation was also presented a uniformly distributed MPL microstructure without large cracks for the MPL which was prepared by bath ultrasonic technique (MPL-C).

The Obtained results in H_2/air PEMFC under various cathode relative humidity (RH_C), 33–100%, revealed that the performance of the MEA made by MPL-C is 20% higher than others; 504 mWcm^{-2} versus 385 mWcm^{-2}at 33% RH_C. More analysis by electrochemical impedance spectroscopy demonstrated that MPL ink preparation by bath ultrasonic techniques resulted a lower ohmic and charge transfer resistance in PEMFC at least in small scale MEA.

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Introduction

Among various kinds of fuel cells, proton exchange membrane fuel cells (PEMFCs) are more considered because operating at low temperature, high energy efficiency and being environmentally friendly [1,2]. In order to commercialize PEMFCs widespread and successful, improvement of the PEMFC power density and durability is needed. Up to now, many strategies have been applied to achieve this purpose in different parts of the PEMFC including membrane electrode assembly, gas diffusion electrodes and bipolar plates. Gas diffusion layer (GDL) has important functions in the PEMFC such as; diffusion of reactants and products, electron and heat conductivity, and mechanical support for the membrane electrode assembly (MEA) [3,4].

A conventional dual layer GDL consists of two layers; macro-porous substrate (MPS) and micro-porous layer (MPL). A MPL is composed of carbon black powder and a hydrophobic agent such as PTFE, which applied on the MPS (usually a slice of carbon cloth or carbon paper). MPL provides a support for the catalyst particles and prevents them to penetrate deeply into the MPS. It also, reduces the contact resistance between the MPS and catalyst layer and helps reactants to distribute toward the catalyst sites homogeneously. In addition, it improves water management in the MEA [5–8].

Up to now, numerous studies have been done in order to improve MPL characteristics such as MPL composition [5,9–14] porosity [15], permeability [16], conductivity [17], hydrophobicity [18], compressibility [19,20], and fabrication process [21,22].

However, most of the investigations have been focused on the MPL composition and its characteristics. One of the major steps of MPL fabrication is its components homogenization. The MPL ink has been usually homogenized by various techniques; probe sonication (pulse and continues) as a common technique [9,10,12,23], ultrasonic bath [5,11,24] and mechanical stirring [13,25,26]. The homogenization technique can change physical properties such as conductivity, permeability, porosity and other features of the MPL.

In summary, sonication is capable to be done in directly or indirectly manners. In direct sonication, which is the most popular way of sonication, a probe is inserted directly into the sample. This method offers high intensity and effective processes for most samples. In the probe sonication technique, the probe, which could be assumed as the wave production source enters into the sample resulting in a high force induction to sample ingredients. Continuous and pulsed operations are two distinctive methods of the probe sonication technique. Needing to a probe is eliminated in indirect sonication. This technique is often described as a bath ultrasonic. In the bath ultrasonic technique, the source of wave production is not in touch to the sample or even its container. Usually, wave conduction to the sample happens through the water in the bath.

Mechanical stirring technique has usually low power output for ink homogenization. Also, this technique needs to a long time period for homogenization.

Regarding to the wide yielded achievements in the MPL composition and manufacturing processes, the effect of the MPL ink homogenization technique on the physical characteristics and performance of the MPL is not systemically studied yet. This is a lacuna aspect in MPL preparation. The objective of the present work is to study the effect of the MPL ink homogenization technique on the intrinsic characteristics of the GDL and H_2/air PEMC performance. In order to do that, several identical inks of the MPL were formed and homogenized by the following techniques; i) Pulse probe sonication, ii) continuous probe sonication, iii) bath sonication and iv) magnetic stirring homogenization. After MPLs ink coating on the MPS, the produced GDLs were characterized in term of gas permeability, electron conductivity, surface morphology and, finally examined in the H_2/air PEMFC by i–v curves and electrochemical impedance spectroscopy (EIS) under different operation conditions.

Experimental

Homogenization of the MPL inks by different techniques

Four identical batches consist of 50 mg of carbon powder (Vulcan XC-72R, Cabot Co.), 20 wt % of PTFE (DuPont) in Isopropyl alcohol (Merck) and DI water in the ratio of 90:10 wt.% were prepared. The samples were dispersed by different homogenization techniques; i) Pulse probe sonication for 10 min, assigned as MPL-A, ii) continuous probe sonication for 10 min, assigned as MPL-B, iii) bath sonication for 2 h, assigned as MPL-C and iv) magnetic stirring homogenization for 2 h, assigned as MPL-D. Homogenizer 2200 HD (Bandelin Co.), ultrasonic bath (Struers Metason, 120T) with applying 50–60 Hz frequency and stirrer (Alfa HS-840) were applied to homogenize samples, respectively.

Four slices of carbon paper (Toray, TGP-H-060ST) were used as the electrode backing substrates. Homogenized MPL inks were coated onto the carbon paper and sintered in two stages; i) 280 °C for 30 min and, ii) 350 °C for 30 min. MPLs loading was controlled at 1.4 mg cm^{-2}.

The morphology of MPLs was studied by scanning electron microscopy (SEM, Ziess model VP).

In-plane gas permeability and total resistivity measurement

The in-plane gas permeability of the GDLs containing MPLs A–D was measured by an apparatus based on the Nitta et al. experimental setup [27]. The measurements were carried out in different compressibility of GDLs. The compression ratios of the GDLs were controlled by the gasket thickness in the experimental set up.

By means of the mentioned apparatus, in-plane gas permeability and through-plane conductivity evaluated simultaneously for all the manufactured GDLs.

Gas flow resistance is considered as the gas permeability which is calculated by Darcy's law [27]. By applying conservation of mass and ideal gas law, the gas permeability can be obtained by the Eq. (1):

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