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Property Changes of Gas Diffusion Layer in a PEFC by Compression

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체결압이 고분자연료전지 가스확산층에 미치는 영향

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ABSTRACT

분자전해질연료전지 내의 다공성 기체확산층은 반응가스의 확산과 전자이동통로의 역할을 수행할 뿐만 아니라 전기화학반응에 의해 공기극에서 생성된 수분(기상 혹은 액상)을 반응면으로부터 분리판 채널 방향으로 이동시켜 배출시키는 중요한 역할을 한다. 따라서 물관리를 통한 성능향상을 위해서는 기체확산층의 구조 및 재료특성에 대한 심도 깊은 연구가 필요하다. 실제 단위전지 체결시 기체확산층은 분리판의 리브(rib)에 의해 눌리게 되며, 그 부분의 기공 크기 분포의 변화를 야기한다. 또한 리브 전단부분에서 탄소 섬유가 손상을 입으며, 탄소 섬유를 감싸고 있는 PTFE coating이 벗겨지게 되어 표면화학적 특성이 달라진다. 본 연구에서는 단위전지 체결 시 분리판에 의해 눌리는 기체확산층의 기공 크기 분포 변화를 측정하였으며, 기공의 소수성에서 친수성으로의 변화를 알아보았다. Mercury 기공 측정기와 PMI 기공 측정기는 큰 기공 분포의 변화에, 질소의 흡/탈착을 이용한 BET 방식은 작은 크기의 기공 분포 변화 관찰에 사용되었다. 체결압에 의한 탄소섬유의 구조적 변화와 아울러 표면의 습윤 정도의 변화를 XPS와 물/알콜 uptake를 이용해 알아보았다. 이 연구를 바탕으로 물관리를 통한 연료전지 성능 향상을 위한 최적 GDL 선정에 기반이 되는 자료를 도출하였다.

KEY WORDS : gas diffusion layer(가스확산층), pore size distribution(기공크기분포), compression (체결압), wettability(습윤성), PEFC(고분자전해질연료전지)

1. Introduction

In recent years, much interest has been put on the gas diffusion layer (GDL) whereas it is essential to have an efficient link between the bipolar plates and active catalyst layers. The porous gas diffusion layer provides humidified gas

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access from flow field channel to catalyst layer including in-plane permeability to regions adjacent to ribs. Also it provides passage for removal of product water to opposite direction of reactant gas[1]. In the polymer electrolyte fuel cell (PEFC) system, water plays an important role in proton conduction. Although high water content in the membrane is desirable for high proton conduction, excess water content is detrimental to the cell performance because it can be condensed to liquid phase water in the electrode and gas diffusion layer. The liquid phase water can drastically diminish the cell performance by hindering gas diffusion as well as by covering the active sites of the electro-catalysts. Consequently, proper water management is vital to ensure sustainable performance of PEFC and researches on two phase transport in gas diffusion layer are required urgently.

For enhancing the knowledge on the material properties of GDL, physical and electrochemical characterization[2] and mechanical behavior of gas diffusion layers properties were studied[3]. In addition, Lin and Nguyen showed the effect of thickness and hydrophobic polymer content of the gas diffusion layer on electrode flooding level[4]. He has provided various approaches to solve the two phase transport phenomena in porous gas diffusion media[5-7].

In the actual clamped condition of cell on operation, the porous media readily deform with the application of external forces. Current understanding of the phenomena involved is limited by the inaccessibility of gas diffusion layer to in-site experimental measurements. Compression effects are commonly disregarded because of their complexity. However to optimize the water management, the in-depth understanding of compression effect on GDL is required in advance. The GDL beneath the rib is more compressed than that beneath the channel, resulting in different pore size distribution depending upon the relative position to the rib and channel. In addition, hydrophobic characteristics of GDL surface beneath the rib is changed due to the broken bonds of PTFE coated carbon fibers. PTFE is currently used as a hydrophobic agent.

In this paper, changes in pore distributions and surface properties of GDL by compression were studied. Since the hydrophobicity of GDL allows water management in a fuel cell, changes in hydrophobic characteristics of GDL beneath the rib of the flow field plate due to the broken bonds of carbon fibers and PTFE coating on the fibers was presented.

2. Experimental

2.1 Description of the sample

Carbon felt type GDL (SGL Inc.) of 370 μm thick was used for the experiments. This commercial GDL consists of gas diffusion medium (GDM) and microporous layer (ML). The GDM and ML were treated with 5 wt% and 23 wt% of PTFE, respectively. Mechanical and electrical measurements such as thickness, contact resistance versus compression were done simultaneously by lab-made device. Three times of successive compression have been applied on the sample. In-plane permeability of GDL is recorded during the compression test. Porosity is estimated using weight per unit area and uncompressed thickness.

The compressed porosity is calculated by correcting for the loss in the thickness under load. Although the thickness change is small during the compression test, both the contact resistance and permeability are drastically decreased. It

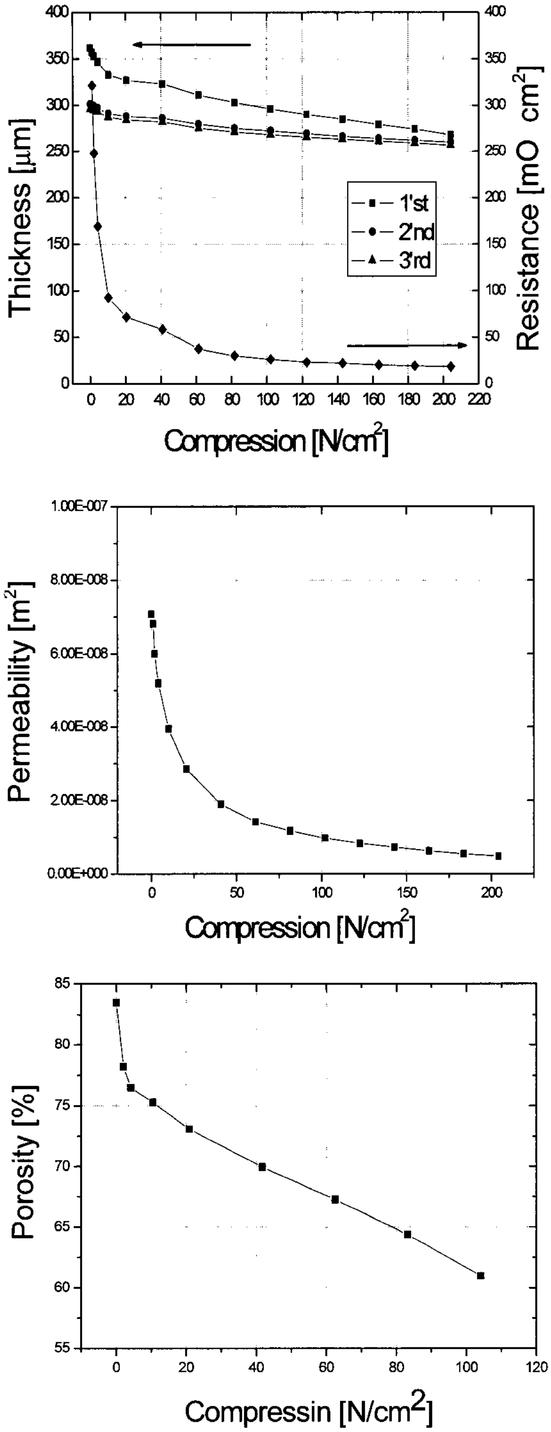

Fig. 1 Generic properties of (a) thickness and contact resistance versus compression (b) permeability versus compression (c) porosity versus compression

necessitates studying the compression effect on physical properties of GDL in detail. In all experiment, GDL was compressed to the 70% of initial thickness, which is generally used value on actual cell operation.

2.2 Pore size measurements

Pore size distributions of fresh and compressed GDLs were measured by mercury porosimeter (Autopore IV9500, Micromeritics, Inc). GDL comprised the two layers of macro porous GDM and micro porous layer (ML). Therefore, GDL generally shows bimodal pore size distribution, for the micro pore region, N_2 adsorption/desorption (ASAP 2400, Micromeritics is utilized and for the macro pore region, Automated Perm Porometer 6.0 (ASTM D 737-96, D737-96, F316-86, F778 and B.S.3321, 6410; P(ASTM D737-75, D737-96, F316-86 F778 and B.S 3321, 6410; Porous Materials, Ins) is applied.

3. Results and Discussion

3.1 Changes in pore size distribution by compression

Pore size distribution of GDL is changed by the compression and results are plotted in Fig. 2. The total pore volume is decreased by 4% after compression. Most of pore volume change occurs in the range larger than 10,000 nm. Considering that the PMI method uses pore wicking liquid and

Table 1 Pore volume of fresh and compressed GDLs, obtained from Mercury Porosimeter

Pore volume Specimen	< 100 nm	> 10,000 nm
Fresh	0.15%	87.65%
Compressed	0.221%	83.49%

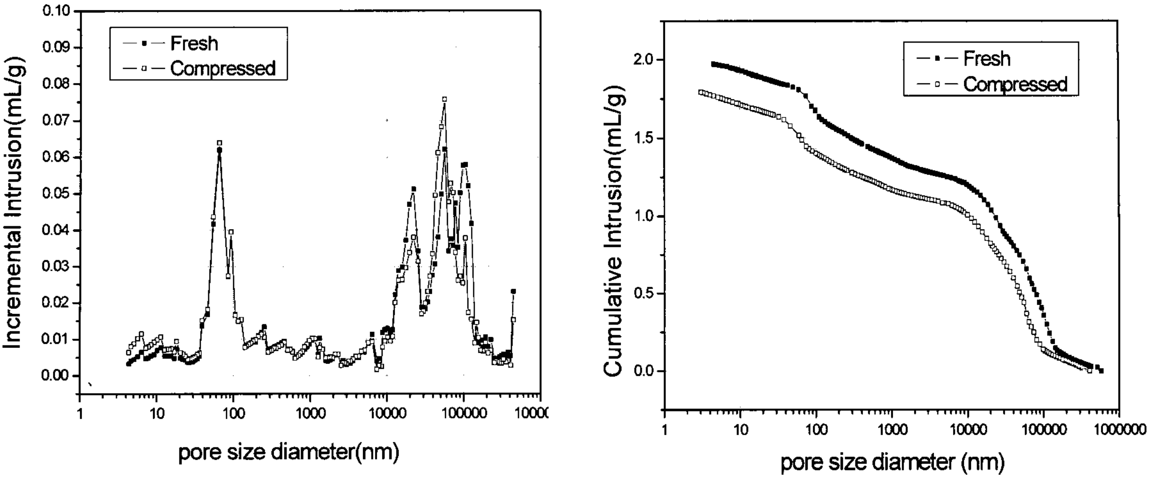

Fig. 2 Pore size distribution of fresh and compressed GDLs, measured by mercury porosimeter (a) Cumulative volume and (b) incremental volume

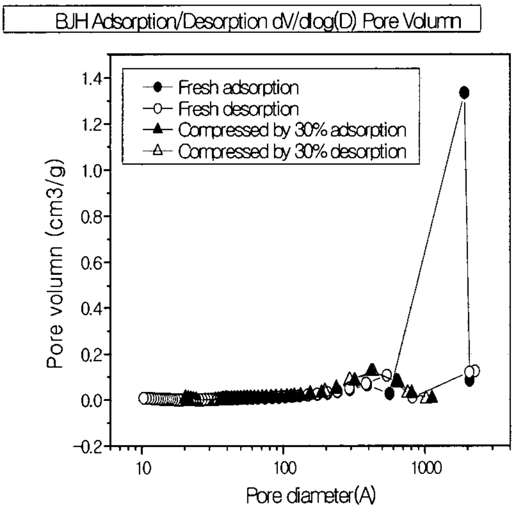

Fig. 3 Pore size distribution of fresh and compressed GDLs, measured by N_2 adsorption/desorption isotherm

it determines the flow mean diameters of pores. The determined bubble breaking pressure and pore diameter depend on not only pore size but also wettability of pore inner surface. The size of flow mean pore (bubble breaking point pore diameter) was measured to be decreased slightly from 15.7 to 14.0 microns by the compression.

The pore volume changes were less than 10% as determined by three independent methods. However, the permeability is drastically decreased by 1 order in magnitude by the compression. It seems to mean that the tortuosity of pores in GDL is increased, thereby blocking the passage of gases. In addition, the compression may cause many broken bonds and failure of water proof property in the GDL.

Table 2 Bubble breaking point pressure and pore diameter of fresh and compressed GDLs determined by PMI method

	bubble breaking pressure	bubble breaking pore diameter
Fresh	0.38 psi	15.7 micron
Compressed	0.48 psi	14.0 micron

Table 3 Changes of surface concentration of fluorine after compression of GDLs measured by XPS

	Fresh		Compressed	
	GDM	ML	GDM	ML
Surface fluorine contents (%)	53.7	46.8	49.5	46.1

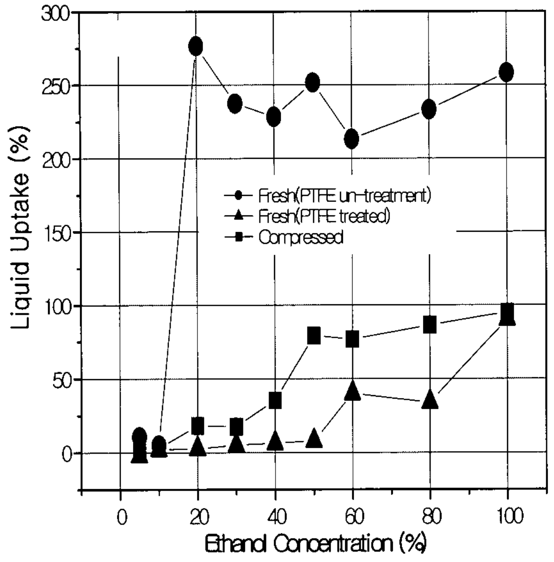

Fig. 4 Liquid uptake of fresh and compressed GDLs

Table 3 presents the loss of fluorine in the GDL by the simple compression process, determined by XPS technique. Most of fluorine loss was recorded only in the GDM and its amount is about 4 wt%. The fluorine was incorporated during the water proof treatment and its loss causes lack of water proof property. Although the loss of fluorine is as small as 4 wt%, the wettability change of GDL was determined to be significantly large as shown in Fig. 4.

3.2 Wettability change of surface by compression

In order to investigate the wettability changes of GDL, liquid uptake for the fresh and compressed GDLs were measured. For the soaking liquid, concentration controlled aqueous ethanol solution were used. For the fresh GDL, most of the liquid uptake occurs only above the ethanol content of 60%. In contrast, the compressed GDL soaks up several time more liquid even in the low ethanol content ranges.

4. Conclusion

The pore volume changes of GDL by compression were less than 10% as determined by three independent methods. In addition, the loss of fluorine is as small as 4 wt%. However, the permeability is drastically decreased by 1 order in magnitude by the compression. The compressed GDL soaks up several times more liquid in the low ethanol contents ranges.

The changes of pore size distributions determined from three independent methods and permeability and liquid uptake results suggest that the compression causes large changes in surface properties probably due to many broken bonds of PTFE and failure of water proof property in the GDL.

Pore size distribution and porosity under compression are two of the important parameters on mass transport in the GDL. These will be used for better understanding of two phase transport phenomena regarding compression effect.

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