Effect of ionomer on mass transfer of PEFC catalyst layer and design of optimal structure

Gen Inoue[1], Takahiro Matsuoka[2],
Yosuke Matsukuma[2] and Masaki Minemoto[2]
[1] Dept. of Chemical Engineering, Kyoto University, Kyotodaigaku Katsura, Kyoto, 615-8510, Japan
[2] Dept. of Chemical Engineering, Kyushu University, 744 Motooka, Nishi-ku, Fukuoka, 819-0395, Japan

INTRODUCTION

It is needed to reduce the cost of polymer electrolyte fuel cell (PEFC) by increasing Pt utilization. Especially, optimization of catalyst layer (CL), which consists of void space, carbon support and ionomer, is effective to increase the performance of oxygen reduction reaction (ORR) from the view point of mass transfer. And its structure can be controlled by fabrication process. In this study, three-dimensional catalyst layer was simulated by numerical analysis, and the relationship between electrode structure and Pt utilization. Moreover, from the results, the effect of CL fabrication process on cell performance was examined experimentally.

SIMULATION MODEL AND RESULTS

The primary structure of carbon black (CB) is aggregate structure. By using our previous method [1-4], heterogeneous CL structure shown in Fig.1 was simulated. Next, ORR and oxygen, proton and electron transport were simulated with these effective diffusion coefficient and conductivities. Our previous model [4] was used for the mass transport simulation in heterogeneous porous media. It is assumed that local oxygen dissolves and diffuses to Pt through ionomer. Fig.2 shows in-plane CL structure and oxygen concentration distribution in the case of ionomer content 20 vol. %. Local oxygen concentration was decreased at small pore space. Fig.3 shows the i-V curve and reaction rate distribution of each ionomer content. In the case of 30 vol. %, many pore were blocked, and reaction rate distribution became ununiform because oxygen concentration was not uniform.

EXPERIMENTAL EVALUATION

In order to examine the effect of ionomer on CL structure and CB agglomeration from fabrication process, the particle size of each ionomer and carbon dispersion with different ionomer carbon weight ratio (I/C) and carbon concentration was measured. Fig.4 shows the CB agglomerate and CL structure. CB agglomerate size in I/C=1.0 dispersion was the smallest. It is inferred that ionomer can not only control CB-CB affinity but also be binder. And from the SEM image, it was found that large CB agglomerate made large pore in CL. These tendencies became stronger by increasing concentration.

Based on the above results, in order to decrease oxygen diffusion resistance and to increase Pt particle exist near large pore, new CL fabrication process with was developed by two step mixing. Firstly the dispersion of I/C=1.5 without Pt was prepared. Next, it was mixed with I/C =0.5 with 50 wt. % Pt dispersion. By this method, many Pt particles were supported around the large CB agglomerate as the core structure Fig.5 shows CB agglomerate core structure obtained by SEM. With this structure, the cell performance was measured with pure H2 and O2 under 80 ^{o}C cell temperature and humidifier. Fig.6 shows i-V curve, at the high voltage condition, compared with usual structure, the effectiveness of this new structure was confirmed.

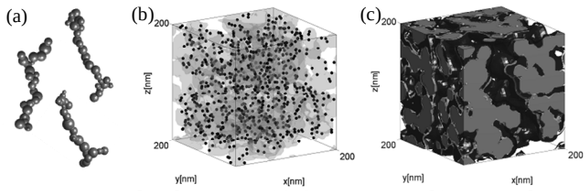

Fig.1 Simulated structure (a) CB aggregate (b) Pt particle (c) ionomer coating on CB surface

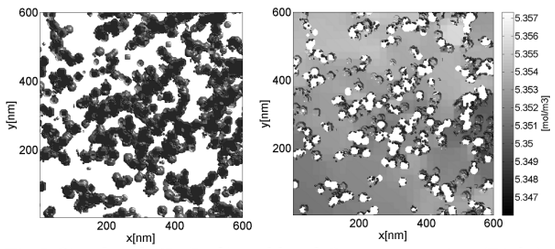

Fig.2 Simulated CL (Left) and local O_2 concentration (Right)

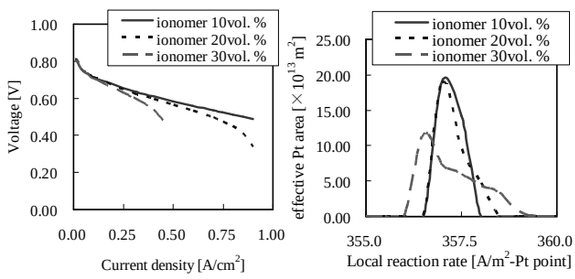

Fig.3 Cell performance (Left) and local reaction rate (Right)

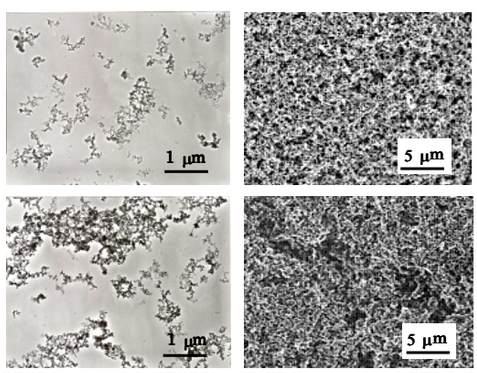

Fig.4 CB aggregate by TEM (Left), CL structure by SEM (Right) (Upper: I/C=0.5, Lower: I/C=1.5)

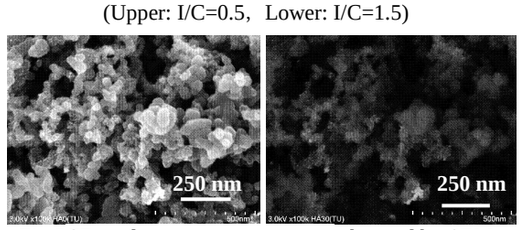

Fig.5CB agglomerate core structure obtained by SEM (Left: Secondary electron image, Right: Reflected electron image)

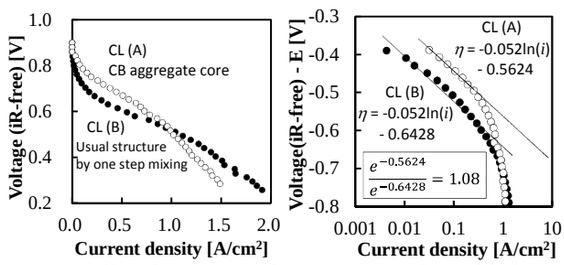

Fig. 6 Cell performance (Left: I-V curve, Right: Tafel plot)

REFERENCE

[1] G.Inoue et al., J. Power Source 175, 145-158 (2008)
[2] G.Inoue et al., ECS Trans., 16 (2) 1661-1671 (2008)
[3] G.Inoue et al., ECS Trans., 16 (2) 769-777 (2008)
[4] G.Inoue et al., ECS Trans., 25 (1) 1519-1527 (2009)