

Development of Mesoporous Carbon Fibers as Novel Catalyst Supports for Polymer Electrolyte Fuel Cell

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論 文 内 容 の 要 旨

In this work, hydrogen energy was focused on as one of the solutions for global environmental problems through the combination with renewable energy. Polymer electrolyte fuel cell (PEFC) generating electricity with a hydrogen fuel was particularly focused on among technologies related to hydrogen energy. Even though PEFC is a commercialized technology, cost and durability are still barriers on the way of further spreading PEFC. Particularly, degradation of catalyst layers is a serious problem since they mainly determine PEFC performance. Catalyst layers are commonly composed of electrocatalysts, Pt particles dispersed on carbon, and a proton conduction ionomer. One of degradation mechanisms of cathode electrocatalysts is known to agglomeration of Pt particles on the carbon support. In the previous work, Pt particles were encapsulated within pores of mesoporous carbon (MC) supports, resulting in increase in durability of electrocatalysts. However, a critical issue that the μm -scale structure was not well controlled although the nm-scale structure was well controlled was found. Since the μm -scale structure of catalyst layers was very important to get high PEFC performance, only slightly low PEFC performance was achieved.

In this study, in order to solve the problem on the μm -scale structure, controlling of the three-dimensional μm -scale structure was proposed by further fiberization of MC using an electrospinning deposition (ESD) method. The objectives of this research were to develop MC fibers (MCFs), to apply to catalyst layers of PEFC, and then to improve PEFC performance.

In chapter 1, a research background, advantages, and critical issues of PEFC technology were first introduced. Then, the previous research approach toward the durability issue of electrocatalysts was explained. Finally, objectives and research approaches of this study were introduced based on the critical issue found in the previous study.

In chapter 2, synthetic and material characterization methods for MCFs were introduced. Then, the methods of material and electrochemical characterization as electrocatalysts were explained after Pt deposition on MCFs. Lastly, for the evaluation as PEFC, preparation and evaluation methods of membrane electrode assemblies (MEAs) were discussed.

In chapter 3, different MCFs were prepared by varying conditions, such as pH, concentration, heat treatment, and so on, and characterized to find an optimum condition in order to get MCFs with the large pore diameter and pore volume. After the optimization of synthetic conditions, MCFs revealed the fiber diameter of 1-2 μm and the mesopore diameter of 4 nm or larger. Because of their fiber structure, the higher elasticity and electronic conductivity was achieved in comparison to MC bulk.

In chapter 4, as electrocatalysts, various MCFs were electrochemically evaluated after Pt deposition. Electrochemical active surface area (ECSA) and oxygen reduction reaction (ORR) activity of MCFs based electrocatalysts were characterized. MCFs with 2.5-3 nm Pt nanoparticles encapsulated into 4-5 nm mesopores showed highest ORR activity among MCF based electrocatalysts, which was comparative to a conventional electrocatalyst. Also, a correlation between ORR activity and mesoporosity was studied. Mesopores with the larger diameter and larger pore volume were necessary for large ECSA and high ORR activity, derived by good dispersion of Pt particles.

In chapter 5, for the evaluation as PEFC, MEAs were prepared with MCFs based cathodes. With MCF with 4-5 nm mesopores, current-voltage (I-V) performance resulted in slightly low in comparison to a conventional electrocatalyst. However, after the further enlargement in mesopore diameter to 6-7 nm, the improvement in I-V performance was found, the current density reached to 615 mA/cm^2 at 0.6 V, which was almost the same current density as the conventional MEA. However, in order to further improve I-V performance, especially to reduce concentration overvoltage, it was found that MCF-based powder was not appropriate to fully control the structure of catalyst layers.

Toward a problem in MCFs powder, MCF sheet was rather applied to the cathode. Pt deposited MCF sheet was first directly used as a cathode, but the I-V performance resulted in very low since the contact between the electrolyte membrane and MCF sheet was not good because of large inter-fiber pores of several micrometers. Then, conventional electrocatalyst powder was added to MCF sheet to partially fill up those inter-fiber pores. With this composite cathode, I-V performance was a lot improved, and slightly less than conventional I-V performance. On the other hand, ohmic resistance was improved than that of conventional MEA because of the sheet structure of MCF sheet. Furthermore, addition of conventional electrocatalyst powder to Pt deposited MCF sheet was tried. However, no improvement in I-V performance was obtained. The reason was probably because introduction of a proton conducting ionomer into MCF sheet was not successful, and further improvement on introduction methods was found to an issue to overcome for the further improvement.

In chapter 6, as conclusions, new findings though this study were summarized, and future outlook for further improving PEFC performance was discussed.