

Development of Novel Nano Carbon-based Fuel Cell Electrocatalysts with High Performance

金, 菜?

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氏 名 : 金 菜 リン

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論 文 内 容 の 要 旨

In Chapter 1, a short introduction on fuel cells electrocatalysts will be given, starting with their properties, continuing with their applications and finishing with the challenges.

In Chapter 2 and 3, a facile one-pot method is developed to load the Pd-Pt core-shell on MWNT and Graphene by the successive additions of Pd salt and Pt salt in the dispersion of catalyst supporting at room temperature. This method requires no reducing additives probably because the MWNT and Graphene serve as the reducing agent for Pd-Pt. Such a simple approach enables the large scale synthesis of Pd-Pt on MWNTs having remarkably high electron conductivity and electrochemical durability.

In Chapter 4, Au-NPs with Pt-shell layer (Au-Pt) on conductive support are promising electrocatalyst for fuel cells due to the minimum use of Pt by replacing the core to Au having lower cost and higher acid tolerance. Here, an easy and novel strategy is proposed to grow ~5 nm Au-Pt core-shell NP on the MWNT sheets without any additional surfactant or reducing agent, in which PBI was used as the binding sites to load the Au-Pt on the surfaces of the MWNTs.

In Chapter 5, a Pd-based electrocatalyst is developed by loading Pd-NPs onto the surfaces of CNTs. The PBI on the CNTs plays a significant role in the effective and homogeneous Pd attachment. After the KOH doping, the PBI layer on the catalyst became anion conductive, and, as a consequence, a triple phase boundary (TPB) structure was formed around Pd. The Pd-based electrocatalyst was employed in the cathode of the anion-exchange membrane fuel cell (AEMFC) and was found to show a remarkably high power density (241 mWcm^{-2}). It is assumed that the formation of the TPB structure may lead to such a high performance.

In Chapter 6, three different electrocatalysts are prepared, including s-Graphene/PyPBI/Au_{6.6}, s-Graphene/PyPBI/Au_{3.3} and s-Graphene/PyPBI/Au_{1.6} with the Au-NP diameters of 6.6 nm, 3.3 nm and 1.6 nm, respectively, in which the Au-NPs were homogeneously loaded onto the surfaces of the s-Graphene having no capping molecules or oxidized functional groups. In the present approach, the PyPBI coated on the surfaces of the s-Graphene acted as an effective anchor that stabilized the very small Au-NPs, and the size of the Au-NPs were controlled by changing the feeding amount of the Au salt ions. By this method, an ultrasmall Au-NPs having a 1.6 nm average diameter was readily prepared, and these

Au-NPs possessed the 4-electron transfer ORR reactivity. Electron donation from the PyPBI to Au-NPs is suggested to be important for such an ideal oxygen reduction to H₂O. Most importantly, the catalyst showed a very low overpotential; indeed, the value was lower than that of the conventional Pt-NPs, indicating that our catalyst has a higher catalytic activity than that of the Pt-NPs. This is the first report describing the preparation of an Au-NP-catalyst having higher ORR activity than that of the conventional Pt-NPs.

In Chapter 7, Sulfonated polymers, such as sulfonated polysulfone and sulfonated polyimide, are promising candidates as a future polyelectrolyte for PEFCs. In PEFC cell systems, the same polyelectrolyte material is often added to the electrocatalyst layer as the ionic path. Partially sulfonated poly(arylene thioethersulfone) (SPTES) and poly[(2,2'-benzidinedisulfonicacid)-*alt*-(1,4,5,8-naphthalenetetracarboxylic dianhydride)] (BDSA-NTDA) were selected as the electrolyte and wrapped MWNTs using the materials, to which Pt were attached onto the polyelectrolyte layer to fabricate new electrocatalysts, where the MWNT and Pt serve as the electron path and reaction site, respectively. For both polyelectrolytes, Pt-NPs with a narrow distribution were found to be homogeneously dispersed on the polymer-wrapped MWNTs. The electrochemical measurements revealed that MWNT/SPTES/Pt and MWNT/BDSA-NTDA/Pt possessed electrochemically active surface areas (ECSA) of 42.3 and 35.6 m²/g of Pt, respectively. Such a high ECSA efficiency ensures the use of these composites for the preparation of a future electrocatalyst.

In Chapter 8, provides the conclusions drawn from experimentation as well as future recommendations.