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主 論 文 の 要 旨

論文題目　**Synthesis of Nitrogen-Doped Graphene-Coated Metal Nanoparticles and Their Catalysis for the Oxygen Reduction Reaction**
(窒素含有グラフェン被覆金属ナノ粒子の合成と酸素還元触媒)

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論 文 内 容 の 要 旨

The proton exchange membrane fuel cell (PEMFC) has the potential to become a future green energy source as an alternative to fossil fuels. The platinum (Pt) catalyst is a practical catalyst for PEMFC owing to its low overpotential for oxygen reduction reaction (ORR) and fast reaction rate. However, the development of alternative materials is still essential due to the remaining issues of the Pt catalyst, including the high cost, low durability and poor tolerance to poisoning. To overcome these drawbacks, I have proposed the possibility of applying nitrogen-doped graphene-coated metal nanoparticles (hereinafter referred to as core-shell nanoparticles) as a solution, in which nitrogen-doped few-layer graphene (NFG) is coated on Pt-based bimetallic nanoparticles. The NFG layer not only protects the metal-core but also causes localization of the π-conjugated system induced by nitrogen doping in graphene. Therefore, the ORR activity can be expected to enhance depending on the relationship between the Fermi level of the core metal. In this thesis, the development of a novel synthesis process for the core-shell nanoparticles by the solution plasma (SP) method as well as their ORR properties and corrosion resistance were evaluated. The possibility to overcome the trade-off characteristics by the proposed structure is verified.

Chapter 1 provides the overview of the current state of the world regarding PEMFC, redox platinum catalysts, and the metal-carbon core-shell structure synthesis techniques and clarifies the superiority of the method applied in this study, namely the SP method for synthesizing both the metal-core and the NFG-shell.

Chapter 2 shows the results of the synthesis, structural analysis and corrosion resistance test of the core-shell nanoparticles with Cu-core (Cu-NFG) prepared by the SP method. The synthesized Cu-NFG consist of a shell of 3-5 layers of nitrogen-doped graphene and a core of Cu nanoparticles. As a result of an immersion test in an acidic solution for 48 hours, the elution of the Cu-core in the presence of NFG-shell was revealed to be reduced to below one-tenth compared to that of bare Cu nanoparticles.

Chapter 3 shows the results of the synthesis and catalytic performance of the core-shell nanoparticles with Pt-core (Pt-NFG) prepared via the SP method. Compared with the commercial Pt/C catalyst, the synthesized Pt-NFG nanoparticles have higher reaction selectivity against the 4-electron pathway for ORR in acidic medium. Furthermore, it demonstrated that the durability in immersion tests is improved compared to that of the commercial Pt/C catalyst.

Chapter 4 shows the results on the synthesis and catalytic performance of the core-shell nanoparticles with Pt-based bimetallic core (PtM-NFG) prepared using the SP method. It was found that a higher ORR performance than the Pt-NFG nanoparticle can be obtained when the Pt-core is replaced with the PtPd bimetallic nanoparticle.

Finally, Chapter 5 summarizes the results of Chapters 2 to 4 and concludes with a general remark. It was shown that the formation of a core-shell structure by coating the metal catalyst with the NFG-shell makes it possible to prevent corrosion from the surrounding acidic solution without deactivating the catalyst properties of the metal-core. Because the results of this study reveal the possibility of developing a novel catalyst with higher catalytic performance and durability than Pt by designing the bimetal for the metal-core and type and amount of dopant in the graphene shell, an alternative catalyst for PEMFC with low-cost must be developed in the near future by utilizing material design methods such as first-principles calculations.