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Article / Book Information

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論 文 要 旨

THESIS SUMMARY

系・コース： Department of, Graduate major in	応用化学 系 応用化学 コース	申請学位（専攻分野）： Academic Degree Requested	博士 Doctor of	(Engineering)
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要旨（英文 800 語程度）
Thesis Summary（approx.800 English Words ）

This thesis, entitled "Structural Effects of Connected Nanoparticle Catalysts on Oxygen Reduction Performance", aims to enhance the performance of oxygen reduction reaction (ORR) catalysts for polymer electrolyte fuel cells (PEFCs). This study focuses on the development of a carbon-free catalyst with a network structure formed by the connection of Pt-based nanoparticles, which is distinct from conventional catalysts, and a systematic investigation of the structural effects of connected nanoparticle catalysts on ORR performance. The thesis comprises five chapters.

Chapter 1, "Introduction" provides an overview of Pt-based catalysts for ORR in PEFCs, showing the structural parameters that influence ORR activity and the importance of modifications in Pt lattice and electronic structures at the catalyst surface to enhance ORR activity. Furthermore, it also explains the issues of catalyst degradation caused by potential changes during PEFC operation and shows the necessity of developing highly durable catalysts. This underscores the structural uniqueness of connected nanoparticle catalysts without carbon support (carbon-free), and the importance of elucidating the structural factors that contribute to their high activity and durability.

Chapter 2, "Investigation of carbon-free connected effect of Pt catalysts on ORR performance", investigates the influence of the carbon-free connected nanoparticle structure, a characteristic of the developed catalysts, on ORR activity. To isolate structural factors, three types of Pt catalysts with similar crystalline sizes have been synthesized: isolated Pt nanoparticles on carbon support, connected Pt nanoparticles on carbon support, and carbon-free connected Pt nanoparticles. The results indicate that both the carbon-free structure and the connected nanoparticle structure contribute to the enhanced ORR specific activity. Furthermore, structural analysis reveals that the connected Pt nanoparticles are found to have locally distorted, shorter lattice spacing where they are connected, indicating that this lattice strain would be a contributing factor to the high activity.

Chapter 3, "Synthesis and characterization of carbon-free connected Pt–Co nanoparticles catalysts with chemically ordered structures", describes the development of carbon-free connected Pt_x–Co_1 catalysts with controlled metal composition and chemically ordered structure, and the investigation of their ORR activity and load cycle durability. These results indicates that the connected Pt_3–Co_1 nanoparticle catalyst with a highly ordered structure exhibits both high ORR specific activity and enhanced load cycle durability. Higher ordered degree effectively suppresses metal dissolution against load potential cycles. Additionally, the surface of connected nanoparticles with a thin atomic Pt rich layer on a Pt_3–Co_1 ordered alloy structure is shown to be stable and highly active.

Chapter 4, "Investigation of metal composition effect of connected Pt–alloy nanoparticle catalysts on ORR performance", further investigates the effect of metal composition on ORR performance by developing connected Pt-based ternary and high-entropy alloy nanoparticle catalysts. The results show that in the connected nanoparticles, high-entropy Pt alloys promote the formation of chemically ordered structures compared to binary Pt-alloys. The connected high-entropy alloy nanoparticle catalysts with a highly ordered structure are demonstrated to maintain highly active surfaces against load potential cycling. Moreover, a volcano-type correlation between lattice spacing and ORR specific activity is observed for carbon-free connected Pt-alloy nanoparticle catalysts, providing guidelines for designing suitable structures for ORR.

Chapter 5, “Conclusion and prospect”, summarizes the achievements of this thesis and provide future prospects based on the results obtained in each chapter.

In summary, this thesis systematically controls the structures of carbon-free connected Pt-based nanoparticle catalysts and clarifies the effects of the structures such as carbon-free, connected nanoparticles, chemical ordering, and metal composition including high-entropy alloys on ORR performance. The findings in this study provide design guidelines for highly active and durable ORR catalysts for next-generation PEFCs and can be applied to other energy conversion catalysts, thus making significant contributions to the field of engineering.