T2R2 東京科学大学 リサーチリポジトリ
Science Tokyo Research Repository

論文 / 著書情報
Article / Book Information

題目(和文)	
Title(English)	Structural Effects of Connected Nanoparticle Catalysts on Oxygen Reduction Performance
著者(和文)	LIAOQiancheng
Author(English)	Qiancheng Liao
出典(和文)	学位:博士(工学), 学位授与機関:東京科学大学, 報告番号:甲第303号, 授与年月日:2025年3月26日, 学位の種別:課程博士, 審査員:山口 猛央,多湖 輝興,横井 俊之,松本 秀行,黒木 秀記
Citation(English)	Degree:Doctor (Engineering), Conferring organization: Institute of Science Tokyo, Report number:甲第303号, Conferred date:2025/3/26, Degree Type:Course doctor, Examiner:,,,,
学位種別(和文)	博士論文
Category(English)	Doctoral Thesis
種別(和文)	要約
Type(English)	Outline

Powered by T2R2 (Science Tokyo Research Repository)

Thesis Outline

Department of Chemical Science and Engineering

School of Materials and Chemical Technology

February 26, 2025

Liao Qiancheng

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 provides an extensive overview of Pt-based ORR catalysts in PEFC applications. It highlights the significance of structural parameters that influence ORR activity, emphasizing the critical role of modifications in Pt lattice and electronic structures at the catalyst surface in enhancing catalytic performance.

Chapter 2 investigates the influence of the carbon-free connected nanoparticle structure 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.

Chapter 3 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 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 summarizes the achievements of this thesis and provides 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.

Flowchart illustrating the content of this study