論文 / 著書情報
Article / Book Information

題目(和文)	理論計算及び実験的検証による窒素ドーピングした炭素層被覆金属微粒子上での電極触媒反応機構の包括的解明
Title(English)	Towards Comprehensive Understanding of N-doped-Carbon Coated Metal Catalysts for Electrochemical Reactions: Theoretical Calculation and Experimental Validation
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Category(English)	Doctoral Thesis
種別(和文)	論文要旨
Type(English)	Summary

（博士課程）
Doctoral Program

論 文 要 旨

THESIS SUMMARY

専攻： Department of	Electronic Chemistry 専攻	申請学位（専攻分野）： Academic Degree Requested	博士 (Engineering) Doctor of
学生氏名： Student's Name	Seunghyo Noh	指導教員（主）： Academic Advisor(main)	北 村 房 男
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要旨（英文 800 語程度）
Thesis Summary（approx.800 English Words ）

Non-precious metal catalysts have attracted a great deal of attention in the field of oxygen reduction reaction catalyst in both acidic and alkaline media due to its relatively cheap material cost compared to highly expensive Pt or noble catalysts. Unfortunately, however, the performance and long-term stability of non-precious catalysts have not reached at the grade of commercialization.

As a new type of non-precious catalyst, N-doped carbon coated metal catalyst is a promising candidate for alternative of Pt catalysts. The encapsulated types own relatively strong resistance for metal dissolution due to their outermost carbon shells. Hou et al. reports that Fe_3C@N-C catalyst has high performance in both acid and alkaline solutions. Deng et al. also presents that Fe encapsulated with N-doped bamboo structure carbon nanotube (Fe@N-CNT) has a higher activity for hydrogen evolution reaction. High activities of M@N-C catalysts are studied by several research groups. However, the active site of M@N-C and the reaction mechanism on it are understood deeply. In addition, the effect of core metal and alloy may suggest an enhanced performance.

This thesis tries to understand the underlying mechanism of catalytic reactions and propose the trend of catalytic design. Chapter 2 explains the principles of experimental characterization and calculation techniques, which are utilized in this thesis. The results and discussion are written from chapter 3 to chapter 7. Chapter 3 introduces why the metal support can enhance the ORR activity on N-doped graphene, and this discussion becomes a main theoretical support to explain the activity of the metal encapsulated at N-doped carbon shells. The actual synthesis of the materials is carried out in chapter 4. In this chapter, copper encapsulated N-doped carbon (Cu@N-C) was fabricated as a promising ORR catalyst based on the theoretical analysis (Chapter 3). Furthermore, the thickness effect was the main issue to enhance the catalytic performance of the carbon encapsulated structure. In practical, Cu@N-C synthesized by annealing process shows a substantial enhancement of its ORR activity in alkaline media by the CO_2 oxidation treatment. Unfortunately, the catalyst, however, has poor ORR activity in acid solution despite the outstanding performance in alkaline. The development of high performance catalysts has been still pointed out as an impregnable barrier in acidic media because it was easy to overcome neither activity nor long-term stability. The carbon encapsulated structure in Chapter 3 and 4 was considered as an effective approach to prevent the degradation of catalytic performance owing to its carbon shells. On the other hand, the improvement of catalytic performance is complicated because of 1) thickness effect of the carbon layer stated in Chapter 4, 2) nitrogen doping level and 3) position included in carbon shell, the species of inner metals (e.g., Fe, Co and Ni) or alloying composition of them. It is difficult to carry out every combination by try-and-error. The computational material design is efficiently utilized to exclude the poor catalytic structures and to achieve the optimal design in Chapter 5. By screening the materials, FeCo@N-C is chosen as the highly active catalyst theoretically and FeCo@N-C/KB catalyst synthesized experimentally presents a high ORR and HER performance in acidic solution.

The main contribution of this thesis is to understand and develop the metal encapsulated with nitrogen doped carbon materials. In order to develop these catalysts, sometimes the study of conventional catalysts such as Pt-alloy may provide us any idea or way. Chapter 6 is the theoretical study related to Pt-based ternary alloy to understand the enhanced durability. This study shows binding energies between metal and oxygen, the size effect of catalyst, information of alloy formation by energy convex hulls. In particular, the technique of energy convex hull utilized in this study gives us an idea to design of alloying metal in Chapter 4. This study showed that inner metals surrounded by Pt skin have an essential effect to determine the activity and degradation of catalytic performance, i.e., the configuration of inner metals can affect a variety of parameters. The view point was underlying hypothesis in Chapter 5. Therefore, Chapter 6 includes underlying information to enhance the performance of non-precious catalyst as well as the design of durable Pt-ternary catalyst.

Chapter 7 is the conclusion of this thesis and synthetically states the understanding of M@N-C catalysts. In addition, further works are suggested to pave the way for an advanced catalyst.

備考 ： 論文要旨は、和文 2000 字と英文 300 語を 1 部ずつ提出するか、もしくは英文 800 語を 1 部提出してください。
Note：Thesis Summary should be submitted in either a copy of 2000 Japanese Characters and 300 Words (English) or 1copy of 800 Words (English).

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