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Title	ナノ触媒におけるグラフェン酸化物の有効活用に関する研究
Author(s)	高, 策
Citation	
Issue Date	2025-09
Type	Thesis or Dissertation
Text version	ETD
URL	https://hdl.handle.net/10119/20095
Rights	
Description	Supervisor: 谷池 俊明, 先端科学技術研究科, 博士

Japan Advanced Institute of Science and Technology

Doctoral Dissertation

Maximizing Potential of Graphene Oxide for Nanocatalysis

Gao Ce

Supervisor: Toshiaki Taniike

Graduate School of Advanced Science and Technology

Japan Advanced Institute of Science and Technology

Materials Science

September 2025

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Graphene oxide (GO) and its derivatives such as graphene oxide frameworks (GOFs) are widely used as supports in the field of catalysis due to their high specific surface area, and abundance of various functional groupsAlthough properties of GO significantly influence the synthesis of their derivatives and interaction with the supported metal nanoparticles (NPs), the relationship between the properties of GO and the performance of the resultant nanocatalysts has not been systematically studied, which corresponds to the aim of this thesis.

In **Chapter 2**, GOFs have great potential as supports for metal NPs, due to their well-defined and engineerable pores. Since GOFs are prepared by bridging GO sheets with linker molecules, their properties depend on those of GO as the starting material. However, the diversity in the physicochemical properties of GO has prevented understanding key parameters for catalyst design. To address this issue, in this chapter, GOFs and GOF-supported Pd NP catalysts (Pd@GOFs) were synthesized from three GO samples with different properties. They were subjected to systematic characterization and catalytic recyclability test in Suzuki-Miyaura cross-coupling. The major findings are: The density of linker molecules in GOF is crucial for enhancing catalytic recyclability, which is influenced by the abundance of reactive in-plane functional groups in the raw GO samples. The gallery spaces between GO layers are important to maintain uniform dispersion of Pd NPs. GO with too small sheets cannot create such gallery spaces, leading to significant aggregation of NPs. Although GOFs have been demonstrated to be useful for various applications, the importance of the quality of the raw material, GO, has sometimes been overlooked. The results presented in this chapter, which assess the quality of GO samples using a multifaceted method, is valuable for fundamental research and design of nanocatalysts.

In **Chapter 3**, the intellectual design of catalysts is pivotal for developing advanced materials and enhancing various catalytic reactions, including electrochemical water splitting for sustainable energy production. Among different catalysts, Pd on GO supports has shown promise for the hydrogen evolution reaction (HER). Although numerous studies have utilized GO as a support material for HER catalysts, research on which specific properties of GO affect catalytic performance remains relatively scarce. Therefore, in this chapter, various GO materials with different characteristics were selected to synthesize GO-supported Pd NP catalysts. The catalysts were evaluated in HER. The properties of the GO materials were characterized using multiple techniques, and their catalytic performance was evaluated through linear sweep voltammetry (LSV) and electrochemical surface area normalization. The LSV results are shown in Figure 2. Firstly, the Pd NP catalyst supported on GOFs did not exhibit promising performance (not shown), but those of GOs showed reasonable performance, which is apparently an opposite trend compared to the Suzuki-Miyaura reaction discussed in Chapter 2, suggesting the importance of diffusion resistance in HER. Among all the catalysts, Pd/GO-GNP (GO prepared from graphene nanoplates (GNP)) exhibited the best HER performance, with the largest electrochemically active

In **Chapter 4**, GOFs using several different linkers with varying functional groups were synthesized, aiming to investigate their effects on the structural and catalytic performance of GOF-confined catalysts. FTIR and XRD analyses revealed that linker types and amounts influence GOF properties, such as interlayer spacing and functional group retention. Pd@GOF catalysts, synthesized by incorporating

palladium NPs into GOFs, were tested for their performance in the Suzuki-Miyaura coupling reaction. Catalytic tests demonstrated that the linker's functional groups significantly affect activity and durability, highlighting the importance of balanced functional group availability and sufficient pillar density for retaining Pd NPs. Comparisons between GOF-confined and GO-supported catalysts indicated that GOFs effectively prevent NP agglomeration while offering enhanced recyclability. The study also noted that commercially sourced GO, with fewer in-plane functional groups, impacted catalytic retention. These findings provide valuable insights into designing high-performance GOF-based nanocatalysts by optimizing linkers and GO surface functionalization to meet diverse catalytic requirements.

In conclusion, this thesis highlights the pivotal role of GO's physicochemical properties and linker design in optimizing nanocatalyst performance. By systematically exploring the relationship between GO characteristics, linker functionality, and catalytic behavior, the findings offer a comprehensive understanding of material design strategies. This work provides a foundation for developing high-performance GOF-based catalysts tailored for specific reactions, bridging fundamental research and practical applications in sustainable catalysis and energy production.

Keywords: Graphene oxide; Nanocatalysts; Graphene oxide framework, Suzuki-Miyaura coupling reaction,