

論文 / 著書情報
Article / Book Information

題目(和文)	
Title(English)	Plasma-in-Liquid Synthesis of Sulfur-Doped Nano-Sized Carbon Materials for Secondary Batteries and Electrocatalysts
著者(和文)	KIMHanvin
Author(English)	Hanvin Kim
出典(和文)	学位:博士(工学), 学位授与機関:東京工業大学, 報告番号:甲第12082号, 授与年月日:2021年9月24日, 学位の種別:課程博士, 審査員:竹内 希,千葉 明,藤田 英明,野崎 智洋,赤塚 洋,Oi Lun Li
Citation(English)	Degree:Doctor (Engineering), Conferring organization: Tokyo Institute of Technology, Report number:甲第12082号, Conferred date:2021/9/24, Degree Type:Course doctor, Examiner:,,,,,
学位種別(和文)	博士論文
Category(English)	Doctoral Thesis
種別(和文)	論文要旨
Type(English)	Summary

論文要旨

THESIS SUMMARY

系・コース：	電気電子	系
Department of, Graduate major in	電気電子	コース
学生氏名：	18D18340	
Student's Name		

申請学位(専攻分野)：	博士	(工学)
Academic Degree Requested	Doctor of	
指導教員(主)：	竹内 希	
Academic Supervisor(main)		
指導教員(副)：		
Academic Supervisor(sub)		

要旨 (英文 800 語程度)

Thesis Summary (approx.800 English Words)

Chapter 1. Introduction

The relationship between the carbon cycle and the environment, and the importance of carbon utilization as an energy material for eco-friendly technologies were introduced. Various approaches focused on changing paradigms were used to prepare carbon materials; among them, the synthesis of carbon using the plasma-in-liquid process was reported as an eco-friendly and efficient route. The breakdown mechanism of the plasma-in-liquid was introduced, and the theoretical background of the SIBs and the ORR was examined with a focus on energy and environmental applications. A more optimized strategy seems to be required for interdisciplinary research linking plasma science, carbon-based materials, and industrial applications. Therefore, in this chapter, new strategies aimed at improving SIBs anodes and ORR catalysts for advanced energy and environmental applications were examined.

Chapter 2. Research trends and literature reviews

The literature on the mechanisms of carbon synthesis via the plasma-in-liquid process was reviewed. Furthermore, the literature reported to date on the applications of carbon-based materials synthesized through the plasma-in-liquid process, especially as secondary battery anode materials and ORR catalysts, was carefully reviewed. Additionally, research trends were summarized by compiling statistics on related scientific papers. Furthermore, to provide perspectives regarding this field, we have tried to briefly summarize the synthesis methodologies and electrochemical performances of these materials for each application, making it easier to understand at a glance. I hope that this thesis will provide a steering wheel for researchers working in this field to set directions for future research.

Chapter 3. Sulfur-Doped Carbon for SIBs Anode Material

SIBs have recently attracted considerable interest due to the plentiful supply of raw materials for their production and their electrochemical behavior, which is similar to that of lithium-ion batteries (LIBs). However, the relatively larger radius of sodium ions than that of lithium ions is not suitable for storage in conventional graphite, which is widely used as the anode. To resolve this issue, in this study, we developed a new harmonized carbon material with a three-dimensional (3D) grapevine-like structure and a sulfur component using an efficient synthesis process. Based on these advantages, the harmonized sulfur-carbon material exhibited a high reversible capacity of 146 mA h g⁻¹ at an extremely high specific current of 100 A g⁻¹ and long-term galvanostatic cycling stability at 10 and 100 A g⁻¹ with superior electrochemical performance. Our results are anticipated to provide new insights into SIBs anode materials that would advance their production.

Chapter 4. Sulfur-Doped Carbon for Two-Electron-Transfer Dominant ORR

We propose a simple sulfur-doped disordered carbon material which catalyzes the ORR predominantly via a 2-electron-transfer process. All the prepared catalysts exhibited a superior HO₂⁻ selectivity of ~75%, and the most positive onset potential, highest current density, and most improved electrocatalytic activation were demonstrated for the sample annealed at the highest temperature. Our results are expected to provide a new perspective on the ORR via the 2-electron-transfer process on sulfur-doped disordered carbon materials.

Chapter 5. Investigation of Carbon Nanoparticles Structure for ORR

Carbon-based materials have been widely studied as promising energy conversion materials that can replace noble metal catalysts. In addition, the plasma-in-liquid process has recently been used to synthesize heteroatom-doped carbon materials for oxygen reduction reactions, such as nitrogen-, boron-, and halogen-doped carbon. However, there has been a lack of insight into pristine carbon, which is the base material for such reactions. In this study, pristine carbon materials (Cx-P) were synthesized using a plasma-in-liquid process, and the material properties and ORR performance were analyzed with respect to heat treatment. Cx-P was successfully synthesized as a carbon nanoparticle and redesigned as partially crystallized carbon with various pore-size distributions using heat treatment. The 2-electron-transfer ORR mechanism was identified as the intrinsic reduction route. On the other hand, the electrochemical activity increased with heat treatment, showing an unexpectedly superior kinetic current density of $\sim 18.30 \text{ mA cm}^{-2}$ (at 0.5 V vs. RHE). Our results are expected to provide an important reference to the ORR performance of the relevant carbon materials, including insights into the basic material properties of partially crystallized porous carbon synthesized using the plasma-in-liquid process.

Chapter 6. Optimized Preparation of Sulfur-Doped Carbon for SIBs and Two-Electron-Transfer-Dominant ORR

Multifunctional disordered sulfur-doped carbon catalyst is demonstrated for use in both sodium-ion-exchange and the 2-electron-transfer-dominant ORR for energy storage and conversion applications. The disordered sulfur-doped carbon prepared via a simple plasma-in-liquid approach showed distinct performance for each electrochemical application, and the underlying fundamental mechanisms were experimentally demonstrated. Benefiting from the enhanced reaction kinetics derived from C-S covalent bonds, superior sodium-ion-exchange performance with a high reversible capacity of $\sim 150 \text{ mA h g}^{-1}$ over 5000 cycles is achieved with a stable rate capability. In addition, an efficient 2-electron-transfer-dominant ORR with a high current density of $\sim 25.66 \text{ mA cm}^{-2}$ at a low overpotential of $\sim 0.890 \text{ V}$ (vs. RHE) is obtained. This work cautiously proposes the importance of sulfur bonded to carbon to boost its electrochemical performance and is expected to provide new perspectives for various interdisciplinary applications.

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 1 copy of 800 Words (English).

注意：論文要旨は、東工大リサーチリポジトリ(T2R2)にてインターネット公表されますので、公表可能な範囲の内容で作成してください。

Attention: Thesis Summary will be published on Tokyo Tech Research Repository Website (T2R2).