

論文 / 著書情報
Article / Book Information

題目(和文)	
Title(English)	Study on novel synthesis of porous carbon microspheres dispersed with iron compound nanoparticles and their application to rechargeable batteries
著者(和文)	ADIAkylbek
Author(English)	Akylbek Adi
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12552号, 授与年月日:2023年9月22日, 学位の種別:課程博士, 審査員:谷口 泉,久保内 昌敏,山中 一郎,平山 雅章,森 伸介
Citation(English)	Degree:Doctor (Engineering), Conferring organization: Tokyo Institute of Technology, Report number:甲第12552号, Conferred date:2023/9/22, 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 : Department of Graduate major in	応用化学 応用化学	系 コース	申請学位 (専攻分野) : Academic Degree Requested	博士 Doctor of	Engineering
学生氏名 : Student's Name	Adi Akylbek		審査員主査 : Chief Examiner	Taniguchi Izumi	

要旨 (英文 800 語程度)

Thesis Summary (approx.800 English Words)

This work developed novel synthesis methods based on spray pyrolysis to prepare porous carbon microspheres dispersed with iron compound nanoparticles, and systematically studied their physical and electrochemical properties as anode and electrocatalyst materials for Li-ion and Li-S batteries, respectively.

Chapter 2 reported on a synthesis of porous carbon microspheres dispersed with Fe_3O_4 nanoparticles (porous C/ Fe_3O_4 microspheres) as an anode material for Li-ion batteries using spray pyrolysis with a sodium nitrate (NaNO_3) additive in the precursor solution. The XRD, SEM, TEM, and CHNS results showed that NaNO_3 catalyzes the carbonization of sucrose and enhances the crystallization of Fe_3O_4 . Moreover, nitrogen adsorption/desorption analysis showed that the aqueous removal of sodium compounds produces porous C/ Fe_3O_4 microspheres with large surface areas and pore volumes. The porous C/ Fe_3O_4 microspheres prepared by spray pyrolysis with a 1.00 M NaNO_3 additive exhibited a reversible capacity of $\sim 780 \text{ mAh g}^{-1}$ in the initial cycles and $\sim 520 \text{ mAh g}^{-1}$ after 30 cycles at a current density of 50 mA g^{-1} . The wide range of pores produced from the removal of sodium compounds might enable easy electrolyte penetration and facilitate fast Li-ion diffusion, leading to the improved electrochemical properties of this sample.

In Chapter 3, a novel synthesis route was developed based on spray pyrolysis to further improve the porous and crystal structure of the carbon microspheres dispersed with Fe_3O_4 nanoparticles (porous-crystalline C/ Fe_3O_4) as anode materials for Li-ion batteries. The porous-crystalline C/ Fe_3O_4 microspheres were prepared by sequentially annealing the spray-pyrolysis synthesized dense-amorphous C/ Fe_3O_4 microspheres under reducing and oxidizing atmospheres. The post-heat treatments provided an easy way to crystallize the amorphous carbon, introduce micro- and meso-pores, and obtain mixed-valence iron oxides such as Fe_3O_4 . The porous-crystalline C/ Fe_3O_4 microspheres demonstrated an improved specific capacity of $\sim 560 \text{ mAh g}^{-1}$ at a current density of 50 mA g^{-1} , which corresponded to 92% of their theoretical value. The porous-crystalline C/ Fe_3O_4 microspheres also demonstrated higher specific capacities at high current densities than the porous C/ Fe_3O_4 microspheres prepared by spray pyrolysis with a 1.00 M NaNO_3 additive in the previous chapter. The significant improvements in their electrochemical properties of the porous-crystalline C/ Fe_3O_4 microspheres were attributed to the synergy between well-developed porous structure and highly conductive graphitic carbon, which provided simultaneously higher Li-ion and electronic conductivities throughout the entire microspheres.

In Chapter 4, the C/ Fe_3O_4 microspheres discussed in Chapter 3 were used to investigate the synergetic

effect of conductive and adsorptive/catalytic interfaces on the adsorption and conversion of polysulfides, aiming to enhance the electrochemical performances of the Li-S batteries. The porous-crystalline C/Fe₃O₄ microspheres had a wide range of micro-and mesopores to physically adsorb polysulfides and highly accessible Fe₃O₄ nanoparticles to chemically immobilize them within the electrochemical interfaces and promote their conversion into short-chain sulfides. As a result, the porous-crystalline C/Fe₃O₄ microspheres enhanced sulfur utilization and accelerated redox kinetics of the Li-S batteries when used as modified separators. This work demonstrated that high-performance Li-S batteries could be achieved by rationally designing conductive and adsorptive/catalytic interfaces of the C/Fe₃O₄ microspheres.

Despite the improved electrochemical performance of the Li-S batteries, the multi-step procedures involved in preparing the sulfur cathodes and modified separators may restrict the large-scale application of these batteries.

Therefore, Chapter 5 introduced the synthesis of the advanced sulfur cathode incorporating porous-crystalline C/Fe₃C microspheres as an electrocatalyst and CFP as a 3D current collector through vacuum filtration, eliminating the complex procedures in the production of cathode materials for Li-S batteries. As 3D current collector, CFP offered transfer of electrons and additional nucleation sites for the deposition of the end redox products. Meanwhile, the porous-crystalline C/Fe₃C microspheres effectively immobilized polysulfides and promoted their fast conversion and nucleation. This study demonstrated the potential for achieving high-performance Li-S batteries without the complex procedures associated with the production of cathode materials.

Overall, Chapters 2 and 3 demonstrated that the porous structure of the C/Fe₃O₄ microspheres facilitates Li-ion diffusion, leading to the better utilization of Fe₃O₄ nanoparticles in Li-ion batteries. Moreover, the crystallization of amorphous carbon into graphitic one can provide higher electronic conductivities throughout the entire microsphere, further enhancing the electrochemical performances of the samples.

Chapters 4 and 5 demonstrated that high-performance Li-S batteries could be achieved by rationally designing conductive and adsorptive/catalytic interfaces of the C/Fe₃O₄ and C/Fe₃C microspheres. Moreover, the porous-crystalline C/Fe₃C microspheres outperformed the porous-crystalline C/Fe₃O₄-microspheres as electrocatalysts in Li-S batteries due to the higher electronic conductivity of Fe₃C nanoparticles.

The obtained results confirm the importance of the developed synthesis methods in tuning the physicochemical properties of the composite microspheres and enhancing the electrochemical properties of the rechargeable batteries. Moreover, these methods could be easily adopted for the preparation of carbon microspheres dispersed with other transition metal oxides. Therefore, further work should focus on synthesizing these composite microspheres and studying the applications of these composite microspheres for various energy storage systems and beyond.

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).