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主 論 文 の 要 旨

論文題目 Solution Plasma-Based Synthesis and Characterization of Carbon Electrode Materials （ソリューションプラズマを用いたカーボン電極材料の合成と評価）

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論 文 内 容 の 要 旨

Abstract

A Specific type of liquid phase electrical discharge processes, named solution plasma process (SPP), have been exhibit an excellent potential of synthesis various types of nanoparticle. The unique properties and novel reaction kinetics of SPP successfully applied to carbon synthesis process. In this process, benzene was served as the carbon precursor, while gold and platinum electrodes produced catalytic metal nanoparticle supported on carbon through electrode sputtering during the discharge process.

A new class of structure-controlled carbon nanoballs (CNBs) were synthesized. The pore structure and graphitation degree can be precisely controlled by the pulse frequency of SPP. The structural characterization revealed that CNBs which synthesized with low frequency have excellent meso-macro hierarchical pore structure, with an averaged diameter size of 14.5 nm and a total pore volume of 1.13 cm3g-1. The CNBs formed by the aggregation of carbon exhibited inter-connected pore channels in different directions on both meso- and macrometer length scales. As for the application in cathode material in Li — O2 battery, the discharge capacity of as-prepared carbon reached 3600 mAhg-1, which exceeded the capacity of commerical carbon material by 30 〜 40 %. The excellent discharge capacity was contributed by the co-existence of high pore volume and meso-macro hierarchical porous structure. The structural and electrochemical properties accompanied with the synthesis mechanism of CNBs were discussed in details.

In addition, high-electrocatalytic-activity noble nanoparticles (NPs) were also successfully fabricated on CNBs from metal electrodes via a one-step solution plasma process in benzene. TEM images showed that spherical Au and Pt NPs were uniformly dispersed over the entire surface of the CNBs, and the average particle size was below 10 nm after heat treatment. Cyclic voltammetry showed clear peaks corresponding to the oxidation and reduction

features in the catalytic reaction. This indicated the high electrocatalytic activity of the Pt/CNB and Au/CNB electrodes in the oxygen reduction reaction, which is an important factor in terms of catalytic reactions. This proved that SPP is a simple one-step method for the large-scale synthesis of NPs/carbon and, and this NPs/CNB is possible to apply as the next generation of electrode materials in Li — O2 battery.
Therefore, SPP holds great potential as a candidate for next-generation synthetic methods for the production of CNBs or NPs/carbon materials, and might bring the application of Li — O2 battery one step forward.

Outline of thesis

Chapter 1 describes the fundamental of solution plasma process, and its advantages, and its applications. Additionally, the overview of Li — O2 battery and porous carbon materials are introduced.

Chapter 2 describes the synthesis carbon nanoparticles by SPP process. Carbon nanoparticles were formed by carbon atoms supplied by two different groups of organic solutions, sugar and aromatic hydrocarbon solutions. Structural properties of these synthesized carbon materials are described, and suggested suitable solution for the synthesis of carbon nanoparticles in solution plasma process. In addition a comparison of fundamental characteristics of plasma discharges under organic solution were conducted.

Chapter 3 focuses on the structure-controlled carbon nanoballs (CNBs) were synthesized by various parameters of SPP. The change of the pore structure and graphitation degree of CNSs are described and have been published.

Chapter 4 presents the structural analysis of CNBs having hierarchical porous structure and its performance in Li — O2 battery system. In addition, Ketjen Black EC-600JD (KB, Mitsubishi Chemical, Japan), of which demonstrated the highest capacity among commercial carbon materials, was used to compared to the electrochemical performance of CNBs and to find out the key factors. The results have been submitted.

Chapter 5 describes the synthesis of high-electrocatalytic-activity noble nanoparticles (NPs) supported on CNBs using SPP to decrease over-potential on ORR/OER. This technique applied a one-step method for the synthesis of NPs on carbon materials. Benzene is used as carbon precursor while gold (Au) or platinum (Pt) nanoparticles are generated instantaneously via sputtering from metal electrodes. The experimental results have also been published.

Chapter 6 summarizes all chapters and concludes the major finding in the thesis.