論　文　の　要　旨

題　目: SYNTHESIS OF NANOSTRUCTURE-CONTROLLED SPHERICAL CARBON PARTICLES AND THEIR APPLICATION TO ENERGY STORAGE DEVICES
(ナノ構造が制御された球状炭素微粒子の合成とエネルギー貯蔵デバイスへの応用)

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The fast-growing energy crises and depletion of fossil fuels as well as the emission of greenhouse gases lead to severe pollution of the environment. Therefore, the development of sustainable energy storage devices (*e.g.* batteries, supercapacitors) is very crucial for solving energy issues, which requires new and advanced materials as well as novel fabrication techniques in order to achieve high electrochemical performance. In recent years, the rational design and synthesis of nanostructured particulate materials in favorable morphologies have attracted significant attention. Among various materials with different morphologies, carbon spheres have received a great deal of interest due to their regular shape, low surface free energy, good fluidity, high packing density, and chemical stabilization.

The research covered in the scope of this dissertation is aimed to address the following objectives: (i) to develop the methodologies for design and synthesis of spherical carbon-based materials using soft template and template-free spray drying method, particularly the precisely tailored structure, pore texture, and surface properties, (ii) to provide a possible mechanism for obtaining an in-depth understanding of the particle formation of carbon-based materials, and (iii) to show how the morphology, pore size, and material composition may influence lithium-ion batteries (LIBs) and supercapacitors performance. This dissertation comprises five chapters, of which content is described briefly as follows.

Chapter 1 summarizes the important properties of spherical carbon-based materials, the several synthetic strategies for producing carbon sphere materials with desirable structures and functionalities for energy storage applications, and the alternative material in response to the problem suffered by carbonaceous materials. This summary and problem statements lead to the motivation of the researches covered within the scope of this dissertation.

Chapter 2 discusses the developed strategy for fabrication of carbon-coated SiO_x (SiO_x@C core-shell) particles through a sol-gel method using the simultaneous hydrolysis-condensation of tetramethyl orthosilicate (TMOS) and polymerization of 3-aminophenol and formaldehyde followed by the carbonization process. The core−shell particles can be obtained with a well-controlled reaction rate and spherical morphology by using TMOS as an excellent silica precursor for the first time instead of the traditionally used tetraethyl orthosilicate (TEOS). When used as the anode material for LIBs, the obtained SiO_x@C core−shell particles exhibited a reversible capacity of 509.2 mAh g^{-1} at 100 mA g^{-1} and the capacity retention was approximately 80% after 100 cycles. The significantly improved electrochemical performance due to the morphology and structure of the material.

Chapter 3 focuses on the synthesis of spherical carbon particles derived from Kraft lignin via the spray drying method followed by the carbonization process. The developed synthetic strategy is highly significant because it allows the utilization of lignin as an abundant and sustainable carbon precursor. The carbon particles generated in this study could be obtained with spherical morphologies and it could be precisely controlled from dense to hollow sphere by varying the potassium hydroxide (KOH) concentration. In addition, to get a better understanding of the particle formation of carbon particles, a plausible mechanism has been discussed in detail, which provides insights with regard to the exploration of lignin-derived carbon materials. The resulting spherical carbon particles exhibited dense structures with a specific surface area (1233 $m^2 g^{-1}$) and tap density (1.46 g cm^{-3}) superior to those of irregular shape carbon particles. The carbon sphere particles having a high tap density as advanced materials can endow them for a broad prospect of applications in the areas of colloids and interface science as well as sustainable development.

Chapter 4 provides the characterization of hollow carbon spheres (HCSs) derived from Kraft lignin with their structural, pore texture, surface morphology, and electrochemical performance for supercapacitors. HCSs with high specific surface area up to 2424.8 $m^2 g^{-1}$ and micro-mesoporous structure were obtained at low KOH-to-lignin mass ratios (below 1.5), which was in accordance with green chemistry principles. In addition, these HCSs can be used as electrode materials in supercapacitors for energy storage applications. This newly developed synthetic process is expected to lead an advanced carbon material and the value-added utilization of Kraft lignin.

Chapter 5 provides a summary of all chapters and suggestions for future research.