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論 文 内 容 の 要 旨

Graphene has recently attracted much interest as a promising electrode material for electrochemical capacitors owing to its theoretically high specific surface area and good electronic conductivity. Although many studies have been reported, the results are still far from expected from the theoretical values due to unavoidable restacking of graphene. This thesis focuses on the application of graphene-based material as electrodes for electrochemical capacitors, aiming mainly to improve the specific capacitance.

Chapter 1 summarizes fundamental aspects of electrochemical capacitors as well as recent research of electrode materials with particular emphasis on graphene-based materials.

In Chapter 2, a cationic polymer (PDDA) was introduced in an attempt to prevent face-to-face restacking of reduced graphene oxide (rGO). Samples of mono- or multi-layered rGO were fabricated by layer-by-layer self-assembly and reduced by H₂ or N₂H₄. Structural and electrochemical analysis revealed that PDDA acted as a spacer

between mono-layered rGO, successfully preventing restacking of the sheets, enabling full utilization of surface of rGO. The electrical double layer capacitance per surface area of rGO was estimated to be roughly $15 \mu\text{F cm}^{-2}$ and a gravimetric capacitance as $\sim 400 \text{ F g}^{-1}$ was obtained. The results suggest that graphene has a high potential as electrode material with high volumetric capacitance by nanoarchitectural design of electrodes.

The lateral size of individual graphene sheets is expected to impact the electrochemical properties of rGO. In Chapter 3, the lateral size effect on the specific capacitance of rGO was investigated by breaking down the size of graphene oxide dispersed in water using high-frequency ultrasonification. The specific capacitance of rGO prepared by hydrogen reduction of graphene oxide with an average equivalent diameter of 280 nm was 242 F g^{-1} in $0.5 \text{ M H}_2\text{SO}_4$ and 205 F g^{-1} in $0.5 \text{ M Na}_2\text{SO}_4$, respectively, which are $\sim 30\%$ higher than that of the samples without ultrasonic treatment with a diameter of 920 nm. The root causes of this size-effect are discussed based on the microstructure of the rGO electrode as well as the edge/plane ratio.

As concluded in Chapter 4, the fundamental investigation with modeled mono-layer graphene showed a specific electric double layer capacitance of $\sim 15 \mu\text{F cm}^{-2}$, which should offer a much better understanding on the fundamental capacitive properties of individual graphene. In addition, the results of the lateral size effect towards the specific capacitance should also provide a new direction of improving performance of 2-dimensional materials.