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研　究　主　論　文　抄　録

論文題目

Electrochemical Properties and Applications of Graphene Oxide

（酸化グラフェンの電気化学特性と応用）

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主論文要旨

Graphene have been intensively researched to open up new devices in physical field due to have unique electronic structures. Graphene Oxide (GO) prepared by exfoliating graphite oxide in solution had been studied as the precursor material of graphene and tried to obtain graphene by reducing it until a few years ago due to be able to easily synthesize in large scale. However, reduced GO (rGO) is different from intrinsic graphene and exist many defects and remained a little functional groups. GO and rGO exhibit various interesting chemical properties due to its heterogeneous chemical and electronic structures and chemists are increasingly drawn to GO and rGO.

In the present study, we focus on elucidation of electrochemical properties of GO and rGO. We take particular note of two electrochemical properties: electrochemical capacitance properties and oxygen reduction reaction (ORR) properties. rGO has attracted significant interest as an electrode material for electrochemical double layer capacitors due to its high electrical conductivity and high surface area. Also, rGO is covered with functional groups on the sheet surface and these functional groups work as a source of pseudocapacitance. Thus, rGO has potential for electrode materials of electrochemical capacitors. As other properties, GO exhibit negative charge and hydrophilic property owing to many oxygen functional groups of sheet surface. GO can easily combine with other materials such as metal cation, metal complex, and hydrophilic nanomaterials. The hybrid materials after reduction lead up to development of new electrocatalytic materials. Moreover, multilayered GO film (GO paper) exhibits relatively high proton conduction. Applying GO paper to electrolyte, we developed two novel cells: graphene oxide fuel cell (GOFC) and graphene oxide lead battery (GOLB).
The thesis is composed of 7 chapters.

Chapter 1 narrates the general backgrounds, preparation method, properties,

and applications of GO.

Chapter 2 presents the electrochemical properties of GO prepared by the new electrochemical method. We present a new electrochemical oxidation method for preparing of GO, in which a high oxidation voltage is applied at the carbon electrode in pure water. This electrochemical oxidation method can be easily controlled oxidation degree. After electrochemical reduction of GO, the rGO has three electrochemical properties: high electrochemical capacitance, photoelectrochemical property, and electrocatalytic activity for ORR. These electrochemical properties of rGO are due to the CH defects and/or OH groups from rGO.

Chapter 3 presents the electrochemical capacitance of GO. We focus on the effects of the electrochemical oxidation/reduction cycle of GO on its composition and electrochemical capacitance. C=C bonds are produced from CH defects by the electrochemical re-oxidation of rGO, while COC bonds are produced from carbon materials by the first electrochemical oxidation. The electrochemical capacitance of rGO is increase with the electrochemical oxidation/reduction cycle because of the increase of the surface area of rGO. Moreover, we discuss effects of other reduction method on the electrochemical capacitance of rGO by using of X-ray photoelectron spectroscopy. The electrochemical capacitance of rGO is increase with the ratio of CH defects.

Chapter 4 presents the electrocatalytic activity for ORR of GO. The novel iron phthalocyanine/GO (FePc/GO) hybrid is prepared by self-assembly. The FePc/rGO hybrid prepared by electrochemical reduction of FePc/GO hybrid exhibits excellent electrocatalytic activity for ORR compared with the commercial Pt/C catalysts in alkaline medium.

Chapter 5 presents the application of GO as an electrolyte in fuel cell. We studied the performance of fuel cell with GO paper as the electrolyte at low relative humidity and room temperature. This fuel cell showed more excellent fuel cell performance than that of fuel cell with Nafion membrane as the electrolyte. Our study opens up GOFC for a cost-effective next generation fuel cell system.

Chapter 6 presents the application of GO as an electrolyte in lead battery. We fabricated the lead battery with GO paper as the solid electrolyte. This lead battery demonstrated comparable performance to lead battery using H_2SO_4 solution electrolyte. GOLB suggests the high potential for development of a small-sized dry lead battery.

Chapter 7 gives the general conclusion of the thesis.