Low Dimensional Ion-Conducting Nanomaterials for Fuel Cell Membrane Applications

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論 文 名　：Low Dimensional Ion Conducting Nanomaterials for Fuel Cell Membrane Applications
（固体高分子形燃料電池電解質膜用の低次元イオン伝導性ナノマテリアルの開発）

区　　分　：甲

論 文 内 容 の 要 旨

Fuel cells are the key technology in the move towards a “hydrogen society” in Japan. Polymer electrolyte membrane fuel cells (PEMFCs) are to date the most commercially successful type of fuel cell. They are currently being utilized to a limited extent in fuel cell vehicles (e.g. FCV MIRAI, *Toyota*), and micro-combined heat and power plants (e.g. ENE-Farm). Their limited commercialization in broader fields is largely due to their high cost, stemming from the materials used, e.g. the platinum electrocatalyst and the Nafion® proton exchange membrane (PEM). Decreasing the cost of fuel cells can be directly achieved by using cheaper materials, or indirectly by improving device performance and lifetime. Here we attempt this by finding novel membrane materials.

Any new fuel cell membrane material must satisfy three major conditions. First, it must be possible to reproducibly manufacture sufficiently mechanically stable membranes in a scalable manner. Second, such membranes must fulfill the basic requirements for fuel cell operation, namely: water uptake, ionic conductivity, electronic insulation, and sufficient gas barrier properties. Third, it should be possible to incorporate the materials into membrane electrode assemblies (MEAs), where they can be evaluated for their fuel cell performance. Here, two novel low dimensional ionic conductors are investigated as new membrane materials for PEMFCs and alkaline anion exchange membrane fuel cells (AAEMFCs); i.e., graphene oxide and nanocellulose.

In Chapter 1 a detailed introduction about fuel cells is given, including the history and operating principles of fuel cells. The background of graphene oxide and nanocellulose is discussed. The experimental methods used in this work are explained in Chapter 2.

Graphene oxide is explored in detail as a membrane material in Chapter 3. First, membranes are prepared by vacuum-filtration, and their morphology, chemical composition, and mechanical properties are investigated. Next, the suitability of graphene oxide membranes for fuel cell applications is determined by measuring the water uptake, conductivity, and hydrogen gas barrier properties, compared with Nafion. In particular the conductivity (and permittivity) is systematically investigated over a wide temperatures and humidity range by impedance spectroscopy and blocking measurements, giving insight into the conduction

mechanisms. Finally, graphene oxide membranes are incorporated into MEAs and their fuel cell performance is measured. The dependence of fuel cell performance on membrane thickness is investigated, and very thin, electrode-supported membranes are investigated using novel spray-based fuel cell fabrication techniques.

In Chapter 4 the application of graphene oxide membranes in AAEMFCs are investigated. First, graphene oxide in dispersion is reacted with potassium hydroxide in order to introduce mobile OH^- ions, then vacuum filtered to make membranes. The morphology, chemical composition, thermal stability and mechanical properties are investigated. Next, the suitability of GO_{KOH} membranes for alkaline fuel cell applications is determined by measuring the water uptake, ion exchange capacity, conductivity, and hydrogen gas barrier properties, compared with the original GO, and a commercially available anion exchange membrane. In particular the conductivity is systematically investigated over a wide temperature and humidity range by impedance spectroscopy, giving insight into the conduction mechanisms. The dominant charge carrier is confirmed using blocking measurements. Finally MEAs are fabricated and investigated for their performance in an alkaline fuel cell.

Chapter 5 deals with the application of nanocellulose as a fuel cell membrane material. Two varieties of nanocellulose are studied, namely: cellulose nanofibers (CNF), and cellulose nanocrystals (CNCs). First, nanocellulose papers are prepared by vacuum-filtration of nanocellulose water dispersions, and their morphology, chemical composition and mechanical properties are investigated. Next, the suitability of nanocellulose paper for fuel cell membrane applications is investigated by hydrogen gas barrier measurements, water uptake, and conductivity and the results are compared with Nafion. In particular, the conductivity is systematically investigated over a wide temperature and humidity range by the aid of impedance spectroscopy, and the conduction mechanism is elucidated via activation energy determination. Finally MEAs are fabricated with nanocellulose membranes and their fuel cell performance is investigated.

In Chapter 6, the main empirical results are summarized, the conclusions of this work are outlined, and the prospects for application of these novel materials are discussed. Finally, the planned future directions of this work are considered.