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Superhydrophobic Fluorinated Carbons for the Microporous Layer of Polymer Electrolyte Fuel Cells

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論 文 名　：Superhydrophobic Fluorinated Carbons for the Microporous Layer of Polymer Electrolyte Fuel Cells

（固体高分子形燃料電池に用いるマイクロポーラス層用超疎水性フッ素系炭素材料）

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論 文 内 容 の 要 旨

Thesis Summary

Polymer electrolyte fuel cells (PEFCs) are a highly efficient hydrogen based clean energy conversion technology. PEFCs show impressive potential to take down the long ruling internal combustion engine hegemony in the transportation sector. However, the problem of water management at high current density PEFC operations decreases the system efficiency drastically. It is thus important to create and design new materials for the gas diffusion layer (GDL) and/or the microporous layer (MPL) to improve water management as well as PEFC performance. Fluorinated carbons can be a promising alternative as MPL material due to their superior water repellent properties. Therefore, the main objective of this thesis is to increase the PEFC system performance under high current density operations by employing superhydrophobic fluorinated carbons with suitable properties as MPL material.

In Chapter 1, types of fuel cells are briefly explained with the general historical information of fuel cell technology. The general information about PEFCs is provided and its limitations are discussed with the focus on its wide-spread commercialization. The importance and the issue of water management in PEFCs are highlighted. Fluorinated carbon black is proposed as an alternative material for the MPL to improve water drainage speed under high current PEFC operations as well as the system performance. Examples of the usage of the fluorinated carbons in PEFC are given and the lack of investigating fluorinated carbon as MPL material is illustrated.

In Chapter 2, the various experimental methods used in this research are explained in detail.

In Chapter 3, superhydrophobic fluorinated carbons are synthesized and characterized intensively. A new unique synthesis of synthesize fluorinated carbons with a unique solvothermal thermal method with fluorotelomer alcohol precursors and sodium metal is presented. The effect of different fluorotelomer alcohols on chemical structure and microstructure is elucidated. Results show that a higher fluorine content of the fluorotelomer alcohol precursor results in a higher fluorine content in the final product.

Interestingly, all four samples show similar microstructures with non-porous graphitized carbon black. The WCA measurement of materials shows the superhydrophobic nature of the products. Surprisingly, synthesized fluorinated carbons show highly graphitic structures even though there is not a catalyst nor high reaction temperature involved during the synthesis process. Thus, a new HF related catalytic reaction for this graphitic formation is proposed. Additionally, fluorinated carbons are defluorinated with a heat treatment process under an inert atmosphere. Then the effect of fluorination on the graphitic structure and WCA angle is investigated. It is concluded that with fluorination the defect in the graphical domain increases due to the sp3 bonding of fluorine atoms. After defluorination, due to the absence of fluorine atoms, products become more hydrophilic.

In Chapter 4, the effect of superhydrophobic fluorinated carbon black as MPL material is investigated and its performance is compared with commercially used graphitized carbon black. The slurry of MPLs were prepared with a low amount of PTFE binder (5 wt.%) and the slurry was coated onto a GDL with a doctor blade method, followed by a heat treatment process to decompose additives as well as sintering the PTFE. To calculate porosity free standing, MPLs were manufactured by coating the slurry onto a glass plate. Carbon black based MPL (CB-MPL) showed a higher porosity with 83% compared to fluorinated carbon based MPL (FC-MPL) with 78% porosity. Water contact angle (WCA) measurement results elucidated the superior water repellent characteristic of FC-MPL with 151° WCA compared to CB-MPL with 131° WCA. Prepared MPLs performance were compared with an in situ PEFC single test. Results clearly showed higher PEFC performance as well as lower oxygen transport resistance in the structure due to the superior water repellent properties of FC-MPL.

In Chapter 5, the findings of the previous chapters are summarized, and future direction of this research is explained.