

題 目: NANOSTRUCTURED PARTICULATE AND FIBROUS MATERIALS FOR FUEL CELL
ELECTRODE APPLICATION

(粒子状及び繊維状ナノ構造材料の合成と燃料電池電極への応用)

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The objective of this thesis is to systematically investigate the synthesis of nanostructured particulate and fibrous materials using spray processes (i.e. spray pyrolysis and electrospray) and their application. Various types of nanostructured particles such as dense, hollow and porous particles and nanofiber have been developed and their application as electrocatalyst was examined subsequently. The structural properties and electrocatalytic activity of the prepared nanostructured electrocatalyst materials are reported thoroughly.

Chapter 1 describes a general summary of the previous researches and the background as well as the objectives and outline of the present thesis.

The synthesis and evaluation of porous carbon derived from phenolic resin using a fast and facile spray pyrolysis method for use as a new electrocatalyst support material was described in **Chapter 2**. By adding polystyrene latex (PSL) nanoparticles as a template to the phenolic resin precursor, self-organized macroporous carbon structure was first developed. The mass ratio of phenolic resin to PSL at 0.625 gave the optimum porous morphology. Pt nanoparticles (~20wt%) were grown on the carbon surface using a standard industrial impregnation method. Well-dispersed Pt nanoparticles of average size 3.91 nm were observed on the surface of porous carbon particles. The high catalytic performance of porous Pt/C electrocatalyst was confirmed by the high mass activity and electrochemically active surface area, which were 450.81 mA/mg-Pt and 81.78 m²/g-Pt, respectively. The porous Pt/C catalyst obtains two times higher mass activity than that of the commercial Pt/C catalyst and performs excellent durability under acid conditions.

The synthesis of nanostructured carbon particle is challenging because of the poor ability of polymeric precursors to self-assemble during spray pyrolysis. Here, in **Chapter 3**, we report a facile method for designing self-organized nanostructured carbon particles. A dual polymer system of phenolic resin and electrically charged polystyrene latex is ultrasonically spray pyrolyzed to produce either hollow or porous carbon particles.

Chapter 4 describes the synthesis of Pt/SnO₂ nanofibers via electrospinning. The unique electrochemical properties were in evidence based on the activity that allowed a hydrogen oxidation reaction and inhibit an oxygen reduction reaction. A high electrochemically active surface area value of 81.17 m²/g-Pt was achieved with ultra-low Pt loading (4.03wt%). The kinetics of a hydrogen oxidation reaction was investigated using a linear sweep voltammetry technique under a hydrogen atmosphere. A diffusion-limited current was achieved at 0.07 V and was stable at a high potential. This preparation technique shows great promise for the design of anode electrocatalyst material for fuel cells.

A facile method to synthesize Pt nanoparticles on the electrospun SnO₂ fiber matrix is presented in **Chapter 5**. Precursors contained tin chloride pentahydrate, hexachloroplatinic acid, polyacrylonitrile, and *N,N*-dimethylformamide was electrospun subsequent calcination at 500°C for 4 h. It is shown that Pt nanoparticles were grown on SnO₂ fiber matrix without any formation of PtO. Pt nanoparticles size was span in

the range of 6-32 nm, by adjusting the concentration of polyacrylonitrile. The electrochemically active surface area of Pt/SnO₂ fibers was found very competitive to the commercially available electrocatalyst. This work demonstrates that Pt/SnO₂ fibers can be generated by a facile electrospinning process.

The researches of preparation of the nanostructured particles and fibrous materials prepared using spray process and their applications as catalyst are studied. The conclusion of this thesis is mentioned in **Chapter 6**.