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## 主 論 文 の 要 旨

論文題目     **Enhancement of durability by silica coating Pt-based catalysts for fuel cell application**  
(シリカ被覆技術を用いた燃料電池用白金系触媒の耐久性向上)

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## 論 文 内 容 の 要 旨

Since global concerns about the environment and human health, cleaner fuels and more effective energy conversion technologies are developed. Fuel cells are electrochemical devices that convert a fuel with oxygen (oxidant) by chemical reaction into electrical energy. There are many types of fuel cells; this work is focused for proton exchange membrane fuel cell (PEMFC) application. PEMFC is promising alternative energy sources because of advantages such as low temperature operation, low emissions, and high energy efficiency. The state of the art electrocatalyst for cathode electrode is Pt-based catalyst dispersed in the form of nanoparticles on a carbon support, in order to achieve a maximum of active sites. Practical performance, however, not only demands high activities per mass for oxygen reduction reaction (ORR), but also durability against an acidic conditions that occur in the fuel cell under operation, particularly on the cathode electrode. Catalyst degradation mechanisms have been suggested to occur in fuel cells such as platinum dissolution, Ostwald ripening, agglomeration of Pt nanoparticles, and detachment of Pt nanoparticles from carbon support.

A high performance fuel cell catalyst needs long lifetime catalytic activity and to avoid degradation under operation, which is affected a loss of the electrochemically active surface area (ECSA). Many researchers studied about platinum-based alloy catalyst to improve the activity and

durability of the catalysts under PEMFCs cathode conditions. However, the metal species added to the Pt catalysts also eventually dissolve under the cathode conditions. The dissolution of the non-noble transition metal also contributes to deactivation of the catalysts. The dissolved metal species are also deposited in the polymer electrolyte membrane, which results in decreasing of proton conductivity.

As mentioned earlier about the motivations and the problem of Pt-based catalyst for oxygen reduction reaction at cathode electrode of PEMFC. The aim of this thesis is to attain the following objectives:

To enhance durability of Pt-based catalyst (commercial Pt/C) and Pt nanoparticles supported on carbon nanotubes by silica coating to avoid the agglomeration of the Pt nanoparticles and to prevent detachment of Pt nanoparticles from carbon supports with different surfactants and benzoic acid.

The thesis is organized into six chapters, describing the details of my scientific research and the experimental results as follows:

Chapter 1 presents a brief introduction of Fuel cell. An overview of types of fuel cell is then given. Additionally, the challenge of catalyst for oxygen reduction reaction at cathode electrode of PEMFC is introduced.

Chapter 2 silica coating synthesis diagram and experimental setup for Pt supported on carbon nanotubes synthesizing are given. The silica coating on commercial Pt supported on carbon and carbon nanotubes procedure are given in detail.

Chapter 3 describes and discusses the effect of different types of surfactant which use for synthesizing silica-coated commercial Pt supported on carbon (Pt/C) to the enhancement of durability in during operation. The investigation of structural morphology and durability are presented.

Chapter 4 describes and discusses the enhancement of durability of silica-coated Pt supported on carbon nanotubes (Pt/CNT) which has difference structure from Pt/C compare to uncoated Pt/CNT.

Chapter 5 describes and discusses the effect of amount of benzoic acid which use in synthesizing silica-coated Pt/C to the enhancement of durability in an acidic electrolyte during operation.

Chapter 6 summarizes all chapters in the thesis.