

博士論文（要約）

Electrophoretic deposition of carbon nanostructured catalyst supports for anode electrodes used in direct ethanol fuel cells

（電気泳動堆積法を用いた直接エタノール型燃料電池のためのカーボンナノ構造触媒担体）

アハマディ ダリアケナリ アハマド

Ethanol, with its high energy density, production from renewable sources, facility in storage and distribution, is almost the ideal combustible for fuel cells wherein its chemical energy can be converted directly into electrical energy. To obtain efficient and economical fuel cells, extensive research efforts have been devoted to the development of highly active and cost-effective electrocatalysts. In this research, a solution-based EPD technique of carbon-nanostructure layer for fabrication of catalyst layers in anode electrodes of direct ethanol fuel cell are developed and investigated. The EPD process is considered to fabricate catalyst layers for an anode electrode of direct ethanol fuel cells without resorting to expensive manufacturing processes and catalyst materials made of precious metals.

The first chapter gives an introduction to the field. The advantages of direct ethanol fuel cells are first given. Then the techniques used in the fabrication of electrodes are reviewed with emphasis on the deposition of catalyst supports. Two strategies are given, namely, co-deposition of the catalyst support together with the catalyst materials and subsequent deposition of the catalyst layer onto which catalyst is deposited. The latter is suitable for noble metal as it uses low amount of materials and is known to deliver large mass activity. The former should be used with non-noble metal as the catalyst material amount is large. Electrophoretic deposition is introduced as a possible technique to obtain catalytic nanoparticles loaded on a nanographitic catalyst support.

Chapter two reports the fabrication technique and optimization of the catalyst support layer. The catalyst support layer is obtained using DC electrophoretic deposition from a colloid consisting of nanographitic flakes dispersed in isopropyl alcohol together with magnesium ions. The magnesium ions control the flake surface charge in the colloid and are used to bind nanographitic flakes together. The packing density and conductivity of

the deposited layer are controlled by the ion concentration in the colloid. The performance of the catalyst support was first evaluated by decorating the fabricated catalyst support layers with palladium nanostructures serving as the catalyst (palladium is thought as a replacement for the standard platinum catalyst in alkaline solution). The fabricated support layer with the highest packing density exhibited improved conductivity and electro-oxidation performance reaching 1.5 A/cm^2 per mg of palladium. This performance is among the best for direct ethanol fuel cells based on palladium.

Chapter three explains the co-deposition of the catalyst support together with non-precious metal catalyst nanoparticles. Using the optimized fabrication conditions for packing density of the catalyst support, the deposition of a non-precious metal catalyst together with its catalyst support in a single-step electrophoretic deposition is demonstrated. The catalyst consists of nickel oxide nanoparticles which are attached on the surface of the nanographitic flakes during the electrophoretic process. The fabricated anode showed a very low onset voltage for the electro-oxidation of ethanol. Furthermore, a better control of the dispersion of the catalytic nanoparticles on the catalyst support is demonstrated using pulsed electrophoretic deposition. Using short pulses help to decrease agglomeration of catalytic nanoparticles on the nanographitic flake support and form well dispersed catalytic particles on the support. As a result of the improved dispersion of the catalytic nanoparticles, a larger electro-oxidation current was achieved.

Chapter four clarifies the mechanism for ethanol electro-oxidation on nanographitic flakes support loaded NiO nanoparticles. To clarify the mechanism, the sample is first modified by annealing (or plasma treatment) to obtain a different electrochemical behavior, then the samples properties such as material characteristics,

kinetics of the electrochemical reactions and conductivity are compared for samples without and with annealing. Temperature dependence measurements indicate a charge transport mechanism based on hopping transport, which is changed to drift transport for the annealed sample. The hopping charge transport is related to the appearance of an ethanol electro-oxidation current peak at low voltage, which is different from the reported literature data.