### - Communication -

# DMFC Catalyst Layer Prepared Using Arc-Soot Nano-Carbon by Dry-Squeegee Method and Its Impedance Analysis

## Shinichiro OKE,<sup>a,\*</sup> Yuuki IzuMI,<sup>a</sup> Takashi IKEDA,<sup>a</sup> Hikaru URUNO,<sup>a</sup> Yoshiyuki SUDA,<sup>a</sup> Hirofumi TAKIKAWA,<sup>a</sup> Sigeo ITOH,<sup>b</sup> Tatsuo YAMAURA,<sup>b</sup> Hitoshi UE,<sup>c</sup> Toshihiro SAKAKIBARA,<sup>d</sup> Shuichi SUGAWARA,<sup>d</sup> Takashi OKAWA,<sup>e</sup> and Nobuyoshi AOYAGI<sup>e</sup>

<sup>a</sup>Toyohashi University of Technology (1-1 Hibarigaoka, Tempaku, Toyohashi, Aichi 441-8580, Japan)

<sup>b</sup>Futaba Corporation (1080 Yabutsuka, Chosei-mura, Chosei-gun, Chiba 299-4395, Japan)

<sup>c</sup>Tokai Carbon Co., Ltd. (394 Subashiri, Oyama-cho, Sunto-gun, Shizuoka 410-1431, Japan)

dENAX Inc. (2-11-19 Otowa, Bunkyo-ku, Tokyo 112-0013, Japan)

eDaiken Chemical Co., Ltd. (1-3-3 Techno Port, Mikuni-cho, Sakai, Osaka 913-0038, Japan)

Received October, 23, 2008 ; Accepted December 17, 2008 ;

Micro-structure of catalyst layer influences performance of direct methanol fuel cell (DMFC). The dry-squeegee method which was a technique proposed for catalyst layer formation, was found to form a dense and uniform catalyst layer. The membrane electrode assembly (MEA) prepared by using the dry-squeegee method (MEA-ds) had a better performance than that prepared by conventional methods including dropper and brush method, since the MEA-ds had a shorter proton path from the catalyst surface to polymer membrane and many active reaction sites than other MEAs.

*Key Words* : Direct Methanol Fuel Cell, Catalyst Layer, Dry-squeegee Method, Electrochemical Impedance Spectroscopy

### 1 Introduction

Direct methanol fuel cells (DMFC) are very promising power sources for portable applications and high efficiency due to simple handling and processing of fuel. However, their performance has not yet been satisfactory. Typically, DMFC electrocatalysts are based on precious metals, and their high cost still remains one of the drawbacks to the widespread use of these energy conversion systems. Carbon-supported Pt-Ru is the catalyst of choice for the DMFC anode, and many research groups has focused on the development of high catalytic activity electrodes for methanol oxidation by pursuing new synthetic routes as well as by developing new catalyst supports.<sup>1,2)</sup> Nano-carbons are believed to be potential materials for an electrode component in a DMFC. The authors have synthesized original nano-carbons, such as carbon nano-coils, carbon nano-twists, arc-soot nano-carbon (AS), and carbon nano-balloon (CNB). AS can support a lot of Pt-Ru catalyst nano-particles, thus, Pt-Ru/AS catalyst was better than Pt-Ru/Ketjen black catalyst for the DMFC.<sup>3)</sup> CNB has a higher conductivity than other nano-carbons.<sup>4)</sup> Through feasibility investigations to date of such nano-carbons to be used as DMFC electrode, it is realized that the application method is important to obtain stable and higher DMFC performance. Moreover, increasing reaction sites in the catalyst layer is important for improving electrode performance.<sup>5,6)</sup> Since the polymer membrane used for the electrolyte is a solid phase, the membrane cannot deeply penetrate into the electrode as does a liquid one, so the reaction area is limited to the contact surface between the electrode and membrane.<sup>7)</sup> When conventional methods are used to form a catalyst layer, it is difficult to produce a perfectly homogeneous catalyst layer in an area with high ionomer density.

In this study, a dry-squeegee method, to form a catalyst layer of a membrane electrode assembly (MEA), was proposed for the improvement of the cell performance of DMFC.

#### 2 Experimental

MEAs were prepared by the proposed dry-squeegee method (MEA-ds) and two conventional methods including a dropper (MEA-dr)<sup>3)</sup> and brush (MEA-br),<sup>8)</sup> respectively. These MEAs contained Pt-Ru/AS and Pt/AS catalysts. Pt-Ru/AS and Pt/AS were used as the anode and cathode catalysts, respectively. The details of AS preparation and catalyst support were described in our earlier paper.<sup>3)</sup> Briefly, AS was prepared using a twin-torch-arc apparatus. Pt-Ru and Pt particles were supported on AS by the colloidic process.<sup>9)</sup> Catalyst slurries were prepared by mixing 40 mg catalysts, 1 ml de-ionized water, 1 ml isopropyl alcohol, and 13 mg carbon nano-balloon. These conventional methods used catalyst slurries for catalyst layer formation. For use with the proposed drysqueegee method, catalyst slurries were filtered though a filter paper, and dried under vacuum at 120 °C for 2 h. Dried catalyst powders were put on carbon papers (SGL carbon, GDL31BA) in an enclosed area with mending tapes, and smoothed by a glass rod. The 58-µm thickness of the catalyst layer was the same as that of the mending tape. Before hot pressing, a 5 wt.% Nafion solution (6 ml) was sprayed onto the catalyst side of these electrodes prepared by dropper, brash, and dry-squeegee