Novel Catalysts (Electrochemical Deposition of Pt Nano Particles on Carbon Nanotubes)

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Introduction
Carbon nanotubes (CNTs) have received increasing interest due to their peculiar properties, such as electronic conductivity, mechanical strength and large surface area. CNTs have been used for conductive and high strength composites, energy conversion devices, field emission displays, radiation sources, batteries, and sensors. Moreover, CNTs have the unique surface structure which prevents the metal particles from agglomerating. However, the idea of using CNTs as catalyst support has rarely been attempted.

The objective of this study is to synthesize Pt nanoparticles using CNTs as support by electrochemical deposition (ECD). ECD has recently been adopted as the catalyst preparation method for their advantages, such as high purity of deposits, simple process and easy control of the loading mass [1].

Materials and Methods
CNTs used in this study were grown directly on graphite paper (2.5 cm x 2.5 cm) by methane decomposition. Ni was used as the catalyst for the growth of CNTs and doped on graphite paper by ECD method. A three electrode cell was used and the deposition was performed at 0.15–0.65 V at a pulse width of 10 ms in 0.5 mol Ni(NO$_3$)$_2$·6H$_2$O (99.999%, Aldrich) + 0.5 mol H$_2$SO$_4$ aqueous solution. Ni/CNTs was grown by methane decomposition at 700°C for 2 h. Pt-ECD was then performed on CNTs/graphite paper at the deposition potential of -0.25 V (SCE) in 0.25 mol H$_2$PtCl$_6$ (99.9%, Spectrum) + 0.5 mol H$_2$SO$_4$ aqueous solution by potentiostatic method (Amel 7060 potentiostat). Pt/CNTs was also prepared by wet impregnation for the comparison of cataytic activity. In this process, H$_2$PtCl$_6$·6H$_2$O (99.9%, Spectrum) dissolved in distilled water was used as Pt-precursor and the catalyst was dried at 110°C for 12 h and calcined in air at 450°C for 4 h.

Pt content of each catalyst sample was measured by inductively coupled plasma-optical emission spectrometer (ICP-OES, Perkin-Elmer). The surface of each catalyst sample was observed by SEM (Hitachi, S-4700) and HR-TEM (JEOL JEM). Mass activity of each catalyst was determined at 0.9 V using a reversible hydrogen electrode (RHE) as a reference. The amounts of CO chemisorbed selectively on Pt catalytic sites were measured by Autochem 2910 (Micromeritics).

Results and Discussion
The micrograph of the Pt/CNTs/graphite paper synthesized by ECD method was obtained by SEM and the corresponding images are presented in Fig. 1. Fig. 1(a) shows highly dispersed Ni particles, which are used as the catalyst for CNTs growth, on graphite paper. Fig. 1(b) shows multi-walled CNTs grown on graphite paper by methane decomposition. Pt nano particles were then electrochemically deposited on to them. As shown in Fig. 1(c), Pt particles are highly dispersed on the surface of CNTs and ranges about 2-5 nm. TEM images were also obtained for the comparison of the Pt particles prepared by ECD method and those synthesized by wet-impregnation (Fig. 2). The image of Pt/CNTs synthesized by wet-impregnation is presented in fig. 1(a) which indicates that the metal particles are randomly dispersed and range about 2-10 nm. In the case of electrochemically deposited Pt/CNTs (fig. 2(b)), however, the metal dispersion of the Pt particles are better and the size of which is also much smaller (2-5 nm) than the case of fig. 2(a).

The mass activity and the amount of chemisorbed CO are 4.5-5 times larger in the case of Pt-ECD compared to the case of wet-impregnation. These results are attributed to the small particle size and high dispersion of Pt particles. Moreover, the direct growth of CNTs on graphite paper is effective for the increase of catalytic sites compared to the present ‘pasting method’. Consequently, Pt-ECD on the CNTs directly grown on the graphite paper is recommended as the preparation method of fuel cell electrode.

Significance
Electrochemical deposition (ECD) is very useful technique for the direct growth of CNTs on graphite paper and also for the preparation of highly dispersed and nano-sized Pt catalyst. This method is effective for the preparation of fuel cell electrode.

References