

Rationalize Synthesis of Pt-Ru/MWCNTs Bimetallic catalysts for Methanol Oxidation Reaction

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Introduction

Direct methanol fuel cells (DMFCs) have been attracting great attention due to their significant advantages over hydrogen-fed proton exchange membrane (PEM) fuel cells in mobile applications in terms of fuel storage and supply. However, the commercialization of DMFCs is still hindered by some technical challenges, mainly slow kinetics of the methanol oxidation reaction. Bimetallic Pt-Ru alloy catalysts have been demonstrated to be more active than pure Pt catalysts in the electrooxidation of methanol. But, there is still room for further improvement of the catalytic activity. In this study, Pt-Ru nanoparticle catalysts supported on different functionalized carbon nanotubes have been studied for methanol oxidation reaction. As carbon nanotubes (CNTs), have been widely employed as support in heterogeneous catalysis, owing to the high electrical conductivity and corrosion resistance. The catalysts were prepared by combination of strong electrostatic adsorption (SEA) and electroless deposition (ED) methods. Our Previous studies have shown that combining SEA and ED methods can increase the surface area of the Pt particles by lowering the particle sizes of the bimetallic particles [1,2]. The synthesized catalysts then characterized by X-ray diffraction (XRD) spectroscopy, HRTEM images, chemisorption and cyclic voltammetry towards methanol electrooxidation reaction.

Materials and Methods

All cyclic voltammetry experiments were conducted in 1M methanol and 0.5M H₂SO₄ solutions sparged with N₂ flow. A potentiostat (263A, Princeton Applied Research with CorrView/CorrWare (Scribner Associated Inc.)) was used to take all data. Chemisorption using hydrogen pulse titration of oxygen-precovered Pt was performed using a Micromeritics Autochem II 2920 automated chemisorption analyzer to determine the concentration of surface Pt sites. X-ray diffraction (XRD) patterns were recorded on a Rigaku Miniflex-II equipped with a D/teX Ultra silicon strip detector using Cu K α radiation.

Results and Discussion

The combination of SEA and ED methods of catalyst preparation, where SEA is used for synthesis of small, uniformly-distributed Pt nanoparticles followed by ED of controlled amounts of Ru on the Pt surface, has been used to prepare extremely active and well-defined Ru-Pt bimetallic particles on functionalized carbon nanotubes. The activities of all ED-derived Ru-Pt catalysts on MWNTs were much higher than the commercial Pt/XC72 and Pt-Ru/XC72. Figure 1 shows the x-ray diffraction patterns for the different catalysts, and the electrochemical surface area (ECSA) of all catalysts are shown in Table 1.

Significance

Bimetallic Ru-Pt/MWNTs catalysts with higher activity for methanol oxidation reaction were prepared using SEA and ED methods. Particles of higher dispersion, compared to commercial catalysts, were obtained with SEA/ED while targeted deposition of Ru using ED resulted in strong interaction with Pt.

Table 1. ESCA of Pt/XC72 and PT/MWNTs samples made by SEA and the commercial 20 wt% Pt/XC72.

Samples	ECSA (m ² /g _{Pt})
6.5 wt% Pt/MWNT-OH (SEA)	61 ±2
7.3 wt% Pt/MWNT-COOH (SEA)	73±7
6.3 wt% Pt/XC72 (SEA)	74±5
20 wt% Pt/XC72 (Commercial)	59±4

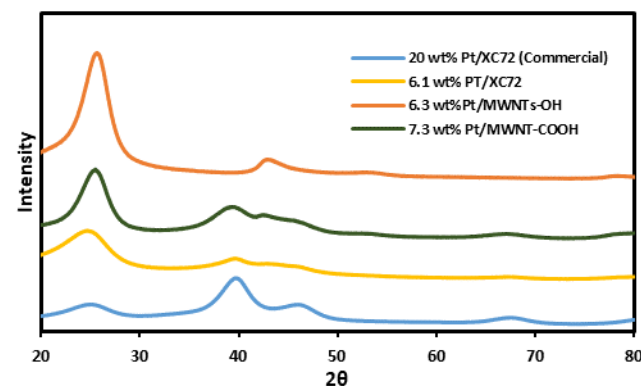


Figure1. Powder X-ray diffraction profiles of Pt/XC72 and PT/MWNTs samples made by SEA and the commercial 20 wt% Pt/XC72.

References

1. Tengco, J.M.M., Tavakoli Mehrabadi, B.A, Monnier, J., Weidner, J.W., Regalbuto, J. R., Catalysts, 6 6 83 (2016).
2. Tavakoli Mehrabadi, B.A, White, R., Shakouri, A., Weidner, J.W., Regalbuto, J. R., Monnier, J. R., Catal Today, In press (2018).