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Abstract
Pt — Pd alloy, Pt, and Pd nanoparticles were synthesized in reverse micellar solutions, employing H2PtCl6/Pd(NH3) 4Cl2 as the precursors, N2H5OH as the reductant, water as the dispersion phase, sodium di-2-ethylhexyl sulfosuccinate (AOT) as the surfactant, and isooctane as the continuous phase. The size of the resultant nanoparticles, which depends on the molar ratio of water to AOT, ω 0, was about 10 nm at ω0 = 9 from TEM images. It was found that the composition of the Pt — Pd alloy nanoparticles had no effect on average particle size.

The nanostuctured electrodes were prepared by electrophoretic deposition of the nanoparticles on Ti supports under a potential field of 1000 V/cm, followed by a heat treatment in air. In this study, we prepared nanosturctured Pt — Pd alloy electrodes, nanostructured Pt + Pd co-deposition electrodes, and nanostructued Pt electrodes; then compared their electrochemical activities towards methanol oxidation in 1.0 M NaOH aqueous solution.

It could be confirmed that the nanoparticles were deposited on the Ti support from FE—SEM images. EDS analysis revealed that the composition on the surface of the nanostructured Pt — Pd alloy electrode was almost equal to that of the preparation solution of the Pt — Pd alloy nanoparticles, indicating that the Pt — Pd nanoparticles had a homogeneous alloy phase.

The electrochemical activity of the nanostructured Pt — Pd alloy electrodes towards methanol oxidation in 1.0 M NaOH + 0.1 M CH3OH aqueous solution was investigated by cyclic voltammetry, CV. The results showed that the current density of the reaction decreased with increasing heat treatment temperature. Although decreasing heat treatment temperature increases the current density, lower temperature would weaken adhesion force between the nanoparticles and the Ti support. In this study, it was found that the optimum heat treatment temperature was 400℃. Furthermore, with increasing the Pd content of the electrode, the current density for methanol oxidation increased first, and then decreased after reaching a maximum at 80 mole % Pd. Also, the onset potential for methanol oxidation decreased with increasing Pd content; then increased after reaching a minimum value of 0.381 V at 80 % Pd.

The potential of the nanosturctured Pt — Pd alloy electrodes was held at 0.825 V; then the dependence of the current density for methanol oxidation on reaction time was examined, an approach known as j — t method. It was found that the initial current density, j value at t = 0.05 sec, increased with increasing the Pd content of the electrode and decreased after reaching a maximum value of 68.7 mA/cm2 at 80 mole % Pd. These results were similar to those obtained by CV method.

The electrochemical activity of the nanostructured Pt + Pd co-deposition electrodes towards methanol oxidation was investigated by both CV and j — t methods. The results showed that the Pd content of the electrode affected the activity of the electrode in the same way as that of the

nanostuctured Pt — Pd alloy electrode. However, the maximum activity of the nanostructured Pt + Pd co-deposition electrode occurred at 40 mole % Pd.

Compared with the other electrodes, the nanostructured Pt — Pd alloy electrode with 80 mole % Pd has the highest activity for methanol oxidation. The series of active electrodes are as follows: nanostructured Pt — Pd alloy electrode > nanostructured Pt + Pd co-deposition electrode > nanostructured Pt electrode > bulk Pt electrode. In the CV method, the ratios of the activity of the nanostructured Pt — Pd alloy electrode with 80 mole % Pd to that of the other electrodes were 1.6 to the nanostructured Pt + Pd co-deposition electrode with 40 mole % Pd, 3 to the nanostructured Pt electrode, and 13.7 to the bulk Pt electrode, based on the peak (P1) current density. Also, in the j — t method, the corresponding ratios were 1.65, 3.3, and 15, based on the initial current density.

The electron-transfer limiting current density, je, for methanol oxidation was obtained by the rotating-disc electrode method. The rate constant of methanol oxidation at a given temperature was obtained by plotting je against methanol concentration. From the rate constants obtained at different temperatures, the activation energy for methanol oxidation catalyzed by electrodes was calculated. The rate constants obtained at 25℃ for the nanostructured Pt — Pd alloy electrode with 80 mole % Pd and for the bulk Pt electrode were 1.38×10 -6 s -1 and 1.41×10-8 s -1, respectively. Then the activitation energies calculated for the corresponding electrodes were 22.8 kJ/mole and 56.9 kJ/mole, respectively.

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