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Effect of Annealing Atomic Rearrangement on Electrochemical Performance of Pd-Ni Catalyst

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The rearrangement of the Pd and Ni atoms in the Pd-Ni nanoparticles is achieved by annealing under a vacuum condition with an absolute pressure of 0.03 MPa, to successfully synthesize carbon supported composite nanoparticles with Pd core and NiO_xH_y doped PdO shell (Pd@PdO-NiO_xH_y/C). This unique structure with a Pd core and a PdO metal oxide shell doped by NiO_xH_y is proved by the X-ray diffraction analysis and energy dispersive spectrometer mapping images. The shell structure of the Pd@PdO-NiO_xH_y nanoparticles is further proved by the X-ray photoelectron spectroscopy. The electrochemical test results indicate that the core-shell catalyst prepared at 400 °C (Pd@PdO-NiO_xH_y/C-400) catalyst has the best catalytic activities and stabilities in all the catalysts for the methanol electro-oxidation (MEO) and ethanol electro-oxidation (EEO). The peak value of the mass specific current density on the Pd@PdO-NiO_xH_y/C-400 catalyst for the MEO reaches to 1118.9 mA mg^{-1} Pd, which is 2.5 and 1.4 times those of the Pd/C (443.1 mA mg^{-1} Pd) and Pd-Ni/C (828.1 mA mg^{-1} Pd) catalysts, respectively. This improvement is attributed to the unique doped structure between NiO_xH_y and PdO in the shell layer of the Pd-based nanoparticles in the core-shell catalyst, the promoting effect of the NiOOH, as well as the bifunctional mechanism. Under the influence of this doped structure, the new Pd nanoparticles generated by the rearrangement of the Pd atoms in the electrochemical activation process, are divided by the NiO_xH_y species, resulting in the new formed Pd nanoparticles have ultra-small size and high stability. These are also the reason that the Pd@PdO-NiO_xH_y/C-400 catalyst reveals good durability for both the MEO and EEO. Moreover, the ratio between its stable current densities for the EEO (86.35 mA mg^{-1} Pd) and MEO (12.57 mA mg^{-1} Pd) after 3600 s at -0.1 V vs. Ag|AgCl is 6.9, showing that the Pd@PdO-NiO_xH_y/C catalyst has broad application prospects in the field of the direct ethanol fuel cells.

Keywords: Direct Alcohol Fuel Cells; Methanol Electro-oxidation; Ethanol Electro-oxidation; Core-shell Structure; Pd based nanocatalyst; NiO_xH_y doped PdO.

FULL TEXT