

The Hydrogen Evolution Reaction Performance of Bimetallic Pt_xPd_y Nanoclusters

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Abstract

Pt and Pt-based nanoparticles (NPs) are widely used as the electrocatalysts for the hydrogen evolution reaction (HER). However, the application of the Pt-based catalysts has the limitation due to its high cost and low utilization of Pt. Therefore, downsizing the catalysts is the key factor for the promotion of the HER performance. In order to solve those problem, in this study, carbon-supported Pt-nanoclusters (NCs) and Pt_xPd_y-NCs (x/y= 1/2, 1/1, and 2/1) are prepared by deposition-precipitation method. In the X-ray absorption near edge spectra (XANES) of the catalysts at Pt L₃ edge, the intensity (H_A) of absorption peak represents the extent of electron transition from 2p_{3/2} to 5d_{5/2} orbitals and the width of 5d band. The oxidation state of Pt atoms is increased with H_A following the trend of PtO₂ > PtPd₂-NCs > Pt₂Pd-NCs > PtPd-NCs > Pt-NPs. Besides, in the X-ray absorption near edge spectra (XANES) spectrum at Pd K edge, the H_A of the PtPd-NCs is lower than those of Pt₂Pd-NCs and PtPd₂-NCs, and it is close to Pd-NPs, implying that the Pt shell may form on the Pd core to prevent Pd from oxidation. In the Fourier transformed extended X-ray absorption fine structure spectra (FT-EXAFS) of Pt_xPd_y-NCs at Pt L₃ edge, it can be observed that the formation of the Pt-Pd bond after the Pd addition. When compared to the FT-EXAFS at Pd K edge, it can be seen that the intensity of the Pd-O follows the trend of Pt₂Pd-NCs > PtPd₂-NCs > PtPd-NCs, saying that the Pt shell structure of the Pt_xPd_y-NCs depends on the Pt/Pd ratio. According, the atomic models of the PtPd-NCs have more complete Pd core/ Pt shell structure, so the Pd core can retain its metallic state without severe oxidation. Moreover, the HER performance results indicate that the PtPd-NCs exhibit the excellent HER performance with the MA_{0.05} and MA_{0.05} decay of 9.57 A/mg_{Pt} and 19 %, respectively, much higher than Pt-NCs (with the MA_{0.05} and MA_{0.05} decay of 1.3 A/mg_{Pt} and 39 %), showing that the Pd core/Pt shell structure can improve the HER performance and stability.

Keywords - hydrogen evolution reaction (HER), PtPd catalysts, nanoclusters, X-ray absorption.