Abstract

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The thesis entitled “Nanostructured Carbon Supported Transition Metal-based Bifunctional Electrocatalysts for Rechargeable Zn-air Battery” describes the design and synthesis of bifunctional electrocatalysts based on transition metal, metal oxide, alloy, and nitrogen doped carbon nanostructures for the development of rechargeable Zn-air battery (ZAB). Electrocatalysts such as reduced graphene oxide (rGO) supported CoFe2O4 (rGO/CoFe2O4), nitrogen and cobalt dual doped mesoporous carbon (Co-N-C), and nitrogen-doped graphitic/onion-like/tubular carbon supported CoFe and NiCo alloy have been synthesized using different catalyst precursors. A supramolecular approach is developed for the synthesis of tubular carbon support. The electrocatalytic activity of these catalysts towards oxygen reduction (ORR) and oxygen evolution (OER) reactions has been evaluated by hydrodynamic voltammetry. The rGO/CoFe2O4 hybrid efficiently catalyzes ORR and is not active towards OER. On the other hand, all the other catalysts are bifunctionally active towards ORR and OER. The effect of catalyst support on the bifunctional catalytic activity is demonstrated with the CoFe alloy catalyst. The encapsulation of CoFe with nitrogen doped graphitic carbon shell imparts high stability during catalysis and the high content of graphitic carbon and nitrogen largely control the overall performance of the catalyst. It is established that post-synthetic structural engineering of as-synthesized NiCo and CoFe catalyst by acid treatment significantly improves overall electrocatalytic performance. Excellent bifunctional activity with potential gap of 0.74□ 0.80 V is achieved with NiCo and CoFe-based catalysts. All the bifunctionally active catalysts have been utilized for the development of rechargeable ZAB. The performance of these catalysts as air-cathode has been evaluated in terms of open circuit voltage, power density, and voltaic efficiency. Open circuit voltage as high as 1.58 V has been achieved with nitrogen doped onionlike carbon supported CoFe catalyst. Among the all bifunctional catalysts, the NiCo-based catalyst delivers the highest power density of 148.8 mW/cm2 and has excellent charge-discharge cycling stability over 48 h with $< 5\%$ loss of voltaic efficiency.

Keywords: Oxygen electrocatalysis; bifunctional electrocatalyst; carbon nanostructure; transition metal alloy; Zn-air battery