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Carbon Nanofibers as the Favorite Conducting Additive for Mn_3O_4 Catalysts for Oxygen Reactions in Rechargeable Zinc-Air Battery

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Abstract : Rechargeable zinc-air batteries (RZABs) have been described as one of the most viable next-generation 'beyond-the-lithium-ion' battery technologies with great potential for renewable energy storage. It is safe, with a high specific energy density (1086 Wh/kg), environmentally benign, and low-cost, especially in resource-limited African countries. For widespread commercialization, the sluggish oxygen reaction kinetics pose a major challenge that impedes the reversibility of the system. Hence, there is a need for low-cost and highly active bifunctional electrocatalysts. Manganese oxide catalysts on carbon conducting additives remain the best couple for the realization of such low-cost RZABs. In this work, hausmannite Mn_3O_4 nanoparticles were synthesized through the annealing method from commercial electrolytic manganese dioxide (EMD), multi-walled carbon nanotubes (MWCNTs) were synthesized via the chemical vapor deposition (CVD) method and carbon nanofibers (CNFs) were synthesized via the electrospinning process with subsequent carbonization. Both Mn_3O_4 catalysts and the carbon conducting additives (MWCNT and CNF) were thoroughly characterized using X-ray powder diffraction spectroscopy (XRD), scanning electron microscopy (SEM), thermogravimetry analysis (TGA) and X-ray photoelectron spectroscopy (XPS). Composite electrocatalysts (Mn_3O_4/CNT and Mn_3O_4/CNF) were investigated for oxygen evolution reaction (OER) and oxygen reduction reaction (ORR) in an alkaline medium. Using the established electrocatalytic modalities for evaluating the electrocatalytic performance of materials (including double layer, electrochemical active surface area, roughness factor, specific current density, and catalytic stability), CNFs proved to be the most efficient conducting additive material for the Mn_3O_4 catalyst. From the DFT calculations, the higher performance of the CNFs over the MWCNTs is related to the ability of the CNFs to allow for a more favorable distribution of the d-electrons of the manganese (Mn) and enhanced synergistic effect with Mn_3O_4 for weaker adsorption energies of the oxygen intermediates (O*, OH* and OOH*). In a proof-of-concept, Mn_3O_4/CNF was investigated as the air cathode for rechargeable zinc-air battery (RZAB) in a micro-3D-printed cell configuration. The RZAB showed good performance in terms of open circuit voltage (1.77 V), maximum power density (177.5 mW cm-2), areal-discharge energy and cycling stability comparable to Pt/C (20 wt%) + IrO2. The findings here provide fresh physicochemical perspectives on the future design and utility of CNFs for developing manganese-based RZABs.

Keywords : bifunctional electrocatalyst, oxygen evolution reaction, oxygen reduction reactions, rechargeable zinc-air batteries.
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