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Co_2Fe LDH on Aromatic Acid Functionalized N Doped Graphene: Hybrid Electrocatalyst for Oxygen Evolution Reaction

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Abstract : Designing highly active and low-cost oxygen evolution ($2H_2O \rightarrow 4H^+ + 4e^- + O_2$) electrocatalyst is one of the most active areas of advanced energy research. Some precious metal-based electrocatalysts, such as IrO_2 and RuO_2, have shown excellent performance for oxygen evolution reaction (OER); however, they suffer from high-cost and low abundance which limits their applications. Recently, layered double hydroxides (LDHs), composed of layers of divalent and trivalent transition metal cations coordinated to hydroxide anions, have gathered attention as an alternative OER catalyst. However, LDHs are insulators and coupled with carbon materials for the electrocatalytic applications. Graphene covalently doped with nitrogen has been demonstrated to be an excellent electrocatalyst for energy conversion technologies such as; oxygen reduction reaction (ORR), oxygen evolution reaction (OER) & hydrogen evolution reaction (HER). However, they operate at high overpotentials, significantly above the thermodynamic standard potentials. Recently, we reported remarkably enhanced catalytic activity of benzoate or 1-pyrenebutyrate functionalized N-doped graphene towards the ORR in alkaline medium. The molecular and heteroatom co-doping on graphene is expected to tune the electronic structure of graphene. Therefore, an innovative catalyst architecture, in which LDHs are anchored on aromatic acid functionalized 'N' doped graphene may presumably boost the OER activity to a new benchmark. Herein, we report fabrication of Co_2Fe-LDH on aromatic acid (AA) functionalized 'N' doped reduced graphene oxide (NG) and studied their OER activities in alkaline medium. In the first step, a novel polyol method is applied for synthesis of AA functionalized NG, which is well dispersed in aqueous medium. In the second step, Co_2Fe LDH were grown on AA functionalized NG by co-precipitation method. The hybrid samples are abbreviated as Co_2Fe LDH/AA-NG, where AA is either Benzoic acid or 1, 3-Benzene dicarboxylic acid (BDA) or 1, 3, 5 Benzene tricarboxylic acid (BTA). The crystal structure and morphology of the samples were characterized by X-ray diffraction (XRD), scanning electron microscope (SEM) and transmission electron microscope (TEM). These studies confirmed the growth of layered single phase LDH. The electrocatalytic OER activity of these hybrid materials was investigated by rotating disc electrode (RDE) technique on a glassy carbon electrode. The linear sweep voltammetry (LSV) on these catalyst samples were taken at 1600rpm. We observed significant OER performance enhancement in terms of onset potential and current density on Co_2Fe LDH/BTA-NG hybrid, indicating the synergic effect. This exploration of molecular functionalization effect in doped graphene and LDH system may provide an excellent platform for innovative design of OER catalysts.

Keywords : π-π functionalization, layered double hydroxide, oxygen evolution reaction, reduced graphene oxide
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