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Nickel Oxide-Nitrogen-Doped Carbon (Ni/NiOx/NC) Derived from Pyrolysis of 2-Aminoterephthalic Acid for Electrocatalytic Oxidation of Ammonia

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Abstract : Nitrogenous compounds, such as NH4+/NH3 and NO3-, have become important contaminants in water resources. Excessive concentration of NH3 leads to eutrophication, which poses a threat to aquatic organisms in the environment. Electrochemical oxidation emerged as a promising water treatment technology, offering advantages such as simplicity, small-scale operation, and minimal reliance on additional chemicals. In this study, a nickel-based metal-organic framework (Ni-MOF) was synthesized using 2-amino terephthalic acid (BDC-NH2) and nickel nitrate. The Ni-MOF was further carbonized as derived nickel oxide and nitrogen-carbon composite, Ni/NiOx/NC. The nickel oxide within the 2D porous carbon texture served as active sites for ammonia oxidation. Results of characterization showed that the Ni-MOF was a hexagonal and flaky nanoparticle. With increasing carbonization temperature, the nickel ions in the organic framework re-crystallized as NiO clusters on the surfaces of the 2D carbon. The electrochemical surface area of Ni/NiOx/NC significantly increased as to improve the efficiency of ammonia oxidation. The phase transition of Ni(OH)2⇌NiOOH at around +0.8 V was the primary mediator of electron transfer. Batch electrolysis was conducted under constant current and constant potential modes. The electrolysis parameters included pyrolysis temperatures, pH, current density, initial feed concentration, and electrode potential. The constant current batch experiments indicated that via carbonization at 800 °C, Ni/NiOx/NC(800) was able to decrease the ammonium nitrogen of 50 mg-N/L to below 1 ppm within 4 hours at a current density of 3 mA/cm2 and pH 11 with negligible oxygenated nitrogen formation. The constant potential experiments confirmed that N2 nitrogen selectivity was enhanced up to 90% at +0.8 V.

Keywords : electrochemical oxidation, nickel oxyhydroxide, metal-organic framework, ammonium, nitrate
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