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Holey Co, N-codoped graphene aerogel with in-plane pores and multiple active sites for efficient oxygen reduction

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Abstract

Non-noble metal electrocatalysts have emerged as promising next-generation electrocatalysts for oxygen reduction reaction (ORR). Despite considerable progress, non-noble metal electrocatalysts are plagued by relatively low efficiency and poor long-term stability. This study reports the design and synthesis of holey Co, N-codoped graphene aerogel with in-plane pores and multiple active sites (CoN-HGA) for efficient four-electron ORR in both alkaline and acid mediums. In particular, we use etched graphene oxide (GO) sheets with abundant in-plane pores to fabricate the aerogel and a porphyrin containing both Co and N elements as the doping precursor. The as-prepared CoN-HGA, thus, not only inherits abundant defective active sites from the etched GO sheets, but also has plenty of Co- and N-doped active sties. Furthermore, its holey structure creates abundant graphene sheet-sheet contacts as electron pathways, thereby greatly diminishing the resistance of the electrocatalyst. Meanwhile, the in-plane pores also serve as efficient mass transport channels, allowing full access to the internal active sites. Compared with commercial Pt/C (20 wt%), CoN-HGA exhibits slightly higher catalytic activity in alkaline medium and comparable activity in acidic medium with much improved stability and methanol tolerance. The synergetic effect from multiple active sites and unique structure provides a new route to design low-cost, high efficient ORR catalyst and beyond.

Keywords: Graphene, non-noble metal electrocatalyst, oxygen reduction reaction, doping

Introduction

Oxygen reduction reaction (ORR) is a key half reaction in many electrochemical energy conversion devices, such as proton exchange membrane fuel cell, metal-air battery, and so on [1,2]. Because of its sluggish nature, ORR requires effective catalysts. Currently, Pt and its alloys are the most widely used ORR catalysts [3-6]. However, Pt-based catalysts are not sustainable for large-scale applications because of their scarcity, high cost, and weak duration [7]. To address this problem, it is highly desirable to develop effective, alternative