

Author's Accepted Manuscript

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PII: S2211-2855(16)30620-6
DOI: <http://dx.doi.org/10.1016/j.nanoen.2016.12.056>
Reference: NANOEN1707

To appear in: *Nano Energy*

Received date: 16 November 2016
Revised date: 28 December 2016
Accepted date: 29 December 2016

Cite this article as: Xiaoqi Chen, Liang Yu, Suheng Wang, Dehui Deng and Xinhe Bao, Highly active and stable single iron site confined in graphene nanosheets for oxygen reduction reaction, *Nano Energy*, <http://dx.doi.org/10.1016/j.nanoen.2016.12.056>

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Highly active and stable single iron site confined in graphene nanosheets for oxygen reduction reaction

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Abstract

Exploring high performance non-precious metal catalysts to substitute Pt for oxygen reduction reaction (ORR) has stimulated wide research interest recently, but it remains a great challenge. Herein, we report a single iron site confined in graphene catalyst via 4N atoms, forming flat FeN₄ structure in the matrix of graphene. The optimized catalyst shows a high ORR activity, almost coming up to the activity of commercial 40% Pt/C catalyst, but a significantly higher stability and tolerance to SO_x, NO_x and methanol with respect to 40% Pt/C. This well-defined structure provides an ideal model to study the catalytic origin of iron-based catalysts. DFT calculations indicate that the high ORR activity origins from highly dispersed and high-density coordinatively unsaturated Fe centres, and the excellent stability origins from the unique confinement of the graphene matrix via 4N atoms. This reaction can proceed easily to H₂O via a four electron transfer path way on the single iron site, which is further confirmed by the experiment. This experimental and theoretical study provides a further insight into the nature of the Fe/N/C catalyst and also introduces a reference for designing high efficient catalysts in electrocatalysis.

Keywords

oxygen reduction reaction; non-precious metals; graphene; single iron site; DFT calculations

Introduction

Oxygen reduction reaction (ORR) is a key process in the cathode of

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