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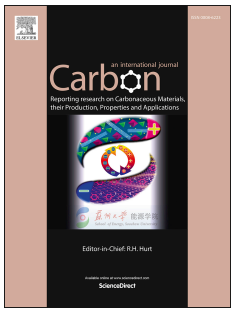

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Surface Activation of Graphene Nanoribbons for Oxygen Reduction Reaction by Nitrogen doping and Defect Engineering: An *ab initio* study

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Abstract

Introducing heteroatoms and creating structural defects on graphene is a common and rather successful strategy to transform its inert basal plane into an efficient metal-free electrocatalyst for oxygen reduction reaction (ORR). However, the intricate atomic configuration of defective graphenes difficult their optimization as ORR electrocatalysts, where not only a large density of active sites is desirable, but also excellent electrical conductivity is required. Therefore, we used density functional theory to investigate the current-voltage characteristics and the catalytic active sites towards ORR of nitrogen-doped and defective graphene by using 8 zig-zag graphene nanoribbons as model systems. Detailed ORR catalytic activity maps are created for ten different systems showing the distribution of catalytic hot spots generated by each defect. Subsequently, the use of both current-voltage characteristics and catalytic activity maps allow to exclude inefficient systems that exhibit either low electrical conductivity or have adsorption energies far from optimal. Our study highlights the importance of considering not only the interaction energy of reaction intermediates to design electrocatalysts, but also the electrical conductivity of such configurations. We believe that this work is important for future experimental studies by providing insights on the use of graphene as a catalyst towards the ORR reaction.

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1. Introduction

The outstanding and versatile properties of graphene nanoribbons make them an interesting class of carbon materials for various applications. Their electronic properties strongly depend on their shape, width, length, and edge morphology.[1-5] Nowadays, it has been proved that all these variables can be fully engineered,[6-7] although controlling them is still challenging. Following the fabrication of a single graphene layer by the Novoselov's method,[8] mass