Exploring the activity of the zigzag graphene nanoribbon edges -and of the graphene quantum dots for the electrochemical Oxygen Reduction Reaction using DFT

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The present study was carried out in the context of the ongoing research aimed at discovering more effective catalysts for the oxygen reduction reaction (ORR) in PEM fuel cells, as the current best-performing ones (Pt -based materials) that both are scarce and expensive. Nanocarbon materials under various forms, undoped or doped, are heavily researched as electrocatalysts for replacing Pt-based catalysts [1]. Nitrogen doped graphene quantum dots (GQD) are gaining attention for this application, due to their abundance in edge sites, low dimensionality, and the fact that ORR is more easily electrochemically catalyzed at the edge than on the basal plane [2]. The primary emphasis in this study is to investigate how dopants and various defects, as well as the reduction in the size of the material affects the activity of the edges towards ORR. To achieve this goal, Density Functional Theory (DFT) was employed to explore the activity of the zig-zag nanoribbon edges, undoped, N-doped or with various functionalized defects – see the activity plot in Figure 1a [3]. Furthermore, as smaller sizes means higher activity in many cases, the study also evaluates the activity of undoped and N doped GQD of various shapes (triangular, rhombohedral and hexagonal) and sizes, for zig-zag or armchair terminated edges – see Figure 1b. The structures placed close to the top of volcanoes show the most promising activity.

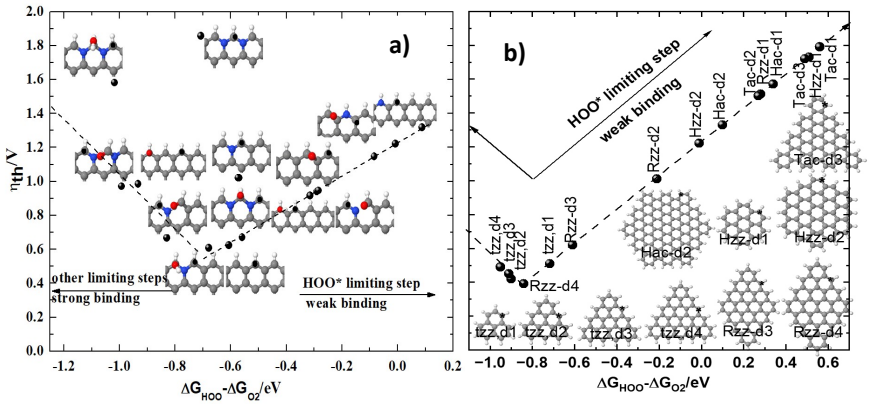

Figure 1 Theoretical calculated overpotential for a) the edges of graphene nanoribbon undoped and N dopped b) the edges of GQD of various sizes and shapes

References

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