Dimensionality-Dependent Oxygen Reduction Activity on Doped Graphene: Is Graphene a Promising Substrate for Electrocatalysis?

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Herein, to understand the low ORR kinetics on the graphene catalysts, the dimensionality of graphene catalysts is sequentially tuned from sheets (2D) to ribbons (1D) and to dots (0D), and then the accompanying changes in terms of physical and electrochemical properties are investigated. In ultraviolet photoelectron spectroscopy, an increment in electropotential is measured as the dimensionality of the graphene catalysts decreases, of which the result infers the enhanced kinetics of the electron transfer from the graphene catalysts to O_2. However, ORR performance does not follow the order of electropotential, and the graphene ribbons show the best activity among the prepared graphene catalysts. Further electrochemical impedance spectroscopy studies demonstrate that ORR kinetics is primarily determined by charge transfer rates in the fabricated graphene electrodes, which are strongly related to the electrode configurations and thus also to the length-to-width ratios of the graphene catalysts. This study suggests the importance of void channels in the fabricated graphene electrode, which have not previously been considered significantly as a factor for improving ORR activity on the graphene catalysts.