

Multiple Heteroatom-Doped Reduced Graphene Oxide as Efficient Oxygen Reduction Reaction Electro Catalysts: The Importance of Doping Sequence

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Opinion

The development of an effective electrocatalyst for the oxygen reduction reaction using multiple heteroatom-doped graphene is of great interest. The competitive doping mechanism produced by the varied atomic sizes of dopants should be developed to maximise the electrocatalytic performance of doped graphene. Using both single-step and two-step procedures, three distinct heteroatoms (e.g., N, P, and B) are competitively incorporated into Reduced Graphene Oxide (RGO). The total number of heteroatoms in ternary RGO synthesised in two steps is lower than in ternary RGO synthesised in one step. Higher ORR electrocatalytic activity for the two-step-synthesized RGO can be explained by: (a): A high amount of P atoms; (b): The fact that B doping itself decreases the less electrocatalytic N moieties such as pyrrole and pyridine and increases the high electrocatalytic moieties such as quaternary N; (c): A high amount of B atoms within the RGO act as a catalyst; It adds to our understanding of how to build heteroatom-doped carbon compounds with high electrocatalytic performance. In recent years, finding effective catalysts for the cathodic Oxygen Reduction Process (ORR) in fuel cells, photocatalytic water splitting, and metal-air batteries has been a major research focus. Noble metals have traditionally been thought to be the most effective ORR catalysts, but they have a number of disadvantages, including expensive cost, poor long-term durability, sluggish electron transfer kinetics, and carbon monoxide poisoning susceptibility. As a result, substantial research has been conducted into the replacement of noble metal-based catalysts with effective and economical non-metal catalysts. Due to their huge surface area, excellent electrical conductivity, and chemical stability, two-dimensional graphenes have been investigated as efficient catalysts among several forms of carbon materials. Due to the low number of active sites, pure graphene with no bandgap had poor electrocatalytic activity. As a result, it is highly recommended to incorporate heteroatom elements (e.g., Nitrogen (N), Boron (B), Phosphor (P), and Sulphur (S)) into graphene to generate electrocatalytic active sites within the graphene and improve its electrocatalytic activity, as heteroatom doping introduces defects, increases both interlayer spacing and electrical conductivity of graphene, and even improves its electrocatalytic activity. When compared to single-atom-doped graphene, dual-doped graphene with two types of heteroatoms (e.g., BN, NS, and NP) showed improved electro catalytic activity due to their synergistic effect. These findings suggest that multiple-doped graphene could be used as a metal-free electro catalyst. The ORR activity of ternary-doped graphene, which uses three distinct heteroatom elements (e.g., NPB), was equivalent to commercially available Pt catalysts. To maximise their electro catalytic efficiency, the competitive doping mechanism of three distinct atoms with regard to the hosting material should be explained, especially by considering the

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doping sequence of the heteroatom elements. The presence of a high density of flaws inside the graphene matrix is the major distinguishing property of graphene oxide. It is critical to understand how the three different atoms are competitively introduced within the graphene network or at the edge of the graphene homogeneously or heterogeneously depending on the doping sequence when considering simultaneous thermal reduction and heteroatom introduction in the thermal doping process, as well as the electrocatalytic reaction occurring on its exposed surface. Using DNA and B₂O₃ as sources, ternary-doped Reduced Graphene Oxides (RGOs) were produced in a two-step procedure and assessed as ORR electrocatalysts. Graphene oxide was thermally decreased by introducing nitrogen and phosphorus atoms at the same time utilising DNA, which contains nitrogen-containing nucleobases and phosphorus groups. Then, using B₂O₃ as the source, additional doping with B was carried out. The relative proportions and configurations of the three distinct atoms inside RGO were influenced by their atomic sizes, resulting in variable electrocatalytic activity of the catalysts for ORRs. To increase the electrocatalytic performance of ternary doped graphenes, the doping sequence should be adjusted. Multiple-doped RGOs were evaluated as electrocatalysts for ORRs using a two-step doping procedure that used DNA and B₂O₃ as dopant sources. Ternary doped RGOs containing N, P, and B atoms were produced by a two-step doping process that used DNA and B₂O₃ as dopant sources. By simultaneously adding N and P atoms from the DNA, dark brown coloured GO was thermally reduced to black coloured RGO at 800 °C in argon. Thermal doping with B₂O₃ at 1000 °C in argon was used for the extra B doping. The overall number of heteroatoms in the two-step synthesised ternary RGO (about 9.3%) is significantly lower than in the single-step synthesised ternary RGO. However, because to the increased amount of P, the two-step RGO had a higher ORR electrocatalytic activity than the single-step RGO. The amount of P within the two-step ternary RGO is estimated to be around 2.36 at percent based on the overlap of the B 1s and P 2p XPS spectra. Even though the heteroatoms in the graphene matrix might cause charge redistribution and create more catalytic active sites, their relative concentration and configurations are critical for maximising the electrocatalytic performance of multi-heteroatom-doped graphenes.