Restacking of N-doped Graphene Layers for the Enhanced ORR Activity in Acidic Media

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Graphene, which is a two-dimensional layer structure of sp^2-hybridized carbon, has been highlighted recently as a promising material for energy conversion. Herein, graphene-derived catalysts are developed for application in oxygen reduction reactions (ORRs) in an acidic media *via* heat-treatment with dicyandiamide and a small amount (< 1 wt.%) of transition metals. In ORRs, bare graphene exhibits 0.58 V (*vs.* RHE) of onset potential; however, it increases to ~ 0.9 V through modification steps and records 1.28 mA mg^{-1} of mass activity at 0.75 V. Through the correlation curve between the ORR activities and the number of graphene layers restacked, it is proposed that the stacking of a few layers (5-7 layers) is desirable in the ORRs rather than a single layer catalyst. The graphene-derived catalysts exhibit graphite properties of facile electron transfer as the restacking of graphene layers increases, without degradation of the pyridinic-N on the graphene edge.