Engineering graphene surface toward design of aggregation-resistant catalyst supports for advanced energy conversion

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Graphene has been considered an emerging fuel cell catalyst support due to its excellent chemical and electrical properties. The evaluation of the unit activity on each catalytic site (intrinsic) of low-loading supported metal nanoparticles (NPs), however, is often hampered by face-to-face aggregation of graphene sheets. Herein, we demonstrate the critical role of the expansion between sheets in a pivotal electrocatalytic process for green energy conversion through ethanol oxidation in acid medium. In order to reduce mass-transport resistances and incomplete utilization of the supported NPs, a one-step design strategy is proposed for tuning a desired physicochemical property of graphene: surface area. This step is based on the principle that the apparent activity is governed by the extrinsic activity, *i.e*., the number of exposed active sites for a particular mass loading. Our design principle is achieved by a two-stage method involving a chemical delamination process of graphite (1) with an *in situ* surfactant functionalization/intercalation–reduction approach (2). As a result, not only an interlayer expansion was attained but also a short-ranged layered structure was assembled. This structural reorganization substantially affected the ethanol oxidation reaction (EOR) over platinum NPs. The unique nanoarchitecture provided a high density of EOR active sites, which incurred in a specific current value increased by about 2.5 and 5.4 times compared to platinum supported on state-of-the-art carbon black and restacked graphene, respectively.

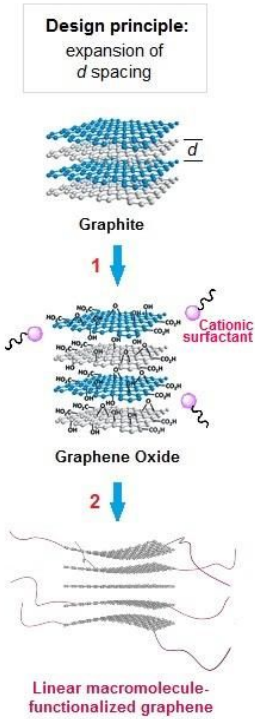

Fig. 1. Scheme for fabrication of aggregation-resistant graphene sheets. Adapted from [1] and [2].

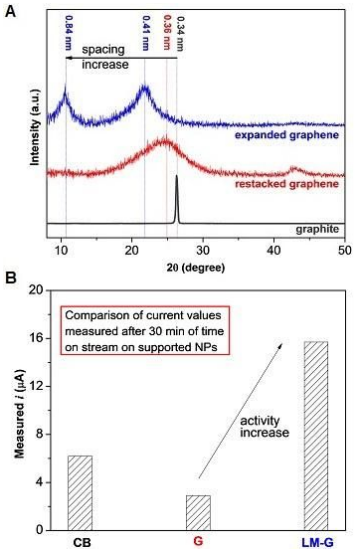

Fig. 2. (A) XRD showing the expansion between sheets. **(B)** Chronoamperometry of the EOR on NPs over carbon black (CB), graphene (G) and linear macromolecule-functionalized graphene (LM-G).

References

[1] Nam *et al*., Carbon, 50 (2012) 3739
[2] Seo *et al*., Adv. Mater., 29 (2017) 1