Materials Science & Engineering Doctoral Defense

Ultrafast, Scalable Manufacturing of Holey Graphene for High-Performance Electrochemical Applications

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Abstract

Nanoholes on the basal plane of graphene can provide abundant mass transport channels and chemically active sites for enhancing the electrochemical performance, making this material highly promising in applications such as supercapacitors, batteries, desalination, molecule or ion detection, and biosensing. However, the current solution-based chemical etching processes to manufacture these nanoholes commonly suffer from low process efficiency, scalability, and controllability, because conventional bulk heating cannot promote the etching reactions. Herein, a novel manufacturing method is developed to address this issue using microwave irradiation to facilitate and control the chemical etching of graphene. In this process, microwave irradiation induces selective heating of graphene in the aqueous solution due to an energy dissipation mechanism coupled with the dielectric and conduction losses. This strategy brings a remarkable reduction of processing time from hour-scale to minute-scale compared to the conventional approaches. By further incorporating microwave pretreatment, it is possible to control the population and area percentage of the in-plane nanoholes on graphene. Theoretical calculations reveal that the nanoholes emerge and grow by a repeating reduction–oxidation process occurring at the edge-sites atoms around vacancy defects on the graphene basal plane. The reduced holey graphene oxide sheets obtained via the microwave-assisted chemical etching method exhibit great potentials in supercapacitors and electrocatalysis. Excellent capacitive performance and electrocatalytic activity are observed in electrochemical measurements. The improvements against the non-holey counterpart are attributed to the enhanced kinetics involving ion diffusion and heterogeneous charge transfer.

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