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Nitrogen-doped graphene hydrogels as potential adsorbents and photocatalysts for environmental remediation

The chemical modification of self-assembled graphene hydrogels with heteroatoms is a topic of emerging interest to harness the unique characteristics of two-dimensional (2D) graphene for macroscopic applications. However, optimization of the concentration as well as the bonding state of heteroatoms at the atomic scale to effectively tune the physicochemical characteristics of such three-dimensional (3D) graphene-based macroscopic assemblies (GMAs) remains largely unexplored. Herein, we synthesized a series of mechanically strong and lightweight nitrogen (N)-doped graphene hydrogels (NGHs), with different doping concentrations, through a simple one-pot hydrothermal reaction and systematically evaluated their performance as both adsorbents and photocatalysts for environmental remediation. Acridine orange (AO) was chosen as a model pollutant. The successful incorporation of N atoms into the carbon lattice of the macroscale 3D graphene-based materials was verified by Raman spectroscopy, Fourier transform infrared spectroscopy (FTIR) and X-ray photoelectron spectroscopy (XPS). Although the N content of the graphene macroassemblies varied inversely with doping density, a conspicuous increase in specific surface area was observed at all doping levels, resulting in a higher adsorption capacity and surface reactivity than the undoped hydrogel. The adsorption equilibrium was best represented by the Langmuir isotherm (with maximum monolayer coverage of 124 mg g^{-1} at 25 °C) while the adsorption kinetics followed both the pseudo-first and pseudo-second order rate expressions. Further, the NGHs could effectively photodegrade 20 mg L^{-1} AO solution by almost 70% within 5 h of visible light irradiation. The good photoreduction ability of NGHs originates from the synergistic effect of N functionalization and 3D interconnected mesoporous network structure, leading to greater uptake of AO, better absorption of visible light and rapid spatial separation of photogenerated electron–hole pairs.

Figures

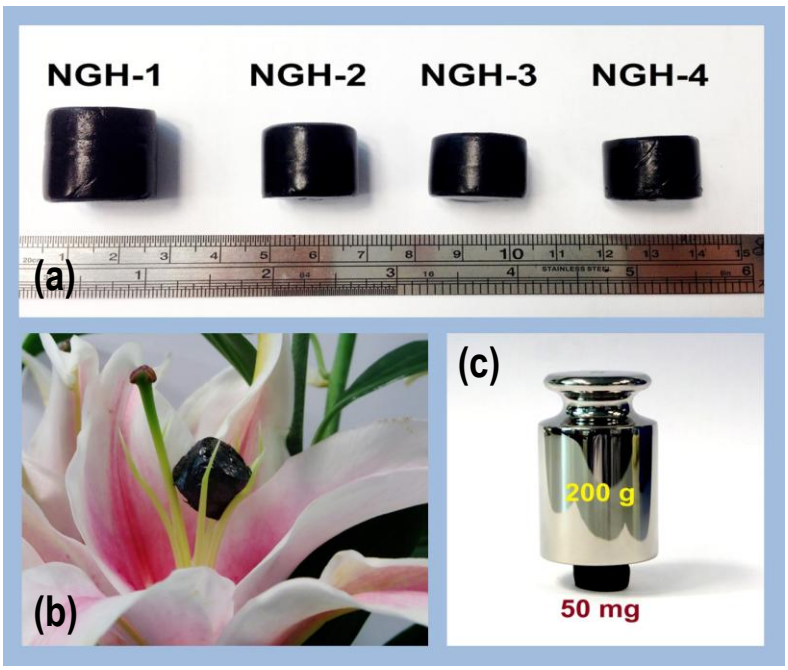

Figure 1: (a) Digital photographs of the as-synthesized NGH bulk materials with different N-doping levels. (b) NGH-2 propped up on the stamen of a flower (*Lillium* sp.). (c) NGH-2 (height × diameter = 1.45 cm × 0.95 cm) supporting a 200 g knob weight, more than 4000 times its own weight of 50 mg.

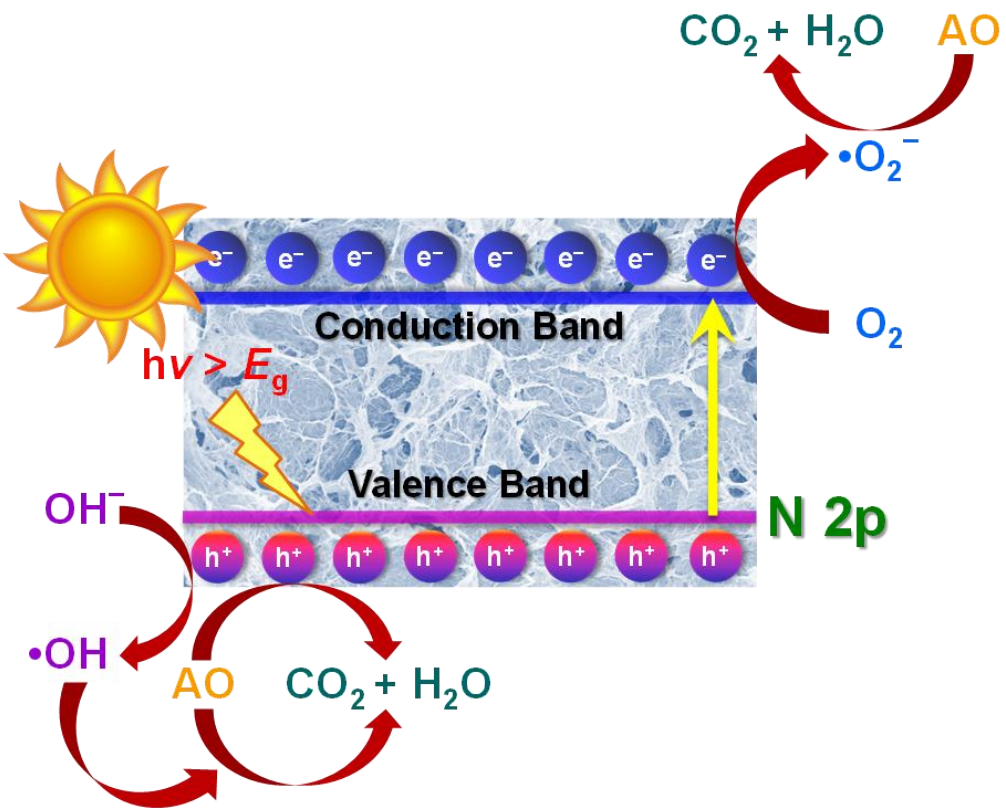

Figure 2: Plausible reaction mechanism for degradation of AO over the N-doped graphene monoliths upon irradiation with visible light.