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Three-Dimensional Hierarchically Porous Floatable MoS_2/Graphene Aerogel for Photocatalytic Oxidation of Micropollutants

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Transforming recalcitrant pollutants into less concerning products under ambient conditions via heterogeneous photocatalysis is desirable from the viewpoint of sustainable development. Despite recent progress in designing highly active photocatalysts, inefficient light harvest, sluggish mass transfer, instability of catalysts, and difficulty in retrieval following decontamination impede their practical large-scale applications. Herein, we overcome these challenges by developing a floatable photocatalytic platform via in-situ growth of MoS_2 nanoclusters on the interpenetrating networks of graphene aerogel (denoted as 'MGA'). Because porosity is a cardinal factor governing the pollutant infiltration rate and hence the kinetics of the degradation process, we further modified the pore architecture and interconnectivity of MGA via a facile and scalable chemical activation technique. The positioning of the hierarchically porous monolithic composite (denoted as '*h*-MGA') at the air–water interface not only enables efficient utilization of incident light energy but also minimizes water-induced light attenuation. In addition, the tailored interpenetrating network morphology facilitates efficient charge transport and rapid ion diffusion, and also doubles as a mechanically resilient and chemically stable scaffold for oxidation and reduction reactions. Consequently, a high photocatalytic degradation of 97% for tetracycline (TC) can be achieved over *h*-MGA under visible-light illumination, without adding any external chemical oxidants. In fact, after 120 minutes of exposure to visible light, up to 67% of the total organic carbon is removed, suggesting that TC may be fully mineralized over an extended reaction period. Additionally, the photocatalysis byproducts do not pose any conceivable toxicological risk, and can thus be safely discharged in the environment, without any post-treatment operations. As a figure-of-merit for the commercial potential of *h*-MGA, we demonstrated the photocatalytic degradation of TC in four different real matrices, viz., tap water, pond water, municipal wastewater, and hospital wastewater, with treatment efficacy several folds higher than contemporary photocatalytic systems. The plausible photocatalysis mechanism of *h*-MGA and the possible degradation pathways of TC were also analyzed via a series of reactive oxygen species-scavenging experiments and liquid chromatography–mass spectrometry findings. Overall, our as-developed *h*-MGA, with exceptional photocatalytic properties, improved durability, and facile retrieval opportunities, represents an appealing option for various water treatment applications.