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Holey structured graphitic carbon nitride thin sheets with edge oxygen doping via photo-Fenton reaction with enhanced photocatalytic activity

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ABSTRACT

Holey structured graphitic carbon nitride thin sheets with edge oxygen doping (HS g-C_3N_4-O) via photo-Fenton reaction were reported. The HS g-C_3N_4-O exhibited high surface area of 348 m^2 g^{-1} and narrow bandgap (2.434 eV), hence producing high photocatalytic activities for both H_2 generation and Rh.B degradation under solar light irradiation.

1. Introduction

Photocatalytic hydrogen generation from water using photo-driven reaction has attracted considerable attention as an ideal green technology [1–3]. Of these, a stable photocatalysts which high effectively converts solar energy into chemical fuels is an appreciated alternative. Among various semiconductors photocatalyst, metal-free graphitic carbon nitride (g-C_3N_4) with a suitable band gap and edges has emerged as a new class of photocatalyst due to its nontoxicity, thermal and chemical stability. However, the latest documented quantum yields for H_2 production using pristine g-C_3N_4 does not exceed 4% [4–7], which is far from satisfactory for further application.

As is known, an excellent photocatalyst must possess a high specific surface area to take place redox reactions and a minimizing recombination to take advantage photo-induced charge. Undeniably, the bulk g-C_3N_4 has poor surface area and the high recombination rate. To improve the essential properties of g-C_3N_4, various strategies including heteroatom doping [8–11], layer exfoliation [12], heterojunction configuration [12–15] and carbonaceous scaffold [16–18] have been adopted. Meanwhile, Oxygen heteroatom has been successfully introduced by a simple H_2O_2 oxidation and its photoactivity has been improved to some extent [19,20]. However, the strongly oxidizing agent of H_2O_2 would be easily break the g-C_3N_4 structure whose is possibly converted into carbon or nitrite oxides. In this sense, rationally controlling the oxidation process would leave behind many vacancies, which could be gradually extended into nanopores texture. The expected oxygen doped g-C_3N_4 with high porosity is able to exhibit impressive photocatalytic activity due to simultaneous doping effect and high active sites.

In this study, a holey structured g-C_3N_4 with edge oxygen doping (HS g-C_3N_4-O) was successfully obtained using photo-Fenton reaction in presence of Fe^{3+}/Fe^{2+} and H_2O_2. First, bulk g-C_3N_4 was oxidized and exfoliated into g-C_3N_4 thin sheets with abundant oxygen-containing groups, which based on Niu et al. report [12]. Furthermore, it has also been demonstrated that the photo-Fenton reaction can generate hydroxyl radicals ($OH^{\bullet}$) under the photoassisted catalysis of Fe^{3+}/Fe^{2+} in present of H_2O_2, and which has been considered as one of the most powerful oxidizing species [21]. Thus, the g-C_3N_4 thin sheets with abundant oxygen-containing groups as active defective sites can be partially oxidized and etched, leaving behind carbon vacancies, which gradually extend into nanopores [22,23]. The as-resulted HS g-C_3N_4-O demonstrated a narrow bandgap of 2.434 eV and high surface area of 348 m^2 g^{-1}, thus presenting highly efficient photocatalytic activity for H_2 generation and Rh.B degradation.

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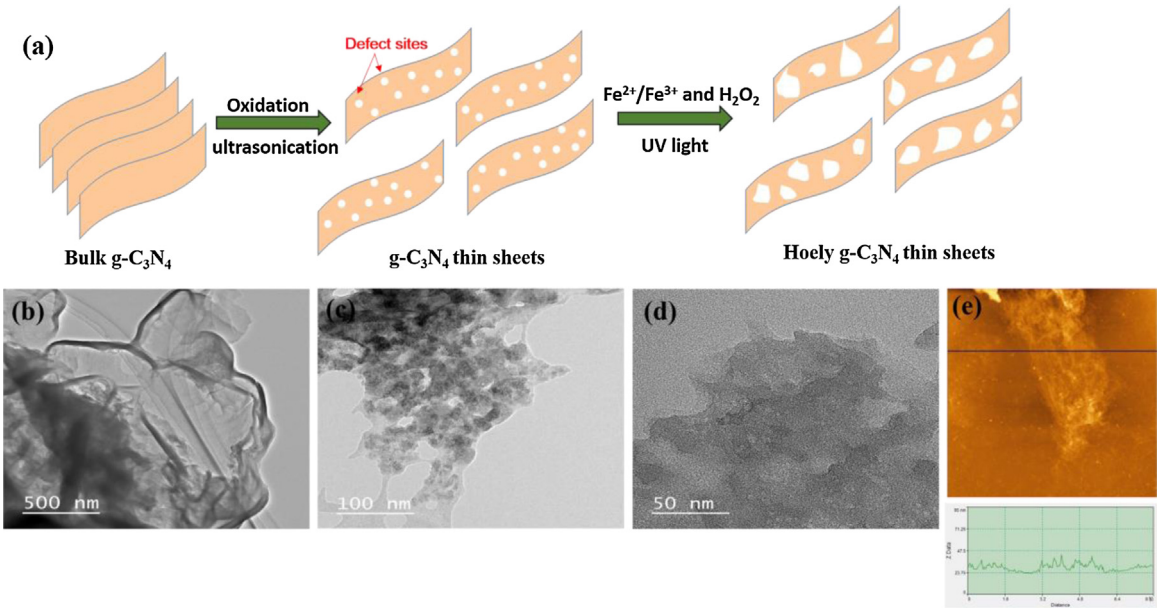

Fig. 1. (a) Schematic illustration of the HS g-C_3N_4-O formation via photo-Fenton reaction. (b) TEM image of oxidized g-C_3N_4 thin sheets. (c and d) TEM image of the HS g-C_3N_4-O. (e) AFM image of the HS g-C_3N_4-O.

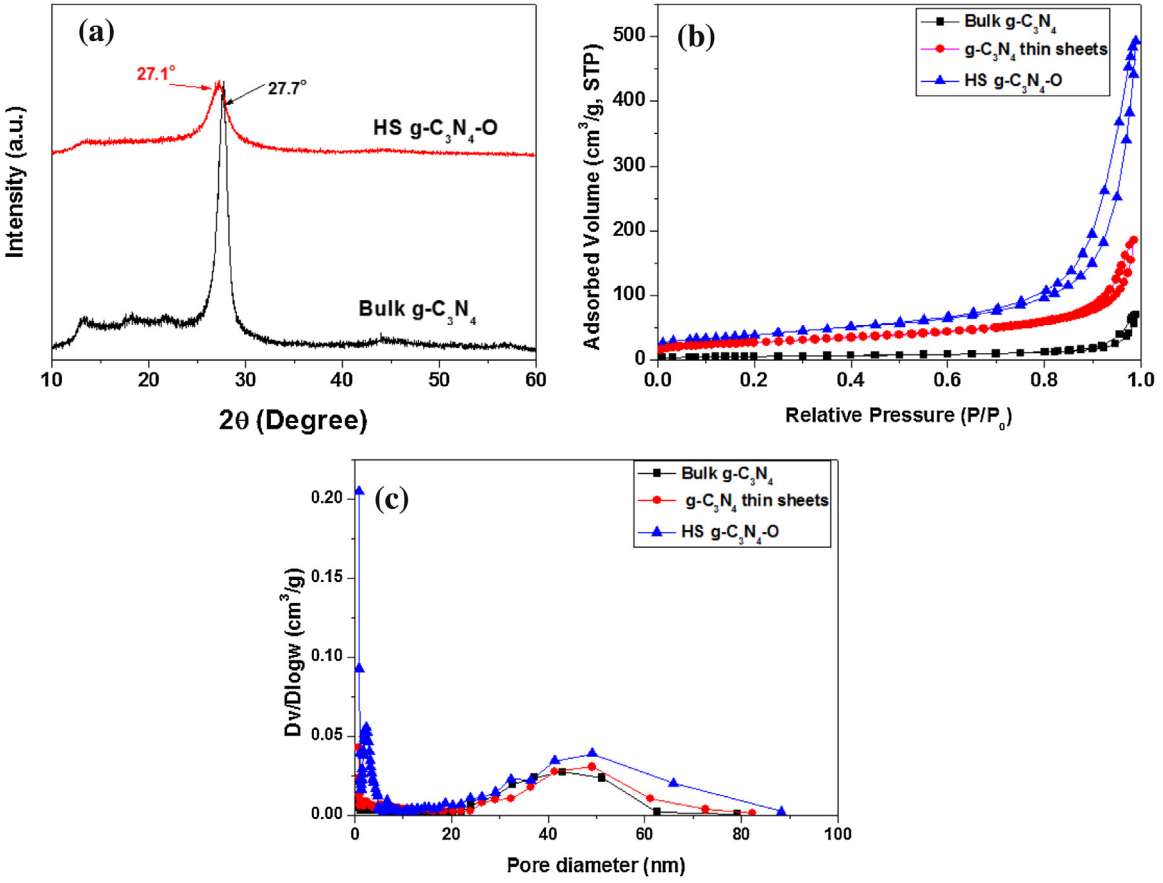

Fig. 2. (a) XRD patterns of the bulk g-C_3N_4 and HS g-C_3N_4-O. (b) Nitrogen adsorption–desorption isotherms and the corresponding pore size distribution curves (c) of the bulk g-C_3N_4, g-C_3N_4 thin sheets and HS g-C_3N_4-O.