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Facile fabrication of amine-functionalized g-C3N4 nanosheets for enhanced nitrogen photofixation

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1.Introduction
The fabrication of g-C3N4 nanosheets is considered to be an effective method for improving its photocatalytic activity. Unfortunately, the time consuming multi-step synthesis and low yields of common approaches limit practical applications of both top-down and bottom-up synthesis methods.[1] Herein, we used a one-pot approach to obtain amine-functionalized ultrathin g-C3N4 nanosheets by collecting the gaseous products from thermal polymerization of urea at 550 ℃.
2.Results and Discussion
As a product of urea decomposition, cyanic acid can spontaneously and rapid polymerize to generate cyanuric acid.[2] The cyanuric acid then reacts with NH3 to produce ammelide, and melamine is obtained by the further reaction of ammelide and NH3. Finally, ultrathin g-C3N4 nanosheets were obtained in the larger porcelain crucible by the copolymerization process of melamine in the gaseous phase (shown in Fig. 1).

Fig.1 The different formation processes of g-C3N4 in gaseous and solid phase.
As shown in Fig. 2, the ultrathin and uniform g-C3N4 nanosheets could be easily achieved and the thickness was about 2.0 nm.

Fig.2 TEM and AFM images of g-C3N4 nanosheets.
As shown in Fig. 3a, the g-C3N4 nanosheets exhibited superior photocatalytic activity compared with bulk g-C3N4. However, the production of NH4+ decreased in the presence of N2, indicating that O2 played a dominant role in the photocatalytic N2 fixation by the g-C3N4 nanosheets (shown in Fig. 3b).

Fig.3 (a) Visible-light nitrogen fixation over bulk g-C3N4 and g-C3N4 nanosheets. (b) Visible-light nitrogen fixation under different atmospheres over g-C3N4 nanosheets.
3.Conclusion
The obtained g-C3N4 nanosheets have large surface area, high reduction potential and enhanced charge-carrier separation rate, thus promoting its activity for photocatalytic nitrogen fixation.

[1] P. Yang, H. Ou, Y. Fang and X. Wang, Angew. Chem. Int. Ed., 2017, 129, 4050-4054.
[2] G. Herzberg, C. Reid, Discussions of the Faraday Society 9 (1950) 92-99.

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