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Soft-chemical synthesis of mesoporous nitrogen-modified titania with superior photocatalytic performance under visible light irradiation

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HIGHLIGHTS

- Nitrogen-modified mesoporous titania was obtained by a soft-chemical route.
- An exfoliation–reassembling strategy was used to prepare the photocatalyst.
- Ethylamine was used as the intercalant and meanwhile provided *N* species.
- The photocatalyst possesses a high surface area and extended visible absorption.
- The photocatalyst show a high visible activity for degrading methyl orange.

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ABSTRACT

Exploring novel titania-based semiconductor photocatalysts with both high specific surface area and expanded visible light response is still a challenge. In this work, we report the first synthesis of mesoporous nitrogen-modified titania by a facile and convenient exfoliation–reassembling strategy. Ethylamine is used for the delamination of layered titanate, and meanwhile, also serves as a source of nitrogen. X-ray diffraction patterns and transmission electron microscopy images reveal that the random reassembling between titanate nanosheets and anatase nanoparticles give rise to its porous structure. N_2 adsorption–desorption isotherms demonstrate that the obtained photocatalyst is fairly high in specific surface area and in mesoporosity (S_{BET} of ~215 $m^2\ g^{-1}$ and pore size of ~5.6 nm) for effective photocatalysis. The mesoporous titania photocatalyst possesses an extended absorption in the visible region due to the successful modification by NH_x species, which is confirmed using the X-ray photoelectron spectroscopic analysis, combined with the Fourier-transform infrared spectra. Photocatalytic tests reveal that the mesoporous nitrogen-modified titania material show an excellent catalytic performance for the degradation of organic compounds under visible light irradiation, which is much higher than those of the commercial P25, pristine protonic titanate, and N-doped mesoporous titania photocatalyst prepared from a template-free route. The present work provides a new way to develop efficient nitrogen-modified oxides materials with high specific surface area and enhanced visible light absorption for solar energy utilization.

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1. Introduction

Semiconductor photocatalysis has attracted increasing interest due to its potential applications for solving grave challenges such as environmental deterioration and energy exhaustion [1–3]. To develop efficient photocatalytic systems, high-quality semiconductor-based materials have been widely studied. Among a wide spectrum of semiconductors, titania has attracted significant attention due to its cheap availability, nontoxicity, hydrophilicity, stability and against photocorrosion. However, the wide band gap of titania confines its application to UV light activation. Given that less than 5% of the solar flux arrived at the earth's surface lies in this spectral region, utilization of natural solar light for practical applications should be improved by tuning the band gap response of titania to the visible region. Among a great variety of strategies for the modification of titania, anion doping [4–8], especially nitrogen doping has shown a great potential in improving its visible light photocatalytic activity.

The high electron–hole recombination rate (ca. 90% of carriers recombine rapidly after excitation [9,10]) of titania is no doubt another serious issue to be addressed in the field. Ever since Antonelli and Ying [11] first synthesized mesoporous titania in 1995, incessant attempts have focused on designing titania materials with

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