

高结晶度氮掺杂介孔 TiO_2 的制备及光催化活性

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摘要 以钛酸丁酯为钛源, 尿素为氮源, 聚丙烯酰胺 (PAM) 和聚乙二醇 (PEG) 为复合模板剂, 采用溶胶-凝胶法, 在氮气和空气气氛中分段煅烧, 制得高结晶度氮掺杂介孔 TiO_2 光催化剂。利用 X 射线衍射、透射电镜、 N_2 吸附-脱附、X 射线光电子能谱和紫外-可见漫反射光谱等技术对其进行了表征。结果表明, 当 PAM 和 PEG 的质量比为 1:4 时, 先在氮气中 600 °C 煅烧, 后在空气中 500 °C 煅烧所得样品是锐钛矿相, 具有良好的孔隙结构和较高的结晶度, 平均孔径为 5.11 nm, 晶粒尺寸为 12.5 nm, 比表面积 110.8 m²/g。掺杂介孔 TiO_2 的氮主要以取代氮和化学吸附分子 $\gamma\text{-N}_2$ 的形式存在, 少量以间隙氮形式存在。氮掺杂使 TiO_2 的能带变窄, 吸收带边明显红移, 且使光吸收强度显著增大。光催化降解甲基橙实验结果表明, 与未掺杂样品相比, 氮掺杂介孔 TiO_2 在可见光作用下表现出较高的催化活性。

关键词: 介孔二氧化钛 氮掺杂 高结晶度 光催化活性 甲基橙

Abstract: Highly crystalline nitrogen doped mesoporous TiO_2 photocatalysts were fabricated by the sol-gel method using tetrabutyl titanate as the Ti source, urea as the N source, and polyacrylamide (PAM) and polyethylene glycol (PEG) as the templates, and then by calcining in nitrogen and air. The photocatalysts were characterized by X-ray diffraction, transmission electron microscopy, N_2 adsorption, X-ray photoelectron spectroscopy, and UV-Vis spectroscopy. When the mass ratio of PAM and PEG was 1:4, the sample prepared by calcining at 600 °C in nitrogen and 500 °C in air had the anatase phase and a mesoporous structure and high crystallinity. The average pore size, crystallite size, and specific surface area were 5.11 nm, 12.5 nm, and 110.8 m²/g, respectively. Nitrogen atoms were incorporated into the TiO_2 lattice mainly as substitutional N and molecularly chemisorbed $\gamma\text{-N}_2$, and a small amount of interstitial N. Nitrogen doping narrowed the band gap and allowed light absorption in the visible light region. Compared with undoped mesoporous TiO_2 , the absorption band edge of nitrogen doped samples exhibited a red shift and the light absorption intensity was increased. Photocatalytic degradation of methyl orange showed that the nitrogen doped mesoporous TiO_2 had a higher photocatalytic activity than undoped mesoporous TiO_2 under visible light.

Keywords: mesoporous titanium dioxide, nitrogen doped, high crystallinity, photocatalytic activity, methyl orange

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