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P 掺杂量对纳米 TiO_2 结构及其光催化甘油水溶液制氢性能的影响

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摘要 以钛酸丁酯为前驱体, 以 NaH2PO4 为掺杂离子给体, 采用溶胶-凝胶法制备了系列 P 掺杂的 TiO2 光催化剂, 运用 N2 吸附-脱附、透射电子显微镜、X 射线衍射、傅里叶变换红外吸收光谱、激光拉曼光谱、紫外-可见光漫反射等技术对催化剂进行了表征. 结果表明, 适量掺杂的 P 可以进入 TiO2 骨架中, 而掺杂量过高时, P 将溶解于 TiO2 晶格间隙中; P 掺杂后的 TiO2 均为具有介孔结构的锐钛矿晶型纳米颗粒, 其晶粒变小, 分散度明显提高. 适量 P 掺杂增大样品的比表面积, 并使得 TiO2 禁带内引入杂质能级, 降低了禁带能量, 增加了光生电子和空穴的分离性能, 提高了 TiO2 的吸光性能. 光催化甘油水溶液制氢反应结果表明, P 掺杂的 TiO2 表现出远高于纯 TiO2 的光催化活性; 5%P 掺杂样品在紫外光和模拟太阳光辐射下, 其最高产氢速率可分别达 1838 和 209 μmol/(g•h). 这与掺 P 后晶粒变小、比表面积增大、禁带能量降低以及光生电子和空穴的分离性能增加有关.

关键词:

Abstract: Using tetrabutyl-titanate and sodium dihydrogen phosphate as raw materials, TiO_2 and P^{5+}-doped TiO_2 semiconductors were prepared by the sol-gel method. Their pore distribution, crystal structure, surface compositions, and photoabsorption properties were investigated by N_2 adsorption-desorption, X-ray diffraction, Fourier transform infrared spectroscopy, FT-Raman, transmission electron microscopy and UV-Vis diffuse reflectance spectrum. The results show that P^{5+}-doped TiO_2 exists in nano-particles of anatase phase with mesoporous structure. P^{5+}-doped TiO_2 samples exhibit much smaller crystallite size and much higher specific surface area than pure TiO_2. P-doped TiO_2 samples show an extension of light absorption into the visible region, which mainly originates from the doping process with the formation of new energy level of P^{5+} between conductor band and valence band of TiO_2 to reduce the energy gap and the electron-hole recombination rate. The P^{5+}-doped TiO_2 samples display improved photocatalytic activity for H_2 production from glycerol solution, and 5% P^{5+}-doped TiO_2 shows a maximum H_2 production rate of 1838 μmol/(g•h) under UV irradiation and 209 μmol/(g•h) under simulated-solar irradiation, respectively, which is related to the decrease of crystal particles, increase of specific surface area, and the reduce of the energy gap and the electron-hole recombination rate.

Keywords: phosphorus, doping, titanium dioxide, nano-particle, photocatalysis, glycerol solution, hydrogen production

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