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Enhanced photocatalytic performance of Ag@TiO₂ for the gaseous acetaldehyde photodegradation under fluorescent lamp



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HIGHLIGHTS

- The Ag@TiO₂ core-shell structure is beneficial to more light absorption and efficient charge separation.
- The adsorptive property of acetaldehyde could reflect the effective reactive sites more straight in comparison to S_{BET}.
- The roles of reactive radicals were investigated through combining ESR tests and scavenger experiments together.
- O_2^- played the predominant role in the photocatalytic process of acetaldehyde.

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G R A P H I C A L A B S T R A C T

The outstanding performance of Ag nanowires@TiO₂ composite and the thoroughly understanding of acetaldehyde photodegradation mechanism would cast light on the purposively design and optimization of TiO₂-based catalysts.



ABSTRACT

The ever increasing problem of air pollution has provoked the research and development of highly-efficient photocatalysts. Herein, Ag nanowires@TiO₂ composite photocatalyst with improved photocatalytic performance was fabricated by a facile one-step solvothermal procedure. The formation of one-dimensional Ag nanowires@TiO₂ core-shell nanostructures could not only broaden the light-absorbing range of TiO₂ catalysts through the surface plasmon resonance effect of Ag nanowires, but also enable the effective separation of photoinduced electron-hole pairs. Under the irradiation of a 260 W fluorescent lamp, the composite with 0.5 wt % Ag nanowires exhibited the highest photocatalytic activity in short contact time (4.8 min), and the corresponding gaseous acetaldehyde removal ratio was 72%, which was much higher than that of bare TiO₂ (37%). The photocatalyst also showed ultrastable activity in the 15 weeks usage, which ensured their practical applications in the air purification field. An in-depth mechanism of the photoecomposition of acetaldehyde was proposed on the basis of the electron spin resonance (ESR) tests and the scavenger experiments. 'O₂⁻ reactive radicals was found to play a predominant role in the oxidation and decomposition of acetaldehyde. The outstanding performance of the composite materials and the thoroughly understanding of the reaction mechanism would cast light on the purposively design and optimization of TiO₂-based catalysts.

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1. Introduction

Nowadays, with the rapid industrial development and the huge consumption of fossil fuels, diverse gaseous pollutants have been released into our living environment, which has resulted in serious environmental problems, including acid rain, photochemical smog and haze [1,2]. Many efforts have been made to solve this troublesome issue. Among all the possible solutions, photocatalysis is an ambient temperature process which can make use of solar energy to photodecompose various organic contaminants without secondary pollution. However, most of the research works today concentrate on the photodecomposition of liquid pollutants such as Methyl Orange (MO) and Rhodamine B (RhB) [3]. Due to the intrinsic differences between the photodecomposition of liquid pollutants and gaseous pollutants, traditional photocatalysts suitable for liquid pollutants cannot be applied to the decomposition of gaseous pollutants directly [4]. The decomposition of pollutants relies on the adsorption of pollutants on the surface of photocatalysts. Unlike liquid pollutants, the concentration of gaseous pollutants is relatively low. The fast and disordered movement of gas molecules further lowered the residue time of gaseous pollutants. The lower concentration of pollutants and the shorter residue time proposed a higher requirement for the photocatalytic capacity and adsorption ability of photocatalysts.

TiO₂ has long been regarded as an ideal photocatalyst to deal with environmental concerns, owing to its non-toxicity, environmentally benign nature, low-cost, abundance, chemical and thermal stability [3-8]. Despite all those advantages, TiO₂ is still being criticized for its poor light-harvesting capacity and the rapid recombination rate of photoinduced electron-hole pairs, which has limited the photocatalyst capacity of TiO₂-based photocatalysts and its applications in the photodecomposition of gaseous pollutants. Only 4% solar energy can be utilized by TiO_2 due to its wide band gap (3.2 eV) [9]. In order to make full use of the abundant solar energy resource, the development of TiO₂ photocatalysts with enhanced light-harvesting capacity is indispensable [10,11]. On the other hand, the rapid recombination of electron-hole pairs, which is harmful to the photocatalytic performance of catalysts, should be suppressed effectively [12-14]. Reviews [15-18] have revealed that the formation of TiO₂ heterostructures often brings some attractive benefits for the photocatalysts. Works have been done to optimize the performance of TiO₂-based photocatalysts. Li et al. [19] and Lu et al. [20] enhanced the light-harvesting capacity of TiO₂ by coupling it with C₃N₄. The utilization of the surface plasmon resonance effect (SPR) of noble metals is an effective way in the optimization of TiO₂ catalysts [21-24]. Under the illumination of light with certain wavelength, the free electrons of the noble metals such as Au, Ag or Pt nanoparticles would couple with the incident light at a resonant frequency, which leads to a localized electronic field around the metal nanoparticles and results in the improved light-harvesting capacity of TiO₂ catalysts. TiO₂ nanoparticles/Au nanobelts [25], Ag-Ag₂O/reduced TiO₂ [26], Ag nanoparticles@TiO₂ nanotubes [27], (Au, Ag, Cu and Pt)/anatase particles [28], Au/TiO2 nanowires [29], Au-decorated ZnO [30] and ZnO-Au-SnO₂ [31] have been proved to be the efficient photocatalysts. To realize the low recombination rate, Yang and coworkers [32] used graphene as electron trap for promoting the separation of electron-hole pairs. One-dimensional electron transport channel also turns out to be helpful for the efficient separation according to the research results reported previously. The usage of carbon nanotubes as the electron traps in the CNT/TiO₂ composite resulted in the effective separation of electron-hole pairs and an improved photocatalytic performance [33-35].

However, most of the efforts reported can only improve either the separation of electron-hole pairs or the light-harvesting capacity of TiO_2 -based catalysts. The fabrication of composite photocatalysts with both low recombination rate and enhanced light-harvesting capacity would be feasible for achieving superior photocatalytic performance. Ag nanowires should be a good choice owing to its SPR effect and high

conductivity. Herein, an Ag nanowires@TiO₂ core-shell heterostructure with enhanced photocatalytic ability was synthesized by a facile one-step solvothermal treatment. Compared with bare TiO₂ catalyst, an enhanced removal ratio of gaseous acetaldehyde was observed when applying the Ag nanowires@TiO₂ core-shell heterostructure as photocatalyst. The related highest decomposition rate constant (0.71959 h⁻¹) is almost 2.6 times larger than that of bare TiO₂ (0.27739 h⁻¹). In the purpose of understanding the decomposition mechanism of acetaldehyde and effect of Ag nanowires, ESR tests and scavenge experiments were carried out. The coupling with Ag nanowires would benefit the generation of \cdot O₂⁻, which was found to be essential for the photodecomposition of gaseous acetaldehyde. The indepth mechanism proposed in this work would shed light on the further optimization of TiO₂-based photocatalysts and the effective decomposition of gas pollutants.

2. Experimental

2.1. Materials

Ag nanowires were purchased from Zhejiang Kechuang Advanced Materials Technology Co. Ltd. Tetrabutyl Titanate (TBOT) was purchased from Sinopharm Chemical Reagent Co. Ltd. p-benzoquinone (PBQ) and 2,2,6,6-tetramethyl-1-piperidinyloxy (TEMPO) were purchased from Aladdin Industrial Corporation. Deionized water was used.

2.2. The synthesis of Ag nanowires@TiO₂ core-shell heterostructure photocatalyst

In a typical process, 0.875 mg, 1.75 mg, 2.625 mg Ag nanowires were dispersed in 30 mL ethanol and 1 mL DI water by ultrasonication for 1 h. 1.5 mL TBOT was then added dropwise into the above mixture under magnetic stirring. The mixture was transferred into a 50 mL Teflon-lined autoclave and aged at 160 °C for 10 h. Afterwards, the suspension was centrifuged and washed with ethanol for several times. The obtained precipitates were dried in a vacuum oven overnight to obtain powder samples. The as-prepared samples were labeled as A0.25 T, A0.5 T, A0.75 T, where A stands for Ag nanowires and T for TiO₂, while 0.25, 0.5, 0.75 represent the mass ratio of Ag nanowires to TiO₂ are 0.25%, 0.5%, 0.75%, respectively. Bare TiO₂ was synthesized without adding Ag nanowires and denoted as A0T.

2.3. Characterization

X-ray diffraction (XRD) patterns were obtained on a D8 ADVANCE X-ray diffractometer (BRUKER AXS GMBH, German) with Cu Ka radiation ($\lambda = 0.15418$ nm, 20 varied from 20° to 80°, 8°/min), the mode is continuous, the voltage is 40 kV, the current is 40 mA. UV-Vis spectra were collected on a Perkin-Elmer Lambda 950 spectrometer (250-800 nm). The Brunauer-Emmett-Teller (BET) specific surface area (SBET) of the samples was analyzed by a Micromeritics ASAP 3000 nitrogen adsorption apparatus (All the samples were degassed at 120 °C for 5h before nitrogen adsorption measurements were taken). The photoluminescence (PL) spectra were recorded on a Perkin-Elmer Luminescence spectrometer 55 (LS55) at an excited wavelength of 320 nm, the emission and excitation slitwidth are both 5 nm, and the scanning rate is 500 nm/min, the PL measurements were performed in powder. Fourier transform infrared spectroscopy (FTIR) spectra were measured by a Thermofisher iN10 iZ10 infrared spectrophotometer with a KBr pellet technique. Raman spectra were collected on a Thermal Dispersive Spectrometer using a laser with an excitation wavelength of 532 nm at laser power of 7 mW. The surface morphology of as-prepared powder was analyzed by using a field-emission scanning electron microscopy (FESEM) equipped with an Energy Dispersive Spectrometer (EDS) (FESEM, Magellan 400) (Firstly, powder was dispersed in ethanol by ultrasonication for 5 min. And then, the mixture was dropped on the

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