

Short communication

Visible-light driven C@TiO₂ porous films: Enhanced photoelectrochemical and photoelectrocatalytic performance



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ABSTRACT

Visible-light driven C-modified TiO₂ film (C@TiO₂ film) was prepared on a conducting substrate and studied as photoanode for photoelectrochemical and photoelectrocatalytic (PEC) properties. The as-prepared material was characterized by SEM, TEM, XRD, and diffuse reflectance spectra. The C@TiO₂ film with multiporous structure exhibited enhanced electrochemical properties and excellent PEC performance, with a seven-fold improvement of photocurrent intensity as compared to unmodified TiO₂. The PEC improvement may be resulting from extended visible light-harvesting, low electron transfer resistance, and promoted photocurrent response. This study could provide a new perspective on preparing high performance photoelectric catalyst films.

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1. Introduction

Titanium dioxide (TiO₂) has been extensively investigated as a photoanode for photoelectrocatalytic (PEC) organic pollution abatement owing to its high activity, good chemical and photonic stability, and low cost [1–3]. However, a drawback, regarding commercially available TiO₂, is that it is mainly active in the UV that accounts for less than 5% of the total solar radiation [4,5]. To improve the optical response and photocatalytic activity of TiO₂ over the entire solar spectral range, numerous efforts have been made to vary the chemical composition and structure of TiO₂ by cation or anion doping, heterojunctions and band engineering that generate donor or acceptor states in the band gap [6–14]. Very recently, surface modification of TiO₂ by carbonaceous species aimed at solving the above problem has been proposed and attracted much attention [15–18]. The most advantageous morphology i.e., a core-shell structure, where nanometer-sized carbon species covers the TiO₂ nanoparticles with a thin homogeneous coating layer, maximizing heterojunction functionality without loss of light absorption, has been achieved by synthesizing nanostructured modified TiO₂ using the sol-gel combustion method [19]. In this system, a porous shell for reactant and product molecules/ions to pass through as well as an intimate interface for charge transfer between the core and the shell have also been realized simultaneously. However, most reported surface-modified TiO₂ photocatalysts for removal of recalcitrant organic

pollutants were used in the format of suspensions mediated by powder species [20,21]. In practical application, TiO₂ based films are more viable due to their easy separation recovery and simple post-treatment process [8,9,22,23]. Furthermore, the application of an additional bias voltage can further accelerate charge separation and reduce unwanted charge recombination processes, consequently promoting the photocatalytic oxidation degradation efficiency of pollutants [3,8,9]. Thus, a systematical and profound investigation on the synthesis, characterization and PEC performance of carbon modified TiO₂ films is highly desired.

In this study, visible-light driven electrode films i.e., C-modified TiO₂ films (denoted as C@TiO₂) by the packing of TiO₂ nanocrystal core nanoparticles wrapped with an amorphous carbon layer on a conducting substrate were prepared and studied as photoanode for degradation of organic pollutant. The C@TiO₂ films exhibited multiporous structure, extended visible light-harvesting, low electron transfer resistance, and promoted photocurrent response. As a proof-of-concept, we demonstrated that C@TiO₂ photoanodes exhibited excellent PEC performance toward the degradation of organic pollutants such as rhodamine B (RhB) and 1-naphthol (1-NP) under visible light by applying a small bias voltage (1.0 V).

2. Experimental

2.1. Preparation and characterization of C@TiO₂ films

C@TiO₂ nanoparticle was prepared by smoldering combustion sol-gel synthesis as previously reported [19]. The experiment details were described in the Supplementary materials. Then the as-prepared

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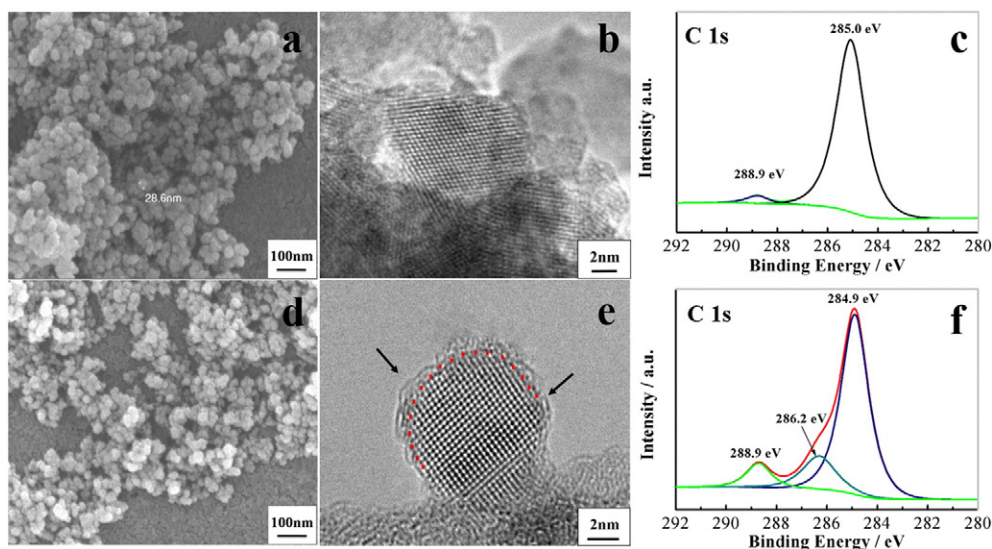


Fig. 1. SEM and TEM images of ref-TiO₂ (a & b) and C@TiO₂ (d & e) particles. Inset showing that an amorphous shell marked by dotted line covers the crystalline TiO₂ particles. Peak deconvolution of C 1s XPS core-level spectral from ref-TiO₂ (c) and C@TiO₂ (f).

C@TiO₂ powder, ethyl cellulose and terpineol with a weight ratio of 1:0.15:3 were mixed to form a viscous mixture. After that the C@TiO₂ films were fabricated by using a doctor-blade method, followed by heat treatment at 350 °C for 1 h and then 450 °C for 20 min. The C@TiO₂ films were observed by FESEM, XRD, UV–vis spectrophotometer.

2.2. Photoelectrochemical characterization of C@TiO₂

The photoelectrochemical performance of the C@TiO₂ films was evaluated using a three-electrode configuration with C@TiO₂ electrode, a saturated calomel electrode (SCE), and Pt wire as working, reference, and counter electrodes, respectively. The supporting electrolyte used here was in 0.5 M Na₂SO₄ solution. All the potentials were referred to SCE electrode unless otherwise stated in this paper. The visible light source was obtained from 300-W Xe lamp with a 420 nm cut-off filter. The light intensity was set to 100 mW/cm² (1 sun). Electrochemical

impedance spectra (EIS) were recorded in the frequency range of 0.1–10⁵ Hz, and the photocurrent densities were measured with the bias potential in the range of –0.4 to 1.0 V.

2.3. PEC degradation experiments

PEC degradation experiments were carried out in a three-electrode system with C@TiO₂ as photoelectrode, and the active area was 2.0 × 2.0 cm² under illumination of the same light sources as the photoelectrochemical characterization experiments. A potentiostat model DJS-292C was employed to bias the C@TiO₂ working electrode. The PEC activities were estimated by the degradation of RhB (0.5 × 10^{–5} M) and 1-NP (1.0 × 10^{–4} M) solution, respectively. Prior to irradiation, the dye solution was stirred in the dark for 30 min to ensure the establishment of an adsorption/desorption equilibrium. At a

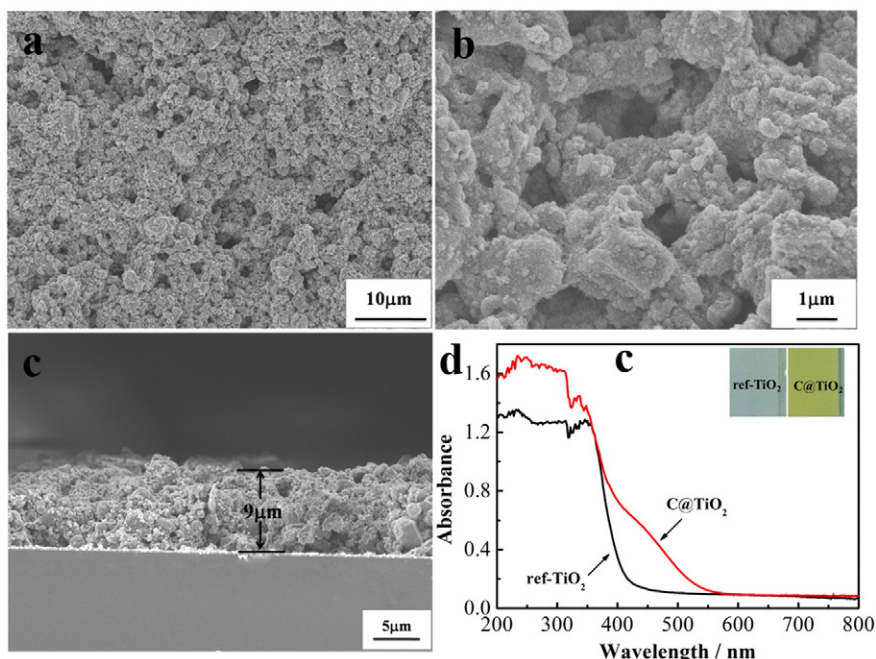


Fig. 2. Top-view (a & b) and cross-sectional (c) SEM images of C@TiO₂ films; (d) optical absorption spectra of ref-TiO₂ and C@TiO₂ films. The inset showing the color change of films.

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