

Original Research

Bionic titania coating carbon multi-layer material derived from natural leaf and its superior photocatalytic performance



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ABSTRACT

Bionic titania coating carbon multi-layer material was fabricated by employing canna leaves as substrate and carbon precursor. Titania nanocrystals were assembled and coated on the natural films. The carbonation treatment under pure N₂ atmosphere yielded the ultrathin multi-film hybrid material. The carbon layer was coated with small anatase titania crystallite (8–10 nm) and possessed a highly specific surface area of 248.3 m² g⁻¹. Examination using UV–visible spectrophotometer (UV–vis) showed that the band gap of the multi-layer material was reduced to 2.75 eV, and the hydrogen production by photocatalytic splitting of water under visible light irradiation was about 302 μmol g⁻¹ after six hour.

1. Introduction

In recent years, the photocatalytic processes, which utilized in waste water control, indoor air purifying, dye-sensitized solar cells and other applications, have attracted extensive interest among researchers [1]. Most especially, the photochemical splitting of water into H₂, identified by Fujishima and Honda in 1972, has emerged as an important reaction for human, because it could easily supply cheap energy [2]. As a common semiconducting oxide material, titanium dioxide (TiO₂) has been the subject of intensive research, because of its low cost, nontoxicity, fascinating chemical and physical properties [3–5]. However, one disadvantage of pure TiO₂ as a wide band-gap semiconductor (3.0 and 3.2 eV for rutile and anatase phases, respectively) is that it could absorb and be motivated only by ultraviolet (UV) light, which occupies about 3% of the solar energy [4]. The separation of photogenerated electrons and holes without recombination is another major drawback. A variety of methods have been investigated for improving the photocatalytic activity of modified TiO₂ in the visible light spectrum [6–10]. For example, the graphene-coupled TiO₂ showed higher activity compared with pure titania under visible-light emitting source, because the graphene sheets acted as an electron transfer bridge or electron sinks, which reduced the recombination rate of the electron-hole pairs [6]. The coupling between TiO₂ and carbon nanotubes was also found to enhance the ability of the photocatalyst to degrade organic molecules under visible light [7].

However, it remains a challenge to design and synthesize novel carbon nanostructures with uniform titania nanocrystal coating, which

possess sufficient photocatalytic activity. Huang and co-workers used natural cellulosic filter paper as scaffold and carbon precursor to prepare titania-coated carbon nanofibers, which has remarkably enhanced photocatalytic ability because of its highly nanoporous structure and synergistic effect [11]. Because of the abundance of unique structures in plants, carbonization of natural biotemplate under proper conditions would produce carbon material with novel structure. In this study, the bionic titania-coated carbon multi-layer material was achieved by coating canna leaves with thin titania layer, followed by calcination in pure nitrogen. The multi-layer structure, rich nanoporous surface and synergistic effect of hybrid material would remarkably enhance the photocatalytic ability under visible-light.

2. Experiment

Natural leaves were collected from canna plants. Tetrabutyl titanate (Aldrich), hydrochloric acid (35% HCl, Fisher) and ethyl alcohol (EtOH, Fisher) were used as the analytical reagents. Deionized water was purified using a milli-Q system. For the experiment 10 g canna leaves were soaked in a mixture of 10 mL hydrochloric acid, 500 mL ethyl alcohol and 490 mL millipore deionized water for 24 h, and this step was repeated two times. The soaked leaves were washed three times in deionized water, followed by being washed three times with EtOH and then dried at room temperature for 24 h. The pretreated canna leaves were then immersed in a solution of tetrabutyl titanate alcohol and water for 24 h and carbonized under a nitrogen atmosphere at a temperature of 700 °C. A comparison sample was prepared

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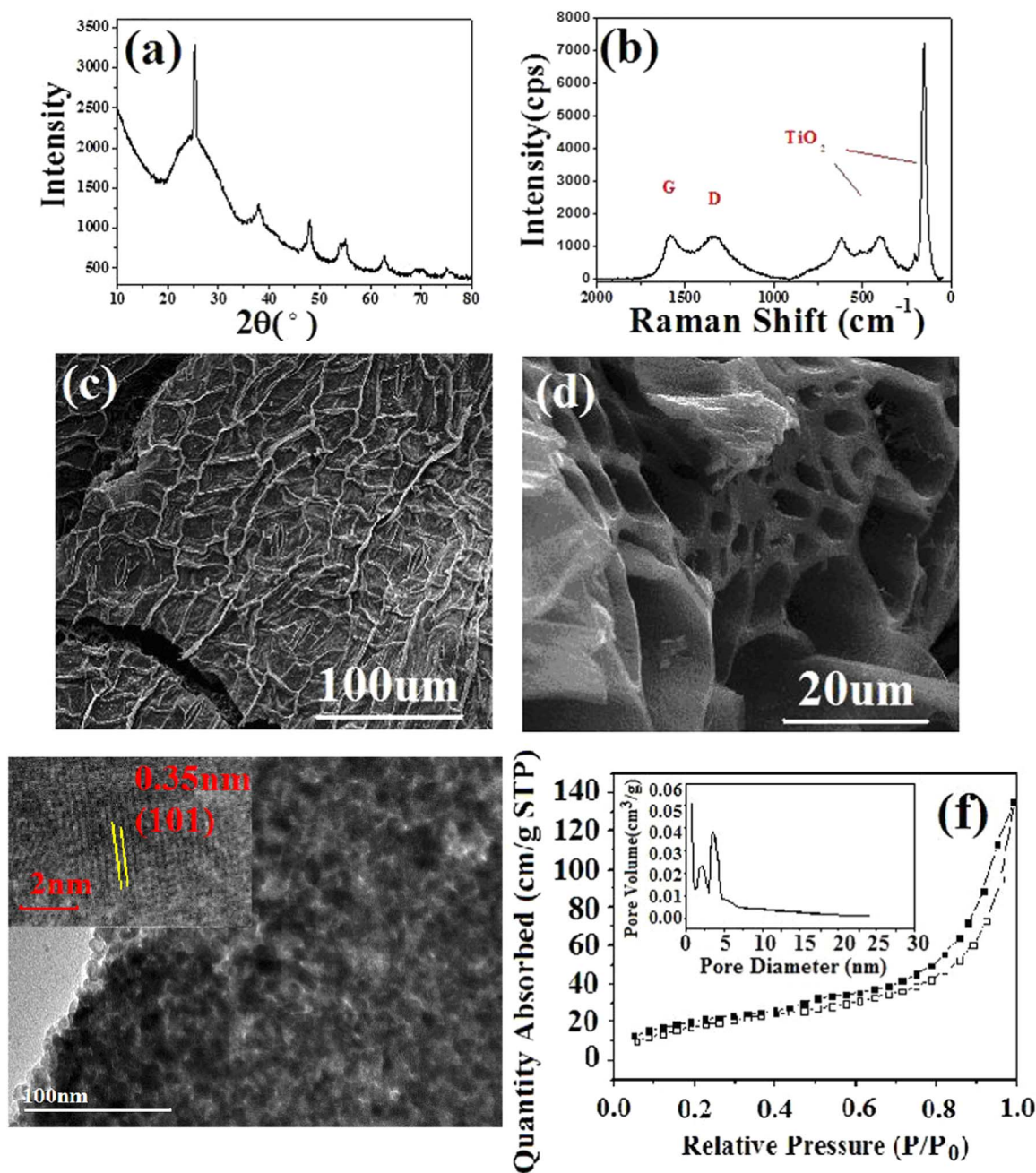


Fig. 1. The XRD pattern (a), Raman spectrum (b), SEM photos (c) and (d), TEM photo (e, inset image is HRTEM) and N_2 adsorption-desorption isotherms (f, inset curve is the pore size distribution) of sample heat-treated by nitrogen.

using the same immersed leaves calcinated under an air atmosphere.

Powder X-ray diffraction (XRD) measurements were taken on a Bruker D8 Advance X-ray diffractometer. The size and surface morphology were analyzed using a FEI Quanta 400 field emission scanning electron microscope (FESEM) and a JEM 2100F transmission electron microscope (TEM). Raman spectra were performed on a Thermo DXR, during which the samples were irradiated by a 532 nm laser beam. The textural properties of the samples were studied using a Micromeritics ASAP-2020 instrument. Optical absorption was recorded by a Puxitongyong T9 double beam UV-vis spectrophotometer in a wavelength range of 200–700 nm. The photocatalytic properties of the materials were reflected through the hydrogen production by photocatalytic splitting of water under visible light irradiation (deuterium lamp, 20 W).

3. Results and discussion

Fig. 1(a) showed the XRD pattern of sample heat-treated by nitrogen. Diffraction peaks were observed at $2\theta = 25.2^\circ, 36.9^\circ, 38.0^\circ, 53.9^\circ, 55.1^\circ$ and 62.8° , which were indexed to (101), (004), (200), (105), (211) and (204) reflection planes of anatase TiO_2 (JCPDS21-1272). A broad band within the 2θ range of $20\text{--}30^\circ$ might be attributed to the amorphous carbon yielded by carbonization of canna leaf [11]. Raman study (Fig. 1(b)) of the sample was performed to determine the existence of TiO_2 and carbon composite. The results showed that there was a significant appearance of D and G bands of carbon in the peak ratio at 1350 cm^{-1} and 1600 cm^{-1} , respectively. Furthermore, the broad peaks of Raman frequency observable at 638 cm^{-1} , 395 cm^{-1} and 151 cm^{-1} correspond to the E_g , A_{1g} and E_g energy levels respec-

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