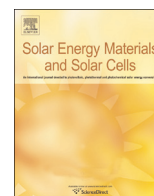




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Performance optimization of self-powered ultraviolet detectors based on photoelectrochemical reaction by utilizing dendriform titanium dioxide nanowires as photoanode

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ABSTRACT

Dendriform titanium dioxide nanowires (D-TiO₂ NWs) with high aspect ratio and density serve as a model architecture for high-performance self-powered ultraviolet (UV) detectors. The photo-sensing performance is characterized by integrating D-TiO₂ NWs into a photoelectrochemical cell. The short-circuit current density value of the D-TiO₂ NWs based UV detector is 176% and 46% higher than that of the bare TiO₂ NW film and TiO₂ nanocrystalline film since it offers excellent ultraviolet absorption, efficient charge carrier separation and fast electron transport. The incident power conversion efficiency can reach 14.5%, which promises a high sensitivity to the UV light. Under UV irradiation, the detector made of D-TiO₂ NWs exhibits a high responsivity of 0.61 A/W, a high on/off ratio of 1903 (incident power density 25 mW cm⁻²) and a fast response time of 5.9 ms. Combined with visible-blind characteristics and photosensitivity linearity in wide light intense range, the D-TiO₂ NWs based self-powered UV detector demonstrates promising application in various fields.

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1. Introduction

Ultraviolet (UV) detectors have been widely used in industrial and scientific applications, such as environmental monitoring, chemical analysis, remote control, memory storage and optoelectronic circuits [1–6]. Among various types of UV detectors, one-dimensional (1D) nanostructure UV detectors based on the photoconductivity have been proposed as excellent candidates, which exhibit high internal gain at room temperature [7–9]. However, their long recovery time [8,10] and surroundings-dependent behavior [11,12] are two major limitations for practical application. In addition, an external power source is usually required indispensably. Recently, self-powered photodetectors have attracted considerable attentions because the disappearance of batteries or external power source which can greatly enhance their adaptability and mobility [13], such as Schottky [14,15] and p–n junction type [3,10,16] and photoelectrochemical (PEC) type [17–21]. Among these self-powered UV detectors, PEC cell stands out competitively due to its plenty of advantages, such as low-cost, environment-friendly, simple fabrication process and abundant raw materials. Moreover, photoelectrochemical UV detector has fast

time response, high photosensitivity and good photosensitivity linearity in a large light intensity range [17,21].

In previous work, a PEC type self-powered UV detector based on TiO₂ nanocrystalline (NC) film has been demonstrated [17], which exhibited a power conversion efficiency (PCE) of 6.41%, a responsivity of 0.36 A/W and a fast decay time of 30 ms for short-circuit current density J_{sc} . The good performance of UV detector based on NC film is attributed to their large specific surface area. Gao et al. demonstrated a self-powered UV detector using ZnO nanos-trawberry aggregates structure which offers large specific area and excellent light absorption [19]. However, the nanoparticle films have high charge recombination loss caused by the electron trapping/scattering at grain boundaries [22]. Wang et al. has reported a self-powered UV detector based on a multilayer TiO₂ nanorod-assembled cloth/nanorod array electrode, exhibiting superior electron transport characteristics [18]. Cho et al. reported a hierarchically branched TiO₂ nanorod structure for efficient photoelectrochemical devices as it simultaneously offers an excellent light-trapping characteristics and a highly conductive path way for charge carrier collection [22]. 1D nanostructures (such as nanorod [23], branched nanorod [24] and nanotube [25]) can offer direct pathways for photogenerated electrons, but poor light absorption, low charge separation and collection efficiency [22] resulting from low density of texture and small interface area restrict their further application. Thus, it is highly

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desirable to design a nanostructure for PEC type UV detector which can combine the advantage of large interface area and efficient electronic transmission network.

Herein, we prepared a dense dendriform TiO_2 nanowire array (D-TiO_2 NWA) to serve as the photoanode of PEC type self-powered UV detectors. Dense nanowires (NWs) and linked branches form a compact interlaced film. This structure provides efficient transport network, excellent light absorption and large TiO_2 /electrolyte interface area, which are desired advantageous features for the PEC type UV detectors.

2. Experimental section

All chemicals were of analytical grade and used without further purification. As comparison, TiO_2 NC films were prepared according to previous work [17].

2.1. Preparation of the TiO_2 NWA

Before the growth of NWs, a TiO_2 polymeric sol described in previous work [26] was spin-coated on the Fluorine-doped tin oxide (FTO) glass substrates (2.2 mm in thickness, >90% transmittance, $14 \Omega/\text{sq}$, Nippon, Japan, ultrasonically cleaned with the mixed solution of deionized (DI) water, acetone and isopropanol for 15 min) and then annealed at 450°C for 1 h as the seed layer. In a typical synthesis process, 0.3 g titanium butoxide (Aldrich Chemicals, 97%) was added dropwise into a mixture of DI water (10 mL) and concentrated hydrochloric acid (14 mL, 38%). After stirring for 10 min, the solution was transferred into a Teflon-lined stainless steel auto-clave (50 mL capacity), then pieces of the seed-coated FTO substrates were placed into Teflon-liner. Hydrothermal reaction was carried out at 150°C for 24 h. After reaction, the autoclave was cooled to room temperature under flowing water. Then the samples were rinsed with DI water and absolute ethanol and annealed at 450°C for 1 h in air.

2.2. Preparation of the D-TiO_2 NWA

For the synthesis of the D-TiO_2 NW, the annealed bare TiO_2 NW was immersed in the aforementioned TiO_2 polymeric sol and annealed in air at 450°C for 1 h in order to form a thin layer of seed onto the TiO_2 NW's surface. Then the seeded TiO_2 NW was immersed in a Pyrex glass bottle with an aqueous solution consisting of DI water (10 mL, $18.2 \text{ M}\Omega$), HCl (0.1 mL), and TiCl_3 solution (0.1 mL, 20 wt% of TiCl_3 in H_2O and HCl solution, Sinopharm Chemical Reagent Co., Ltd., Shanghai, China), and then kept at 80°C for 30 min in an oven. The D-TiO_2 nanostructure has been formed after this process. Finally, the samples were rinsed with DI water and ethanol, and subsequently annealed at 450°C for 1 h in air.

2.3. Assembly of PEC type UV detectors

Assembly process of the self-powered UV detectors have been described in our previous work [17]. In brief, the sintered D-TiO_2 NWA electrodes and the platinized counterelectrodes were assembled into sandwich-type cells. The platinized electrodes were prepared by spin-coating a 4.5 mM isopropanol solution of $\text{H}_2\text{PtCl}_6 \cdot 6\text{H}_2\text{O}$, and then heated at 400°C for 20 min. The interelectrode space was filled with a liquid electrolyte consisting of LiI (0.1 M), 1,2-dimethyl-3-propylimidazolium iodide (0.6 M), I_2 (0.05 M) and 4-tert-butylpyridine (0.5 M) in acetonitrile.

2.4. Characterization

The morphology of the samples was characterized by field emission scanning electron microscope (FESEM, Hitachi S-4800) and transmission electron microscopy (TEM, FEI Tecnai F30). X-ray diffraction (XRD, Philips, X'pert pro, $\text{Cu K}\alpha$, 0.154056 nm) and micro-Raman spectroscopy (JY-HR800, YAG laser, 532 nm and spot diameter $\sim 600 \text{ nm}$) were used to analyze the phase and component. Photo-voltaic performance and electrochemical impedance spectroscopy (EIS) were performed by an electrochemical workstation (RST5200, Zhengzhou Shiruisi Instrument Technology Co., Ltd., China). EIS measurements were carried out in the frequency range

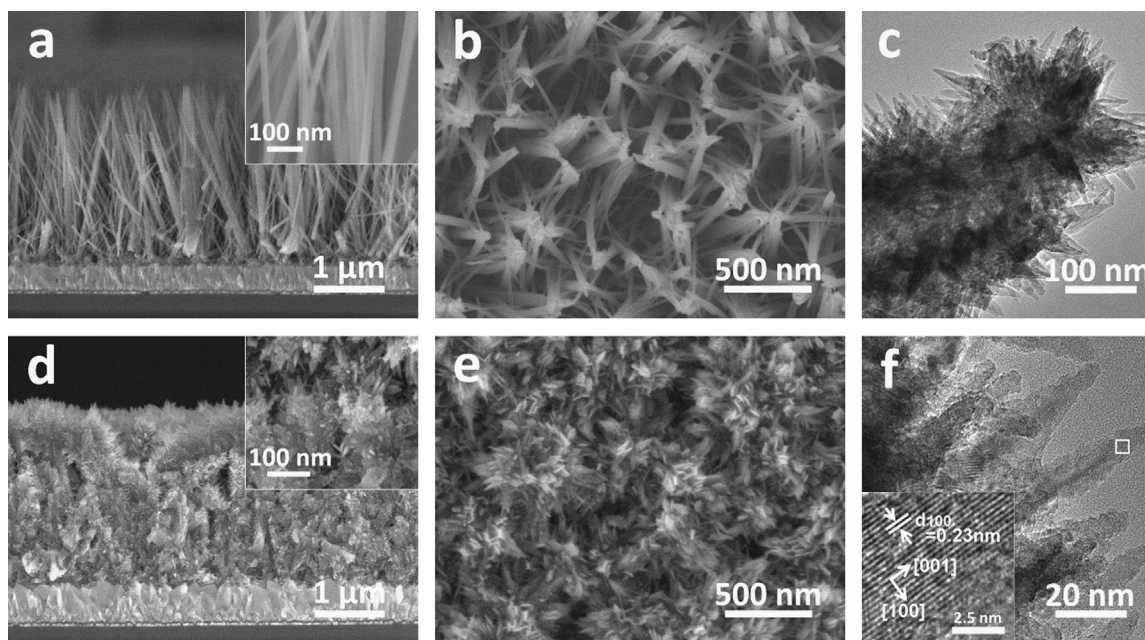


Fig. 1. SEM images of (a, b) oriented rutile TiO_2 nanowire array film and (d, e) dendriform TiO_2 nanowire array film grown on FTO substrate. (a, d) Cross-sectional view, (b, e) top view. The insets of a and d show high-magnification SEM images of TiO_2 NWA and D-TiO_2 NWA, respectively. (c, f) TEM image of discrete dendriform TiO_2 nanowire, the branches densely cover the surface of the NWs. The cone shape branches have an average length of 80 nm and a base diameter about 15 nm. The inset shows a HRTEM image of the rectangular region, indicating that the braches grow along the [001] direction.

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