



Short communication

Speedy photoelectric exchange of CdSe quantum dots/mesoporous titania composite system

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ABSTRACT

Two classes of important nano-materials, mesoporous titania (MT) and CdSe quantum dots, are prepared via chemical methods. Rapid immobilization of CdSe quantum dots into mesoporous titania has been achieved and the composite system is characterized by high-resolution transmission electron microscopy and photoluminescence. The band-to-band photoluminescence emission of CdSe quantum dots is blue-shifted for 20 nm after the dots are immobilized into mesoporous titania. Loading CdSe quantum dots into mesoporous titania leads to speedy photoelectric exchange process of the system and a significant rise in photoconductivity (for a factor of 8), which would attract great deal of attention in quantum dots solar cell.

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Mesoporous materials and semiconductor quantum dots (QDs) are two different classes of nano-materials, both being under intensive study for their potential usefulness in chemical catalysis, opto-electronics, biosensing, and biolabeling [1–3]. Most studies on QDs were focused on synthesis as well as the quantum confinement and surface effects. QDs prepared by chemical methods bear the merits of simplicity, low cost, possibility of large scale of production, as well as availability for a large number of materials. The major problem met in the application of quantum dots grown in liquids is that the dots are randomly distributed and movable, which significantly limits their application. It is essential to immobilize the QDs, so that one can trace the change of a given quantum dot having experienced some process. Currently, immobilization of QDs has become a research hotspot [4]. Macropore templates have a typical pore size of 50 nm or larger, in contrast to the QDs diameters typically around 5 nm. Mesoporous materials have ordered nanopores structure in the size range of 2–50 nm and, more importantly, large and accessible pore surface [5,6]. Mesoporous materials are therefore the excellent candidates for immobilizing QDs. Incorporation of quantum dots in pores of mesoporous materials could lead

to fabrication of quantum-dot arrays and composites with various functions.

Recently, mesoporous titania (MT) has attracted great deal of attention due to its strong oxidizing and reducing ability under UV light irradiation. The material has been used as solar cell electrodes and photocatalyst, and used to fabricate devices including gas-sensors and electrochromic displays [7–11]. Compared with mesoporous SiO₂, MT is difficult to prepare since the material has a strong tendency to precipitate and crystallize. The crystallization of TiO₂ often leads to collapse and destruction of the mesostructures during the removal of the organic template supporting the TiO₂ mesostructure framework. Current works that use mesoporous materials as templates to immobilize QDs are focused on *in situ* QDs synthesis [12,13], which often leads to low photoelectric exchange efficiencies and significant problems related to stability of the materials. Here we report the CdSe QDs immobilization in MT via a simple and safe impregnation at room temperature and investigation on the photoconductive property of the QDs. Such composite system forms a good heterojunction, which could be useful for the development of solar cells with improved efficiency and photo-catalytic materials.

The MT was synthesized by sol-gel reaction of tetrabutyl titanate (Ti(OBu)ⁿ₄, TBOT) in the presence of triblock copolymer surfactant of pluronic P123. P123 was kindly donated by BASF with a molar weight of 5750. TBOT, acetylacetone (AcAc), concentrated hydrochloric acid, and absolute ethanol were purchased commercially without further purification prior to use. 4.3 g P123 was firstly dissolved in 50 ml ethanol absolute and 0.6 ml 1 M

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hydrochloric acid solution was added under vigorous stirring for 3 h. Separately, 10.2 ml TBOT and 1.5 ml AcAc were mixed by stirring for 0.5 h, and then added to P123 and ethanol solution. After being placed at room temperature for 0.5 h, 16 ml distilled water was added into the mixed solution. Subsequently, the solution was aged with vigorous stirring at room temperature for nearly 6 h and then aged at 40 °C until it became gelatin. As-synthesized gelatin was calcined at a rate of 1 °C/min to 150 °C and kept for 2 h. The temperature was then raised to 340 °C and preserved at this temperature for 4 h to remove the P123 template entirely and get the MT powder. CdSe quantum dots were synthesized following Ref. [14]. The growth times of the QDs were chosen to be 30, 90, 240, and 600 s, corresponding to QD sizes of 2.9, 3.6, 4.1, and 4.2 nm, respectively. Loading QDs into MT were accomplished by mixing MT with a controlled amount of CdSe QDs in organic solvent (toluene) under ultrasonic process. The loading process was completed in about 60 min, until there is no free CdSe dots left in solution, as evidenced by UV–vis absorption measurement. The CdSe-QDs-doped MT (MT:CdSe-QDs) was then washed five times with pure toluene.

To perform current–voltage (I – V) and current–time (I – T) measurements, MT, CdSe QDs, or MT:CdSe-QDs was mixed with poly(vinyl butyral) solution (about 10 mg MT and 1 mg poly(vinyl butyral) in 1 ml DMF) and films were prepared by coating the mixtures on ITO glass. The films were then dried in vacuum oven for 16 h. ITO on glass is used as electrodes with two electrodes being separated by a gap of 0.2 mm in width and 10 mm in length. Keithley 4200 semiconductor characterization system was used to measure the current–voltage and current–time characteristics of the samples, and a 250 W tungsten lamp (light spectrum from 340 to 2500 nm) was used as the excitation light source. In the I – V measurements, the distance between the light source and the film was kept at 15 cm and the time to scan the voltage from –20 to

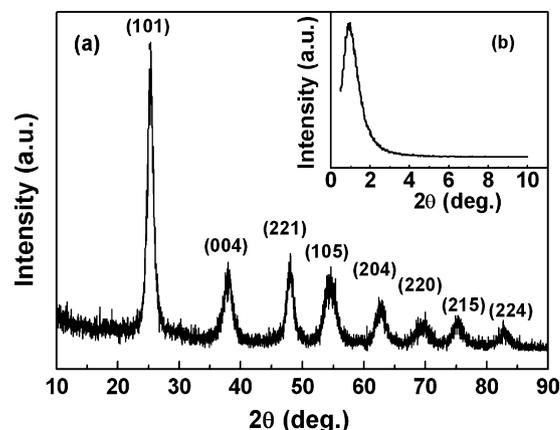


Fig. 1. High-angle X-ray diffraction curve (a) and low-angle X-ray diffraction curve (b) of mesoporous titania.

20 V was 6 s. Note that the quick scanning is to avoid heating the samples.

The power X-ray diffraction (XRD) patterns of mesoporous titania were examined by a Rigaku D/max 2550 diffractometer using a high power Cu $K\alpha$ source operating at 40 kV and 200 mA. Fig. 1(a) and (b) presents XRD curves of mesoporous titania and small-angle XRD, respectively. The XRD patterns in Fig. 1(a) clearly clarify the presence of nanocrystalline anatase framework. From the (1 0 1) diffraction peak, the average crystallite size of 7.6 nm was calculated using Scherrer equation. In Fig. 1(b), the low-angle XRD curve of the mesoporous titania shows a sharp peak at $2\theta = 0.93^\circ$, corresponding to the well-organized mesostructure.

Fig. 2 presents high-resolution transmission electron microscopic (HR-TEM) images of MT:CdSe-QDs obtained using Japan

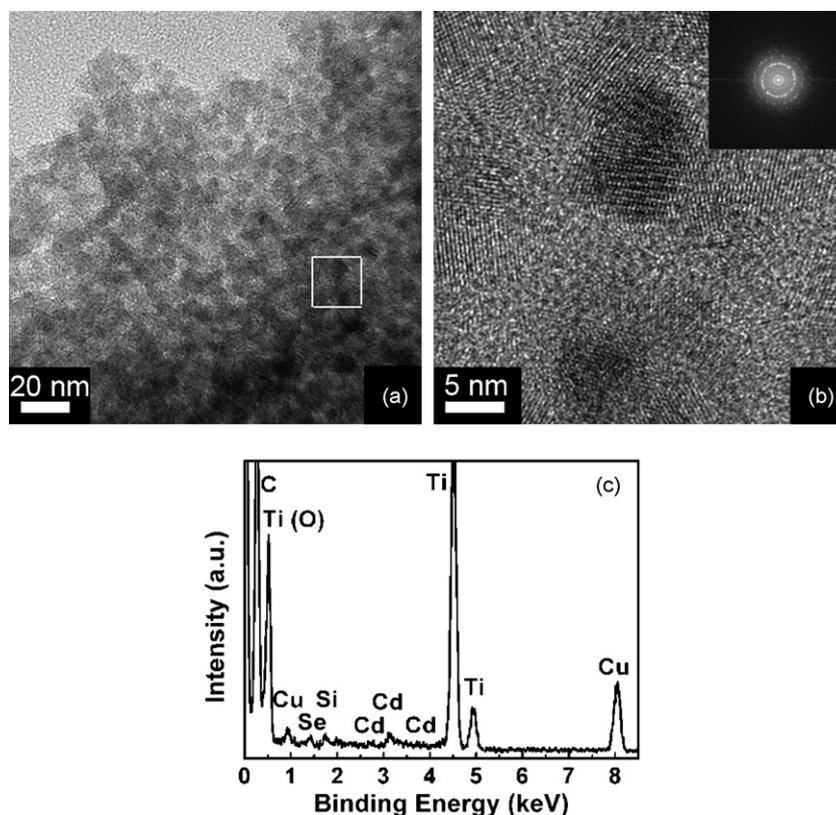


Fig. 2. HR-TEM (a) and (b) and EDS (c) of the QDs-immobilized mesoporous titania.

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