



The optoelectronic properties and applications of solution-processable titanium oxide nanoparticles

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ABSTRACT

In this study, the surface modified TiO₂ nanoparticles have been prepared through microwave-assisted reaction with oleic acid as surfactant. The as-synthesized TiO₂ nanoparticles reveal uniform particle size distribution and high dispersibility in organic solvents which can be easily integrated into solution-process. The organic capped TiO₂ (OC-TiO₂) has been applied as filler for LED encapsulant and electron collection layer for organic solar cells. With 1.0 wt% loading of TiO₂ nanoparticles, the refractive index (RI) of TiO₂/silicone composites increase from 1.51 for neat silicone to 1.575. As a result, a high-power LED encapsulated with this composite showed more than 9% increase in the light output. Moreover, the organic solar cell with the OC-TiO₂ as electron collection layer shows an enhanced long-term durability due to the hydrophobic property of OC-TiO₂.

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1. Introduction

Metal oxides (MOs) have been attracted much attention due to their unique electronic, magnetic and optical properties. The MOs can be easily deposited as a thin film to exhibit good physical adhesion, transparency in the visible range and superior stability. These properties drive them as a promise material to be integrated into optoelectronic devices. Most importantly, the MO reveals much environmentally stable compared with other organic materials. The unique optoelectronic and stable properties of MOs

make them suitable for applying as filler for polymer based composites.

Nanocrystalline TiO₂ is one of the most studied MOs, due to its widespread applications in photocatalysis [1–3], solar cell devices [4,5], sensors [6] and filler for encapsulant with high refractive index [7–10]. The most common methods to TiO₂ crystals mainly comprise sol–gel process, hydrothermal route and surfactant-assisted methods. Unfortunately, these templates typically provide insoluble TiO₂ with diameter ranged from a few tens to several tens. The insolubility of TiO₂ is a critical obstacle to the fabrication of thin films with smooth surface. To prepare well-dispersed TiO₂ suspension, the physical grinding approach has been demonstrated. The smooth and transparent TiO₂ films have been successfully produced and integrated into organic optoelectronics via the grinding route [11,12]. However, the hydrophilic nature of TiO₂ is not compatible with organic materials. The homogeneous distribution of TiO₂ within

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polymer is still challenging owe to the incompatibility between TiO_2 and organic media. The aggregated TiO_2 nanoparticles in the composites tend to decrease the mechanical, optical properties and limit its load-bearing applications. Recently, grafting polymer chains onto nanoparticles to shield the surface has proved to be one of the most promising strategies to overcome this problem [13]. The grafted TiO_2 nanoparticles have been applied for filler of high RI encapsulant [7], high-k dielectrics for organic thin-film transistor [14–17] and electron transfer material for solar cells [18]. The prepared nanocomposites can be processed into highly transparent coatings and self-standing films. The developed encapsulant showed a tunable RI by varying the loading fraction of TiO_2 . Moreover, the grafted TiO_2 still can exhibit high electron mobility despite the surface was covered with polymer chains. This makes it possible to fabricate the solar cells based on grafted TiO_2 and conjugated polymers [19]. Although the grafted TiO_2 nanoparticle exhibits excellent compatibility and processability with organic media, the synthesis and purification were complicated with multistep procedures. The grafting density [20,21], structure and molecular weight of the macromolecules anchored onto the particle surface are also additional variation should be concerned. It is still a challenge to directly synthesize soluble TiO_2 nanoparticles applying for preparation of polymer based composite films.

In this study, we prepared solution-processable TiO_2 nanoparticle through microwave-assisted reaction. With oleic acid as surfactant, the surface property of the TiO_2 can be easily modified under the microwave-assisted condition in a short period of reaction time. The as-prepared TiO_2 reveal excellent solubility and processability in common organic solvents such as chloroform, toluene and 1,2-dichlorobenzene. As-synthesized OC- TiO_2 nanoparticles were incorporated into silicone based encapsulant to achieve TiO_2 /silicone nanocomposites with enhanced optical and conductive properties. Moreover, the X-ray photoelectron spectrometry (XPS)/UPS spectra indicate that the work function of the surface modified TiO_2 is unchanged. To demonstrate the potential use of OC- TiO_2 as hole blocking layer, the organic solar cells having an inverted structure were also fabricated.

2. Experimental

2.1. Preparation of OC- TiO_2

In a typical synthesis, 10 mmol of titanium tetraisopropoxide was added to 35 g of degassed oleic acid and allowed to stir for 5 min. A 10 mmol of triethylamine in 6.4 g of ethyleneglycol was subsequently added. Then, the solution was mixed with benzyl alcohol (vol. 1:1). Subsequently, the solution was maintained in a close system at 150 °C for 45 min through microwave-assisted reaction.

2.2. Preparation of OC- TiO_2 /silicone encapsulant

The as-prepared TiO_2 solution (toluene) was added directly into the OE-6550A under stirring for 30 min. Then, a rotary evaporator was used to remove the solvent within

OE-6550A. Consequently, the mixture was blended with OE-6550B until a homogeneous mixture was obtained. The mixture was vacuum vented until the bubbles exploded and the mixture was clear and transparent. The mixture was poured into a stainless steel mold and heated in an oven for 2 h at 150 °C. After this curing process, the mold was taken out of the oven and the sample was removed from the mold.

2.3. Fabrication of organic solar cell

The polymer solar cells in this study consists of a layer of poly(3-hexylthiophene) (P3HT) and [6,6]-phenyl-C61-butyric acid methyl ester (PCBM) blend thin film sandwiched between transparent anode indium tin oxide (ITO) and metal cathode. A modified ITO surface was obtained by spin-coating a layer of OC- TiO_2 . After baking at 100 °C for 1 h, the substrates were then transferred into a nitrogen-filled glove box. The devices were fabricated by spin-coating blend of P3HT:PCBM on the OC- TiO_2 modified ITO surface. A 20 and 100 nm thick of MoO_3 and aluminum was thermally evaporated under vacuum at a pressure below 6×10^{-6} Torr thorough a shadow mask.

2.4. Characterization

The crystalline structure of the TiO_2 was studied by XRD (Philips X'Pert/MPD). The morphologies of the TiO_2 were investigated using TEM (Hitachi S-4700). Chemical composition was measured by FTIR spectrum. The UV–vis spectra of the TiO_2 solutions were obtained using a Jasco-V-670 UV–Vis spectrophotometer. Particle sizes and zeta potentials were measured using a particle size analyzer (Brookhaven 90 Plus Sn11408). The surface morphologies of the TiO_2 films were investigated using atomic force microscope (AFM, Digital Instrument NS 3a controller equipped with a D3100 stage). The RI of the hybrid materials was determined by an Abbe-refractometer (model:WAY). The water vapor transmission rate (WVTR) was obtained from a commercial PERME-W3/330 instrument. The barrier property was measured under 40 °C and the relative humidity was controlled at 90%. XPS/UPS spectra were recorded using a PHI 5000 VersaProbe (ULVAC-PHI, Chigasaki, Japan) system. Current–voltage (J – V) characteristics were measured in the glove box under nitrogen atmosphere with simulated AM 1.5G irradiation at 100 mW/cm² using a xenon lamp based solar simulator (Thermal Oriel 1000 W).

3. Results and discussion

The crystalline structure of the as-synthesized TiO_2 is studied by XRD. As shown in Fig. 1a, all the peaks can be readily indexed to the pure anatase phase which is in good agreement with the reported values. No characteristic peaks of other impurities were found. The optical property of the OC- TiO_2 was also investigated through measuring its UV–vis spectrum as shown in Fig. 1b. The onset of the UV–vis spectrum of TiO_2 is around 390 nm leading to a band-gap of 3.2 eV. This value is very close to the previous report suggests that the capped molecular has no effect on the

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