



Nanocrystal V_2O_5 thin film as hole-extraction layer in normal architecture organic solar cells

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ABSTRACT

Nanocrystal V_2O_5 dispersion processed thin films are introduced as efficient hole extraction interlayer in normal architecture P3HT:PCBM solar cells. Both thin and rather thick interlayers are studied and demonstrated to work properly in organic photovoltaic. Nanocrystal V_2O_5/V_2O_5 layers effectively block electrons and effectively extract holes at the ITO anode. Very constant and high V_{OC} (above 0.56 V) are easily achieved. Comparable J_{SC} and PCE are demonstrated for nanocrystal dispersion-processed devices when compared with amorphous sol-gel processed devices. The excellent functionality of nanocrystal V_2O_5 interlayers in Si-PCPDTBT:PCBM devices further demonstrates the broad application potential of this material class for photovoltaic applications.

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1. Introduction

Photovoltaic (PV) is considered a promising technology to overcome the energy resource crisis [1], in which bulk-heterojunction (BHJ) organic photovoltaic (OPV) has attracted tremendous interests [1–3] due to advantages such as low manufacturing costs, high production throughput, large area and facile shape processing. Recently, high efficiencies greater than 10% have been reported for single junction OPV devices [4,5]. Up to date, poly(3-hexylthiophene) (P3HT) and [6,6]-phenyl-C61-butyric acid methyl ester ([60]PCBM) blends are the most popular active composite model for OPV studies, where poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) has been most widely used as interface layer between the active layer and electrode [3]. However, PEDOT:PSS can cause substantial degradation in standard architecture solar cells due to its intrinsic acid and hygroscopic properties [6,7]. To solve this problem, solution-processed transition metal oxides such as NiO [8], MoO_3 [9], WO_3 [10], and

V_2O_5 [11–14] have been studied and demonstrated to be promising alternatives in the past few years. Especially, solution-processed vanadium oxide as interlayer can effectively prevent the leakage current at the organic/electrode interface by blocking electrons at the anode contact in either inverted [13,14] or normal [12] architecture. Promising device performance has been demonstrated [11–15] due to its intrinsic optical and electronic properties [16–18]. Compared with thermal vacuum-deposited [19,20] layers, low temperature coating solution-processed V_2O_5 interlayers are more facile and favorable for mass production, which is crucial for the target of low-costs and mass production like “roll-to-roll” printed thin film solar cells.

In 2009, spin-of vanadium oxide dispersions was reported to give anode interlayers for inverted polymer solar cells hybridized with ZnO nanorods by Huang et al. [14]. It was reported that the power conversion efficiency of the P3HT:PCBM cells fabricated with this vanadium layer was improved from 2.5% to 3.56%. In 2012, spin-coating V_2O_5 nanobelt-buffered polymer-fullerene solar cells was studied, and a PCE of 2.71% was achieved [11] with normal architecture devices. Differently, sol-gel-processed amorphous V_2O_5 layers based on a precursor of vanadium (V)

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oxitriisopropoxide, have been reported both in normal [12] and inverted [13] structure in 2011. Cell performance with PCE 2.4–3%, V_{OC} 0.52–0.53 V, J_{SC} 8.4–9.6 mA cm⁻², and FF 55.6–58.9% was achieved in normal structured P3HT:PCBM devices by Zilberberg et al. [12]. In the inverted architecture, a performance of PCE 3.9%, V_{OC} 0.57 V, J_{SC} 10.1 mA cm⁻² and FF 67% was reported for P3HT devices, and a PCE 5.0%, V_{OC} 0.82 V, J_{SC} 11.6 mA cm⁻² and FF 0.53 for a-PTPTBT based devices [13].

Despite the considerable amount of work already performed on V₂O₅, little is known about nanocrystal dispersion-processed V₂O₅ thin films as electrode buffer layers in organic devices. Commercial V₂O₅ powder (ordered from Aldrich - 223794, >98%) was tested for Ref. [11], but only very weak performance (1.08% of PCE) in P3HT:PCBM based normal devices has been reported. Nevertheless, such systems would be of specific interest, since they may offer a pathway towards physically thick film with superior mechanical stability and electronic conductivity. Previous studies reported on a proper and optimum thickness of an electrode buffer layer of a few tens of nanometers, for PEDOT:PSS as well as metal oxides [21]. But in a view of durability and reliability, a relatively thick electrode buffer layer may be desirable, especially for thin film photovoltaic products with an active layer in the size of 100 nm.

In this work, crystal V₂O₅ nano particles were synthesized by hydrolysis of precursor vanadium (III) acetylacetae ($V(C_5H_7O_2)_3$). Based on the prepared nanocrystal V₂O₅ particles, dispersion-processed V₂O₅ nanoparticle (V₂O₅-P) layers were investigated as anode buffer layer in normal structure devices with organic blend P3HT:[60]PCBM. Further it is demonstrated that a relative thick V₂O₅-P layer can still work properly for OPV devices. Sol-gel processed VOx (VOx-S) buffer layers (from vanadium (V) oxitriisopropoxide) in a same device structure were studied for comparison and benchmarked with additional organic blend composites based on poly[(4,40-bis(2-ethylhexyl)dithieno[3,2-b:20,30-d]silole)-2,6-diyl-alt-(4,7-bis(2-thienyl)-2,1,3-benzothiadiazole)-5,50-diyl] (Si-PCPDTBT) [22]:[70]PCBM.

2. Experiments

Nanocrystal V₂O₅ particles were synthesized with a revised hydrolysis method according to literature [23]. In detail, 0.766 g vanadium (III) acetylacetae ($V(C_5H_7O_2)_3$) was dissolved in 200 ml ethanol (99.9%) in a flask, till a transparent solvent was obtained under agitation. Then 40 ml mixture of ammonium hydroxide solution (25% NH₃ in H₂O) and ethanol (1:4 v/v) was added at a speed of 1 drop/s. Precipitates appeared after about 20 h agitation, and were collected by centrifugation and washed with water/ethanol. The precipitates were crystallized at 500 °C for 2 h in atmosphere condition.

The architecture of ITO/V₂O₅/P3HT (or Si-PCPDTBT):PCBM/Al (or Ca/Ag) was used to fabricate OPV devices (Fig. 1A). The devices were processed in ambient atmosphere. Pre-structured ITO coated glass substrates (as obtained from Osram) were subsequently cleaned in acetone and isopropyl alcohol for 10 min each. After drying, the substrates were coated with a hole-extraction layer, either with the dispersion of

V₂O₅ nano particles in ethanol (3.3 mg/ml, ultrasonic treated) or with the liquid of vanadium (V) oxitriisopropoxide sol (ordered from Sigma, mixed with isopropanol at ratio of 1:70 v/v), and dried at 110 °C for 10 min. The absolute value for the work function of V₂O₅ on ITO substrate is determined by referenced to highly ordered pyrolytic graphite (HOPG) at -4.65 eV [24] through a Kelvin Probe measurement (SKP5050, Kelvin Probe Technology Inc.). P3HT (purchased from Merck) and [60]PCBM (purchased from Solenne) (1:1 in wt.%) were blended in chlorobenzene at a concentration of 2 wt.%. Si-PCPDTBT (provided by Konarka) and [70]PCBM (1:1.5 in wt.%) were blended in ortho dichlorobenzene at a concentration of 2 wt.%. The active layer was deposited by doctor-blading on top of the hole-extraction layer for all devices. The P3HT:[60]PCBM devices were annealed at 140 °C for 5 min on a hot plate after evaporation of a 100 nm thick Al electrode. UV-treatment was carried out by a 100 W UV irradiation for 10 s in a UVACUBE 100. Series resistance (R_s) and shunt resistance are defined and determined from the dark J - V curves near 1.5 and 0 V respectively. A Ca/Ag (20 nm/100 nm) cathode electrode was vacuum-thermal evaporated on top of the Si-PCPDTBT:[70]PCBM active layer, and without any other treatment step after evaporation. The active area of the investigated devices was 10.4 mm². Current density-voltage (J - V) characteristics were measured with a source measurement unit from BoTest. Illumination was provided by an OriolSol 1A Solarsimulator with AM1.5G spectra at 0.1 W/cm².

3. Results and discussion

XRD pattern of the prepared V₂O₅ nanocrystals is shown in Fig. 2A. Peaks were indexed according to the orthogonal symmetry of V₂O₅ (space group: Pmmn (59), $a = 11.516$, $b = 3.566$, $c = 3.777$ Å; JCPDS card No. 41-1426), and demonstrates the crystalline phase of our V₂O₅. Transmittance of the V₂O₅-P layers (ca. 22 nm and ca. 210 nm) on ITO substrate were studied by a UV-Visible spectrometer equipped with an integrating sphere. Transmittance of ca. 95% and ca. 83% (ITO substrate background subtracted) were obtained for 22 and 210 nm films respectively in the visible regime (Fig. 2B). Polydisperse nanocrystals of V₂O₅ with particle size smaller than 30 nm were confirmed by transmission electron microscopy (TEM) (Fig. 2B, insert). With these polydisperse V₂O₅ nanocrystals, we believe that a V₂O₅-P layer with the tiny particle at the bottom and bigger particles on top can be obtained (Fig. 1A), with which the following advantages could be expected. The big particles on top can effectively collect the generated holes due to its high conductivity. And the tiny particles at the bottom can form a thin compact layer on top of ITO, which can effectively block the electrons and avoid current leakage. This supposition is confirmed by the atomic force microscopy (AFM) image of the 1-coating working film (Fig. 2C). The electronic functionality of which in device is characterized and discussed below. Fig. 2D and E show the surface morphologies of the V₂O₅-P processed films (ca. 22 nm and 210 nm) on ITO substrate tested by AFM. A roughness of ca. 4.5 nm and ca. 5.7 nm was measured respectively. A high conductivity of ca. 1.3×10^{-4} S/cm of crystal V₂O₅-P film on a glass sub-

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