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Contents lists available at ScienceDirect

Solar Energy Materials & Solar Cells

journal homepage: www.elsevier.com/locate/solmat

Brush painted V_2O_5 hole transport layer for efficient and air-stable polymer solar cells

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ARTICLE INFO

Article history:

Received 17 April 2014

Received in revised form

8 August 2014

Accepted 18 August 2014

Keywords:

Polymer solar cells

Hole transport layer

V_2O_5

Brush painting

ABSTRACT

As an alternative hole transport layer (HTL) to poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS), we investigated V_2O_5 thin films fabricated by simple brush painting without any post-treatments. Polymer solar cells (PSCs) with V_2O_5 fabricated by brush painting showed superior performance to that of conventional PEDOT:PSS based system; in particular, higher FF was obtained. Because more hydrophobic oxide surface induces better physical contact and better wetting of active blends, lower R_s is obtained; thus, the resulting device shows higher performance. PSC with brush painted V_2O_5 at 50 °C using 0.99 vol% vanadium (V) oxytriisopropoxide in isopropyl alcohol exhibited the best power conversion efficiency (PCE) of 3.83%. Furthermore, employment of V_2O_5 as HTL leads to significant improvement in long-term stability in air (dropping of PCE of conventional PEDOT:PSS system to ~0% and retention of PCE of V_2O_5 based PSC to over 60% after exposure in air for 96 h without any encapsulation).

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1. Introduction

PSCs have attracted tremendous attention due to their excellent potential of low temperature and low cost solution process, light weight, large area fabrication, and flexibility [1,2]. PCEs of PSCs have rapidly increased through the development of new polymers, morphology control of active layer, and improvement of device structure, hence they have been recently reported up to ~10% [3–6]. In general PSCs, active layer, which consists of a conjugated polymer and fullerene derivatives, is sandwiched between transparent anode and metal cathode such as Ca/Al and Li/Al. In this case, insertion of suitable hole transport layers (HTLs) between anode and active layer is required to restrict direct contact of acceptor material with hole collecting transparent anode, resulting in large leakage current. In other words, HTLs block the photo-generated electrons and induce efficient hole transfer to anode. Poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) is commonly used as HTL in PSCs to facilitate hole collection due to its high conductivity, high work function, and good optical transparency [7,8]. However, water soluble PEDOT:PSS is sensitive to moisture and can corrode indium thin oxide (ITO) because of its highly acidic property. In addition, electrical inhomogeneity,

resulting from formation of inhomogeneous film morphology, causes serious problem in the reliability of devices [9–11].

Therefore, it is necessary to replace PEDOT:PSS for commercialization of PSCs, and as alternative materials, interest in two-dimensional carbon nanostructures such as graphene and graphene oxide (GO) and n-type transition metal oxides such as molybdenum tri-oxide (MoO_3), tungsten tri-oxide (WO_3) and vanadium pent-oxide (V_2O_5) has been increased [12–21]. At the initial stage of metal oxide based PSCs, to demonstrate high efficiency PSCs, metal oxides were usually fabricated by expensive and complicated vacuum processes. Although, very recently, reports on PSCs containing solution processed oxide HTLs fabricated by sol-gel method or spin-coating of synthesized nanoparticle have been published, spin-coating is known to be unsuitable for the roll-to-roll process and large area devices [22–28]. One of the urgently demanded issues for commercialization of low cost PSCs, showing continual enhancement of efficiencies, is development of cost-efficient printing technologies for each component in PSCs. Demonstrations of highly efficient PSCs containing printed photoactive layers or transparent electrodes have been reported by many groups, while only few studies on printed metal oxide HTLs have been published by several groups [29,30]. Because electrical properties of oxide HTLs, determining selective carrier transport, are strongly dependent on the thickness and surface morphology of oxide, optimization of printing process of oxide HTLs may be difficult.

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In this study, we demonstrate PSCs with V_2O_5 fabricated by simple brush painting. With diverse printing processes such as screen printing, spray process, and inkjet printing, brush painting is also very compatible process with roll-to-roll based mass production to achieve ideal coating on a variety of substrates, even on non-flat substrate. In addition, because direct brush painting of limited solution on thermally controlled substrate induces fast solidification of film, critical control of film thickness is possible. Several groups including our group have reported PSCs with brush painted active layer or metal nanowire/conducting polymer hybrid electrode [31–34]. To our knowledge, this is the first report demonstrating facile fabrication of oxide HTL via direct brush painting. Electrical and optical characterization of V_2O_5 layers and their effect on the performance of PSCs were investigated.

2. Experimental

As the first step in the preparation of HTLs, indium tin oxide (ITO, Samsung Corning Co, Ltd.) coated glasses were cleaned in deionized water, acetone, and isopropyl alcohol under ultrasonication and then dried in an 100 °C oven for 30 min. Subsequently, cleaned ITO surfaces were treated with UV-O3 plasma for 20 min to improve wettability of ITO. For the use of V_2O_5 HTLs fabricated by brush painting, vanadium (V) oxytriisopropoxide (Sigma-Aldrich) was mixed in isopropyl alcohol. Here, the concentration of solution, which affects thickness and morphology of film, was controlled to 0.25 vol%, 0.99 vol%, 4.95 vol%, 9.09 vol%, and 45.45 vol%. Additionally, considering boiling point of the solvent, brush painting was performed at varied temperatures (50 °C, 80 °C, and 110 °C). Also for comparison, V_2O_5 HTL was fabricated by spin coating at 3000 rpm for 40 s using optimized precursor solution (0.25 vol% in isopropyl alcohol). For the reference device structure, PEDOT:PSS was spin-coated at 5000 rpm onto the UV-O3 treated ITO-glass substrates followed by annealing at 120 °C for 10 min. After that, the active layer was spin-coated onto HTLs at 700 rpm for 60 s/1500 rpm for 5 s using blend of 0.02 g P3HT and 0.02 g PCBM in 1 ml 1,2-dichlorobenzene (DCB). Then, to induce self-organization of active layer, samples were kept in a jar for 1 h in nitrogen filled glove box, followed by thermal annealing at 110 °C for 10 min. Finally, metal electrode consisting of LiF/Al (0.7 nm/80 nm) with an area of $\sim 4.64 \text{ mm}^2$ was deposited in vacuum at a pressure of 10^{-7} Torr.

Current density–voltage (J – V) characteristic was measured using a Keithley 2400 equipment under 1 sun and AM 1.5 G filter. A reference Si solar cell certified by the International System of Units (SI) (SRC-1000-TC-KG5-N, VLSI Standards, Inc) was used for accurate measurement. The film morphologies of V_2O_5 HTLs were observed using a field emission scanning electron microscope (FE-SEM, Hitachi S-4800). The optical transparency of V_2O_5 HTLs was measured using a UV–vis spectrophotometer (Varian AU/DMS-100S) and the changes in the ITO work-functions by the use of V_2O_5 HTLs were measured using ultraviolet photoelectron spectroscopy (UPS, ESCALAB 210) with a He I (21.2 eV) source. The chemical characterizing of the V_2O_5 layer was performed using X-ray photoemission spectroscopy (XPS, ESCALAB 210). Furthermore, the surface wetting angle of various HTLs on ITO was measured with deionized water as test materials. Also, efficiency was recorded as a function of exposed time in air to analyze the effect of HTLs on long-term stability of devices.

3. Results and discussion

As mentioned, many groups have reported on the PSCs fabricated by various printing processes, but most of the studies focused on the fabrication of active layer or electrodes with

~ 100 – 200 nm thickness. In case of interfacial layer, especially oxide layer, which has emerged as a promising selective charge transporting layer for improvement of organic based devices, because thickness of layer critically affects electrical property, control of film thickness to a few nm–a few tens of nm is very important. Thus, it is difficult to demonstrate highly efficient PSCs with printed oxide buffer layers. As shown in Fig. 1, because fast

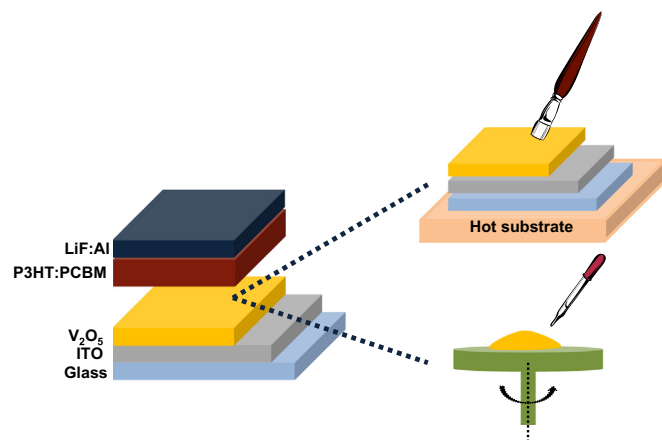


Fig. 1. Schematics of PSCs and methods for the fabrication of V_2O_5 HTLs.

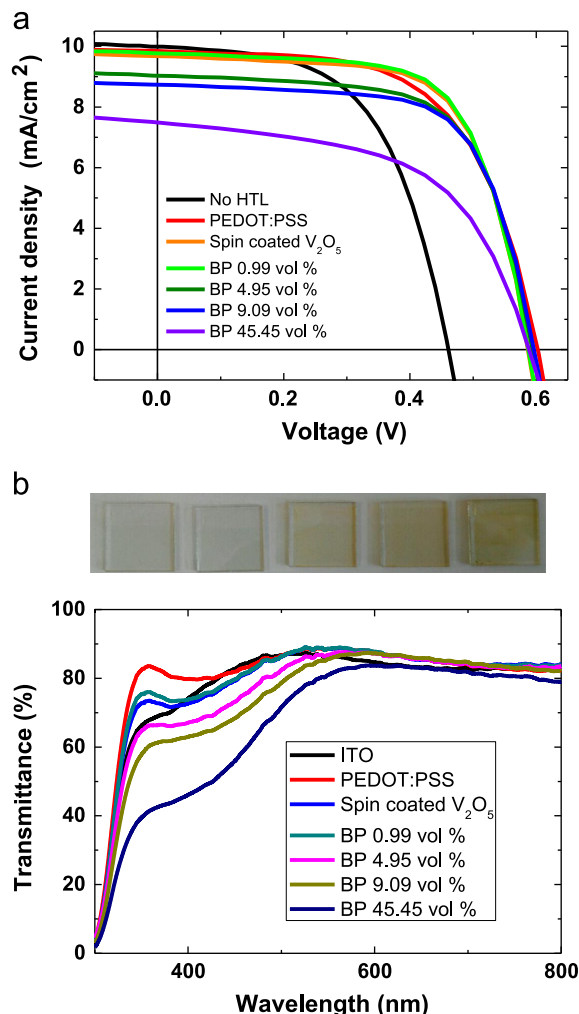


Fig. 2. (a) J – V characteristics of PSCs without and with various HTLs and (b) optical transmittance of bare ITO and ITO coated with PEDOT:PSS and V_2O_5 HTLs.

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