



Vacuum-deposited interconnection layers for tandem solar cells



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ARTICLE INFO

Article history:

Received 28 February 2014

Received in revised form 30 April 2014

Accepted 5 May 2014

Available online 29 May 2014

Keywords:

Organic solar cell

Small molecule

Tandem cell

Interconnection layer

ABSTRACT

Two different types of vacuum-deposited interconnection layers (ICLs) were investigated for tandem solar cells: (1) a pure metal oxide and (2) an organic matrix doped with conductive dopants. The optical and electrical properties of these ICLs were systematically studied and compared. Taking the characteristics of ICLs into consideration, optical design methodology for balancing the photocurrent of each sub-cell in the tandem cell is presented. According to the design, highly efficient small-molecule tandem solar cells with power conversion efficiencies up to 7.3%–7.4% were experimentally demonstrated in both devices utilizing pure metal oxide and organic matrix ICLs.

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1. Introduction

Over the past few years, organic solar cells (OSC) have drawn a great deal of attention in both research and industrial fields. Advantages such as low weight, flexible form factor, potential for low-cost mass production, and low energy consumption in manufacturing make OSCs one of the most-promising next-generation energy-harvesting technologies [1]. Power conversion efficiencies (PCE) beyond 8% have been achieved in single polymer solar cells [2–5]. Solution-processed small-molecule solar cells (SMOSCs) have lagged polymer solar cells for several years; however, the PCE of these types of devices have been boosted to 9.02% in the past year [6]. In addition, the PCEs of vacuum-deposited single-structured SMOSCs have also reached ~7% [7,8]. Moreover, tandem OSCs with certified PCEs greater than 12% were recently reported by Heliatek GmbH [9].

A tandem cell configuration is one of the most promising strategies to extend beyond the 10% PCE milestone required for valuable commercial applications [10,11]. There are already many successful cases of tandem polymer solar cells demonstrating the potential of this approach [12–15]. Nevertheless, solution process utilization for producing a multilayer structure brings challenges in reproducibility for mass production because of under-layer corrosion. Intermixing is not an issue in vacuum sublimation fabrication, and hence, is a much simpler way to form layer-by-layer stacking in tandem structures [16–18]. However, because of the lower mobility compared to that of polymer materials, the active layer thicknesses of efficient vacuum-deposited SMOSCs are typically between 40 and 60 nm [7,8,19–22]. As illustrated in Fig. 1, careful design of the device architecture to place the 40–60 nm absorbing layers of the sub-cells at two individual resonance peaks of the optical field is essential to balance the photocurrent in sub-cells and maximize the short-circuit current density (J_{SC}) of the tandem device [18,23]. Once the photocurrent of each sub cell were not balanced, the tandem cell efficiency may be lower than

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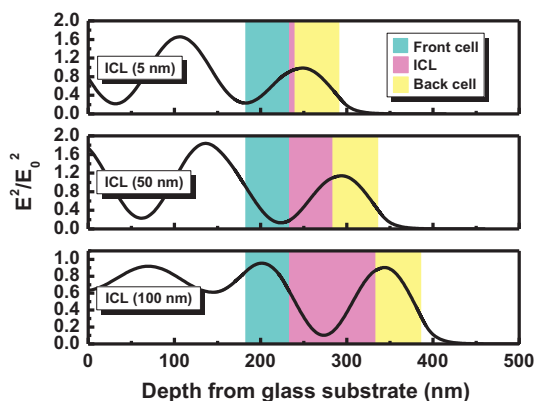


Fig. 1. The optical field distributions of tandem cells with different ICL thicknesses at a wavelength of 550 nm. The active layer thicknesses of both the front cell and the back cell were kept constant at 50 nm.

an optimized single cell [16]. Since the distances between the two resonance peaks are around 150 nm for visible wavelengths [19], accounting for the thin active layers (~50 nm) of SMOSCs, an inter-connection layer (ICL) with thickness ≥ 100 nm is needed to connect the two sub-cells without efficiency loss caused by imbalanced photocurrent. This 100 nm ICL should be transparent and possess excellent carrier transport properties to reduce the photocurrent losses from ICL absorption and charge recombination. To meet these requirements, in previous studies, high conductivity solution processed (3,4- ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) and ZnO nanoparticle layers were usually utilized [8,24,25]. However, the fabrication method is not compatible with vacuum deposited SMOSCs. Leo et al. demonstrated a small molecule based ICL structure constructed by three different doping layers [18]. The method is complicated and may rise the cost in mass production. Our goal was to find a vacuum-process compatible simplified ICL structures for highly efficient tandem SMOSCs.

In this work, we systematically investigated three different types of hole transport layers (HTLs) in ICLs, which may hold the above-mentioned characteristics, and integrated them into a tandem architecture. The first material investigated was a metal oxide (MoO_3), which is commonly used in OSCs and organic light emitting diodes (OLEDs) as a HTL with hole extraction/injection properties [8,19,20,26–30]; however, the thicknesses of MoO_3 used in OSCs and OLEDs are usually thinner than 30 nm [8,19,20,26–30], which is much thinner than the 100 nm thickness ICL required for vacuum-deposited tandem solar cells. We investigated the characteristics of thick MoO_3 films and verified their suitability as HTLs. In OLEDs, a conductive doping technique has been used to increase total device thickness and therefore production yield [31–34]. Two different HTLs with conductive dopants, namely, organic matrix–metal oxide dopant (TAPC: MoO_3) and organic matrix–organic dopant (BF-DPB:F4TCNQ) were examined as HTLs. The chemical structures of the materials used in this work are shown in Fig. 2. The optical properties of these two HTLs were carefully inspected by spectroscopic ellipsometry. The extracted optical constants were

then coupled to optical simulations and checked using single-structured devices. The best HTLs were found to be a neat MoO_3 film of acceptable thickness and the BF-DPB:F4TCNQ system, resulting in impressive PCEs of up to 7.3%–7.4% with same active solar absorbing materials in both the front and back cells.

2. Experimental details

2.1. Thin film and device fabrication

Fullerenes, 4,7-diphenyl-1,10-phenanthroline (Bphen), 1,1-bis[(di-4-tolylamino)phenyl]cyclohexane (TAPC) and 2,3,5,6-tetrafluoro-7,7,8,8-tetracyanoquinodimethane (F4TCNQ) were purchased from *Nichem Fine Technology* and purified at least once by temperature-gradient sublimation before use. *N,N'*-((diphenyl-*N,N'*-bis(9,9-dimethyl-fluoren-2-yl)-benzidine (BF-DPB) and 1,4,5,8,9,11-hexaazatriphenylene hexacarbonitrile (HAT-CN) were purchased from *Lumtec* and used without purification. MoO_3 (*Sigma-Aldrich*), organic thin films, and silver electrodes were deposited on the pre-cleaned indium tin oxide (ITO) coated glass substrates in a high vacuum chamber with base pressure $\sim 1 \times 10^{-6}$ Torr. The sheet resistance of the ITO was $\sim 15 \Omega/\text{sq}$. The deposition was performed at a rate of 2–3 Å/s with the substrate held at room temperature. Thicknesses of the thin films were monitored using a spectroscopic-ellipsometry-calibrated crystal oscillator during deposition. The active area of the cells had an average size of 5 mm² (intersection area between Ag cathode and ITO anode) and each device active area was carefully measured using a calibrated optical microscope. The devices were encapsulated using a UV-cured sealant (*Everwide Chemical Co.*, *Epowide EX*) and a cover glass under an anhydrous nitrogen atmosphere after fabrication and subsequently characterized in air. The single MoO_3 -based cells were configured as follows: ITO/ MoO_3 (40–100 nm)/2-[[7-(4-*N,N*-ditolylaminophenyl-1-yl)-2,1,3-benzothiadiazol-4-yl]methylene]malononitrile (DTDCPB) (7 nm)/DTDCPB: C_{70} (1:1.6 by volume, 40 nm)/ C_{70} (7 nm)/Bphen (6 nm)/Ag (120 nm). The single TAPC: MoO_3 -based cells were configured as follows: ITO/TAPC: MoO_3 (MoO_3 20, 35, and 50 vol%, 100 nm)/DTDCPB (7 nm)/DTDCPB: C_{60} (1:1.6 by volume, 40 nm)/ C_{60} (20 nm)/Bphen (6 nm)/Ag (120 nm). The single BF-DPB:F4TCNQ-based cells were configured as follows: ITO/BF-DPB:F4TCNQ (F4TCNQ 2.5, 5, and 10 vol%, 100 nm)/ MoO_3 (5 nm)/DTDCPB (7 nm)/DTDCPB: C_{70} (1:1.6 by volume, 40 nm)/ C_{70} (7 nm)/Bphen (6 nm)/Ag (120 nm). The energy level diagram of single cells are illustrated in Fig. S1. The tandem cells were configured as: (i) ITO/ MoO_3 (5 nm)/DTDCPB (6 nm)/DTDCPB: C_{70} (1:1.6 by volume, 45 nm)/ C_{70} (6 nm)/Bphen (6 nm)/Ag (0.3 nm)/HAT-CN (1.5 nm)/ MoO_3 (60 nm)/DTDCPB (6 nm)/DTDCPB: C_{70} (1:1.6 by volume, 45 nm)/ C_{70} (20 nm)/Bphen (6 nm)/Ag (120 nm); (ii) ITO/ MoO_3 (5 nm)/DTDCPB (6 nm)/DTDCPB: C_{70} (1:1.6 by volume, 45 nm)/ C_{70} (6 nm)/Bphen (6 nm)/Ag (0.3 nm)/HAT-CN (1.5 nm)/BF-DPB:F4TCNQ (5%, 98 nm)/ MoO_3 (5 nm)/DTDCPB (6 nm)/DTDCPB: C_{70} (1:1.6 by volume, 40 nm)/ C_{70} (6 nm)/Bphen (6 nm)/Ag (120 nm).

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