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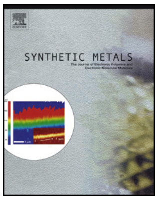

Reversed organic–inorganic hybrid tandem solar cells for improved interfacial series resistances and balanced photocurrents

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ABSTRACT

We report a promising approach to fabricate organic–inorganic hybrid tandem photovoltaic devices with a reversed configuration. High bandgap hydrogenated amorphous silicon (a-Si:H) was deposited with plasma-enhanced chemical vapor deposition onto a solution-processed low bandgap organic photovoltaic (OPV) subcell. Two important factors for efficient tandem solar cells, interfacial series resistance ($R_{S,int}$) and balanced photocurrents, were investigated. The intimate interfacial contact eliminated the $R_{S,int}$, and sufficient transmittance of the OPV front subcell led to the well-balanced photocurrents. As a result, this reversed hybrid tandem device showed an enhanced efficiency of 3.3% and an open-circuit voltage of 1.51 V.

1. Introduction

To surpass the theoretical limit for the power conversion efficiency (PCE) of single-junction solar cells proposed by Shockley and Queisser [1], a tandem device strategy, in which multiple solar cells with different bandgaps are stacked to utilize the broad solar spectrum, has been considered [2,3]. In this regard, there have been much efforts to develop highly efficient tandem photovoltaic (PV) devices including micromorph tandem cells [4] using hydrogenated amorphous silicon (a-Si:H) and microcrystalline silicon (μc-Si:H), and double-junction organic photovoltaic (OPV) tandem devices [5,6] fabricated by solution-processes. However, in the micromorph tandem cell, the formation of the thick μc-Si:H absorber layer induces high production costs, and the solution-processed multi-junction OPV tandem cells present difficulties in stacking multiple organic layers in turn because the deposition of an upper subcell might dissolve or damage an underlying subcell [7]. To overcome these drawbacks, hybrid tandem PV devices that combine organic and inorganic semiconductors have emerged as advanced multi-junction solar cells with the low production cost and multi-layer processing robustness [8–11]. Although the low-cost inorganic semiconductor such as a-Si:H has high charge mobilities and robust mechanical properties, the a-Si:H single-junction solar cell suffers from the narrow absorption range compared to the solar spectrum.

Previously our group created an organic–inorganic hybrid tandem solar cell by combining an a-Si:H front cell and an OPV rear cell with a low bandgap polymer to utilize the broad solar spectrum [8]. However, the previous hybrid tandem solar cell showed a low PCE due to the unbalanced photocurrent and an unfavorable interfacial series resistance between the a-Si:H and polymer layers. To resolve the unfavorable interfacial contact, when we employed a thermally evaporated MoO_3 layer instead of the polymeric hole transporting layer on the a-Si:H layer, the interfacial contact was improved moderately [8]. Furthermore, we were able to eliminate the interfacial series resistance completely by inserting a highly conductive indium–tin-oxide (ITO) interlayer between the subcells [9]. However, these interlayers requiring the additional vacuum-involved procedure might increase the complexity of the device production process.

Tandem solar cells conventionally utilize a high bandgap material as the front cell and a low bandgap material as the rear cell to reduce the thermalization loss of excess photon energy [3,12]. However, in the previous hybrid tandem solar cell with the conventional configuration as illustrated in Fig. 1a (left), highly unbalanced photocurrents of subcells (7.7 mA/cm^2 for the a-Si:H front cell and 2.0 mA/cm^2 for the OPV rear cell [8]) limited the total photocurrent of the tandem solar cell. Moreover, the low

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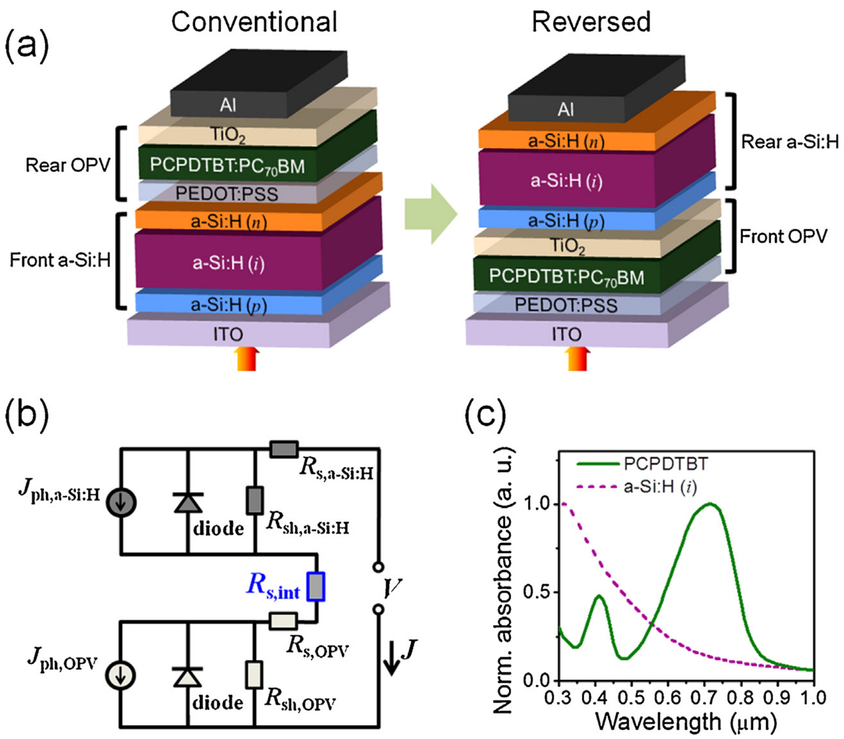

Fig. 1. Illustrations of the hybrid tandem devices. (a) Schematic diagrams of the conventional and the reversed hybrid tandem device configurations. (b) An equivalent circuit diagram for the hybrid tandem device showing $R_{S,\mathrm{int}}$. (c) Normalized absorption spectra of the PCPDTBT and a-Si:H as active materials used in this work.

bandgap OPV devices showed the trade-off between light absorption and charge collection with regard to the film thickness. In other words, when the active layer thickness of the OPV device was increased above 100 nm for the higher light absorption, the descent of PV performance of the OPV cell was inevitable due to the significant charge recombination loss [13]. The main causes of the recombination loss can be attributed to inefficient exciton dissociation and charge collection [14]. In contrast, photocurrent generation of the a-Si:H is non-excitonic [15] and the charge collection from the a-Si:H is efficient because of the relatively high charge mobilities ($\mu_p = 0.3\ \mathrm{cm^2/V\,s}$ and $\mu_n = 2\ \mathrm{cm^2/V\,s}$ for the a-Si:H layer [16], $\mu_p = 1.8 \times 10^{-3}\ \mathrm{cm^2/V\,s}$ and $\mu_n = 3.5 \times 10^{-3}\ \mathrm{cm^2/V\,s}$ for a PCPDTBT:PC$_{70}$BM blend layer [17]). Thus, J_{SC} of the a-Si:H single cell increased with the film thickness up to 400 nm without severe recombination loss.

Taking these material characteristics into account, herein we present a reversed hybrid tandem device configuration with the lower bandgap bulk heterojunction blend layer as the thin front cell and the higher bandgap a-Si:H layer as the thick rear cell as shown in Fig. 1a (right). In this reversed device configuration, two important limits to efficient tandem PV devices were ameliorated. First, the $R_{S,\mathrm{int}}$ depicted in Fig. 1b was significantly reduced by merely altering the layer deposition sequence without inserting any additional interlayer. Second, by using the reversed configuration that placed the semitransparent OPV as the front cell, sufficient photocurrents were generated at the current-limiting rear cell so that the photocurrent-matched condition was almost fulfilled.

2. Experimental

The hybrid tandem solar cells were fabricated mainly with the reversed device configuration shown in Fig. 1a. A hole transporting poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS, Clevios$^{\mathrm{TM}}$ P VP AI 4083) layer with a thickness of 30 nm was formed by spin-coating on an indium-tin-oxide (ITO) coated glass substrate at 4000 rpm and then dried at 110 °C for 10 min. Poly[2,6-(4,4-bis-(2-ethylhexyl)-4*H*-cyclopenta[2,1-b;3,4-b′]-dithiophene)-*alt*-4,7-(2,1,3-benzothiadiazole)] (PCPDTBT, Lumtec Corp., Taiwan) and [6,6]-phenyl-C$_{71}$ butyric acid methyl ester (PC$_{70}$BM, Nano-C, USA) was employed as a low bandgap polymer ($E_g = 1.4$ eV) [18] and an electron acceptor, respectively, in the active layer of the bulk-heterojunction OPV cell. Normalized absorption spectrum of a PCPDTBT polymer film is shown along with an a-Si:H film in Fig. 1c. The PCPDTBT:PC$_{70}$BM bulk heterojunction active layer with a thickness of 70 nm was formed by spin-coating on the dried PEDOT:PSS layer from a chlorobenzene solution with a blending weight ratio of 1:3.5 and a polymer concentration of 6 mg/ml. On top of the active layer, a TiO$_2$ nanoparticle layer was spin-coated from a colloidal solution dispersed in ethanol with a concentration of 0.4 wt%. [19,20] The a-Si:H layers, with total thicknesses of 150 nm or 300 nm, were utilized as high bandgap active materials ($E_g = 1.8$ eV). They were deposited onto the TiO$_2$ nanoparticle layer by using a parallel-plate very-high frequency plasma-enhanced chemical vapor deposition (VHF PE-CVD) system with the *p*–*i*–*n* sequence at 150 °C. The VHF PE-CVD process was operated at RF frequency of 80 MHz and RF power density of 40 W/cm^2 with SiH$_4$ (flow rate of 20 sccm), H$_2$ (60 sccm), and Ar (10 sccm) as source gases at the operating pressure of 0.4 torr with the film growth rate of 0.2 nm/s. B$_2$H$_6$ and PH$_3$ gases were used for *p*-type and *n*-type doping, respectively. The gap distance between the RF electrode and the substrate was maintained to be 20 mm. The *p*-type and *n*-type a-Si:H layers were 5 nm and 25 nm thick, respectively. Single cells were also fabricated with configurations of ITO/PEDOT:PSS/PCPDTBT:PC$_{70}$BM/TiO$_2$/Al for the OPV cell and ITO/a-Si:H(*p*–*i*–*n*)/Al for the a-Si:H cell. The current density–voltage (*J*–*V*) characteristics were measured by a Keithley 2400 source meter with a 150 W solar simulator (Yamashita Denso Corp., YSS-50 A) under a light intensity of 100 mW/cm^2 (1 sun, air mass 1.5 global (AM1.5G) illumination) or in the dark. Optical properties of samples prepared on slide