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# Organic tandem solar cell using active inter-connecting layer

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### article info

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# 1. Introduction

Organic solar cells (OSCs) have attracted much attention due to its potential for fabricating low-cost and large-area flexible solar cells [\[1–5\].](#page-4-0) The performance of the OSCs has obtained huge progress in the past years and power conversion efficiencies (PCEs) over 8% has been achieved [\[6\]](#page-4-0). In order to reach viable efficiencies for industry application [\[4\]](#page-4-0), OSCs need to go beyond the limit of the single heterojunction solar cell [\[7\]](#page-4-0). However, the further improvement in OSCs' performance is limited by the essential properties of the organic semiconductors which are narrow absorption bandwidth, short exciton diffusion length and low charge carrier mobility. Tandem organic solar cells are considered to be an effective way which can potentially lead to an efficiency of about 15% [\[8\]](#page-4-0). In tandem solar cells, two photoactive layers are separated via an inter-connecting layer (ICL). Also, for better absorption and energy conversion, each of the photoactive unit is designed to convert different spectral regions. Therefore, the ICL used to connect the subcells always plays an important role in both electronic and optical properties of tandem solar cells.

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#### **ABSTRACT**

We developed an active inter-connecting layer (ICL) composed of  $SnCl<sub>2</sub>Pc/Al/F<sub>16</sub>CuPc/ZnPc$ to achieve an effective organic tandem solar cell consisting of complementary absorbing layers. This ICL provides a new function to improve the light response of the top cell to enhance current matching between bottom cell and top cell. Meanwhile, the ICL is highly transparent and has efficient charge collections to realize electric connection in series. Finally, in the tandem cell, the open-circuit voltage of 1.52 V is obtained that is the summation of the single cells (1.08 V and 0.46 V), and the power conversion efficiency of 3.21% under 100 mW/cm<sup>2</sup> is achieved that is higher than those of the single cells.

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So far, many ICLs have been reported [\[9\],](#page-4-0) such as: thin gold layer [\[10,11\]](#page-4-0), Ag nanoclusters [\[12\]](#page-4-0), ITO/poly(3,4-ethylene dioxythiophene):(polystyrene sulfonic acid) (PED-OT:PSS) [\[13\]](#page-4-0), metal-oxides/PEDOT:PSS [\[14,15\],](#page-4-0) Al/Au/ PEDOT:PSS [\[16\],](#page-4-0) thin metal layer/doped transport layer [\[17,18\]](#page-4-0), thin metal layer/metal-oxides [\[19–22\],](#page-4-0) and highly doped organic layers [\[23\]](#page-4-0). All these ICLs can give a good electronic connection and have high transparence to reduce optical loss. However, for a high performance tandem cell we always hope the ICL to give an extra function to improve the device performance in the whole tandem cell by some other effective methods. For example: one common method is using the ICL as an optical spacer layer to modulate the optical field distribution due to the interference effect and a nearly current matching can be realized [\[18,24,25\].](#page-4-0) The other way is using the Ag nanoclusters to enhance light absorption due to the plasmonic effects and an improved light absorption can be obtained [\[12,26\].](#page-4-0) Thus it is implied that exploring the multi-function ICL is a feasible method and urgent need to improve the performance of the organic tandem solar cells. In this work, we choose phthalocyanine tin(IV) dichloride (SnCl<sub>2</sub>Pc)/Al/hexadecafluorinated copper phthalocyanine  $(F_{16}CuPc)/zinc$  phthalocyanine (ZnPc) as the active ICL. This ICL provides a new function to improve the light response of the top cell and enhances current matching between bottom cell and top cell. Meanwhile, the ICL is highly transparent and has efficient charge

(a)

<span id="page-1-0"></span>collections to realize electric connection in series that the open-circuit voltage (1.52 V) of the tandem cell is the summation of the subcells (1.08 V and 0.46 V). Finally, in the optimized tandem cells, a  $J_{\rm SC}$  of 3.98 mA/cm<sup>2</sup> is obtained which is near the  $J_{SC}$  of the single cell and a PCE of 3.21% has been attained which is higher than that of single cells.

# 2. Experimental

# 2.1. Device fabrication

The ITO-coated glass substrates with a sheet resistance of 15  $\Omega\,\square^{-1}$  were used as anode. The substrates were cleaned with detergent, then ultrasonicated in acetone, alcohol, and deionized water in sequence and, subsequently, dried in pure  $N_2$ . All organic materials were purchased from Aldrich Corp. and purified twice by using thermal gradient sublimation before used. Al was used as the cathode. The organic and metal materials were thermally evaporated at a base pressure of 10 $^{-4}$  Pa at rates of 1–2 and 5–10 Å/s, respectively. Their thicknesses were monitored by a quartz crystal microbalance during film deposition. A shadow mask was used to define the area of cathode,  $0.0314 \text{ cm}^2$  and  $0.2826 \text{ cm}^2$  for the measurements of current–voltage (I–V) and external quantum efficiency (EQE), respectively.

The layer stack of the tandem structure is schematically shown in Fig. 1(a). Chloroboron subphthalocyanine (Sub- $Pc$ )/ $C_{60}$  cell is as the bottom cell and lead phthalocyanine  $(PbPc)/C_{60}$  cell is as the top cell. The two subcells are sandwiched between a 140 nm thick ITO layer and 60 nm thick Al layer. The ICL to connect two subcells is  $SnCl<sub>2</sub>PC/Al/$  $F_{16}CuPC/ZnPC$ . The thickness for SnCl<sub>2</sub>Pc, Al,  $F_{16}CuPC$ , and ZnPc is 3, 1, 1, and 2 nm, respectively.

#### 2.2. Characterization

The I–V curves were measured with a keithley 2400 source measure unit under 100 mW cm<sup>-2</sup> illuminations with an AM 1.5G filter (SS 150 W solar simulator, Sciencetech Inc.). The illumination intensity was calibrated with a standard silicon photovoltaic traced to the National Renewable Energy Laboratory (NREL). The external quantum efficiency (EQE) was measured with Q Test Station 2000 (Growntech Inc. USA). The measurements were carried out at room temperature in air. Absorption spectrum was taken using the Jasco V-570 UV–visible–near infrared spectrophotometer. The  $n$  and  $k$  values for the different layers in the tandem cell were measured using an ellipsometer (HORIBA Jobin Yvon) and the values obtained were fed into the software to get the optical field profile.

# 3. Results and discussion

#### 3.1. Tandem cell structure

In our tandem cell, SubPc/ $C_{60}$  cell is used as the bottom cell, PbPc/ $C_{60}$  cell as the top cell. The energy level diagram is shown in Fig. 1(b). When light passes through the tandem cell, high energy photons can be absorbed first by



Al/buffer layer

Fig. 1. (a) Device configuration and (b) energy level diagram at flat-band condition.

wide bandgap material SubPc (2.0 eV) and low energy photons do not lose, then the low energy photons can be absorbed by the low bandgap material PbPc (1.3 eV). Therefore, this is a reasonable device structure which ensures the minimal thermalization [\[27\].](#page-4-0) The absorption spectra of SubPc, PbPc and  $C_{60}$  are shown in [Fig. 2\(](#page-2-0)a). SubPc shows an absorption range between 500 and 610 nm and PbPc between 600 and 900 nm, which are completely complementary and give a wide spectral coverage. To further investigate the effect of the ICL on the tandem cell, we fabricate the different device structures without metal cathode: tandem cell 1 with Al as the ICL, tandem cell 2 with Al/ZnPc as the ICL. Here, Al is used as the recombination center as usual [\[19\]](#page-4-0), and ZnPc is considered to be the buffer layer of the top cell which has been used to improve light absorption in the single cell [\[28\]](#page-4-0). The absorbance of the two devices is shown in [Fig. 2](#page-2-0)(b). The absorption spectrum of the tandem cell 1 with Al as the ICL completely comes from combination of the SubPc and PbPc single cell except a weak absorption peak appearing at 910 nm. However, in the tandem cell 2 with Al/ZnPc as the ICL the new absorption peak at 910 nm is further enhanced from 0.1 optical density (O.D.) to 0.2 (O.D.) compared to the tandem cell 1. It can be seen this change must come from the ZnPc which has an impact on the top cell [\[28\].](#page-4-0) Therefore, it is the evidence that the ICL of Al/ZnPc works effectively as a new function like the buffer layer to enhance the light response of the  $PbPc/C_{60}$  top cell.

In order to further confirm the spectrum enhancement, the external quantum efficiency (EQE) of the reference single cells: SubPc/C<sub>60</sub>, PbPc/C<sub>60</sub> and ZnPc/PbPc/C<sub>60</sub> were tested as shown at [Fig. 2\(](#page-2-0)c).  $ZnPC/PbPc/C<sub>60</sub>$  single cell shows a wider and higher EQE in the whole spectrum than

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Fig. 2. (a) Absorption spectra of PbPc, SubPc, C60 and the transmittance spectra of the inter-connecting layer (blue). (b) Absorbance (optical density, O.D) of tandem cell 1 with Al as inter-connecting layer and tandem cell 2 with Al/ZnPc as inter-connecting layer. (c) EQE for the devices: ITO/SubPc (18 nm)/C<sub>60</sub> (40 nm)/buffer layer (BL)/Al (squares), ITO/PbPc (35 nm)/C60 (40 nm) /BL/Al (circles), and ITO/ZnPc(2 nm)/PbPc (35 nm)/ $C_{60}$  (40 nm)/BL/Al (triangles). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

the PbPc/ $C_{60}$  single cell, thus at the same thickness of the PbPc, the ZnPc/PbPc/C<sub>60</sub> single cell can get a larger  $J_{\text{SC}}$ . Meanwhile, SubPc/ $C_{60}$  single cell gives a good performance between 350 and 650 nm. Altogether, the two single cells show the good performance between 350 and 1000 nm which almost cover the whole solar spectrum.

When the tandem cells use the Al/ZnPc as the ICL, the J-V curve is an S-shape with a  $V_{OC}$  only 1.04 V which is much smaller than double of the single cell (in Fig. 3). Thus it indicates that there is a potential barrier between the subcell and ICL, which reduces the  $V_{OC}$  of the tandem cell. As shown in [Fig. 1](#page-1-0)(b), the barrier should be formed between Al (Work Function 4.2 eV) and ZnPc (HOMO 5.0 eV). Herein, the  $F_{16}$ CuPc layer was introduced into the ICL, since the



Fig. 3. The comparison of the *I–V* characteristics of the tandem solar cells with different combinations of inter-connecting layers.

F16CuPc/ZnPc interface can form accumulation heterojunction with high conductivity which has been extensively used as an interface buffer layer to form a good ohmic contact [29-31]. Meanwhile,  $SnCl<sub>2</sub>PC$  is inserted into the interface between  $C_{60}$  and Al, which is a good electron transport material to reduce the contact resistance [\[32,33\]](#page-4-0). Finally, as shown in Fig. 3, the tandem cells get a good performance that a  $V_{OC}$  of 1.52 V is obtained which is the summation of the subcells (1.08 V and 0.46 V). And the fill factor (FF) of 0.53 is obtained which also indicates that the ICL of  $SnCl<sub>2</sub>Pc/Al/F<sub>16</sub>CuPc/ZnPc$  has low resistance. The transmittance spectral of this ICL is also shown in Fig. 2(a) and there is more than 90% transmittance between 300 and 1000 nm. Hence, as the ICL,  $SnCl<sub>2</sub>PC/Al/F<sub>16</sub>CuPC/$ ZnPc has high transparency and efficient charge collections to realize electric connection in series. Furthermore,  $SnCl<sub>2</sub>PC/Al/F<sub>16</sub>CuPC/ZnPC$  is active to enhance the light absorption of the top cell and extend the spectral coverage of the tandem cell from 300 to 1000 nm ,which is the widest spectral coverage ever reported in the organic solar cells.

#### 3.2. Optimizing device performance

In order to acquire the best efficiency of the tandem cell, we need to optimize the thickness of the each layer of the tandem cell. In series solar cell it must be considered to balance the current of the bottom cell and top cell, and the  $J_{SC}$  in the tandem solar cell is decided by the smaller one of the subcells. Fig.  $4(a)$  shows the  $J_{SC}$  of the single cell PbPc/C<sub>60</sub> and ZnPc/PbPc/C<sub>60</sub>. It can be seen that a larger  $J_{\rm SC}$ can be obtained in ZnPc/PbPc/ $C_{60}$  (J<sub>SC</sub> = 7.33 mA/cm<sup>2</sup>) compared to PbPc/ $C_{60}$  (J<sub>SC</sub> = 5.13 mA/cm<sup>2</sup>), which is consistent with the EQE (Fig. 2(c)). Besides, in SubPc/ $C_{60}$  single cell, it has a shorter spectral response and gives the  $J_{SC}$  that is only 4.80 mA/cm<sup>2</sup>. Thus, in the optimized process of the tandem cell, the SubPc layer was arranged as the optimal thickness of its single cell. The thickness of PbPc at the top cell was changed to modulate the  $J_{SC}$  of the tandem cell. From the [Fig. 4\(](#page-3-0)b), it can be seen that the optical electric field at SubPc is decreased with the increase of the PbPc's thickness. Therefore, it is necessary to choose the thin PbPc film in order to get a larger  $J_{SC}$  in the subcell (SubPc/C<sub>60</sub>). When the active ICL was used in our tandem cell, the light response of the top cell PbPc/ $C_{60}$  is enhanced and at the same thickness of PbPc a larger  $J<sub>SC</sub>$  can be obtained. From

<span id="page-3-0"></span>

Fig. 4. Calculated optical field distribution of the tandem cell with cell structure ITO/SubPc (18 nm)/C<sub>60</sub> (16 nm)/inter-connecting layers/PbPc (25 nm)/C<sub>60</sub> (20 nm)/buffer layer (5 nm)/Al according to the method referred in the paper [\[21\]](#page-4-0). (a) J<sub>SC</sub> of single cell PbPc/C<sub>60</sub> and ZnPc/PbPc/C<sub>60</sub> with various PbPc thicknesses (b)  $|E|^2$  in SubPc layer with various PbPc thicknesses at  $\lambda = 590$  nm. (c)  $f_{SC}$  and FF of tandem solar cell with various PbPc thicknesses (d) Optimized |E|<sup>2</sup> in tandem cell at  $\lambda$  = 590 nm (black) and at  $\lambda$  = 750 nm (red). The blue line shows the |E|<sup>2</sup> in the single cell ITO/SubPc (18 nm)/C<sub>60</sub> (40 nm)/ buffer layer (5 nm)/Al at  $\lambda$  = 590 nm. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

the Fig. 4(a), a  $J_{SC}$  of 6.56 mA/cm<sup>2</sup> can also be achieved at the PbPc's thickness of 25 nm. Fig.  $4(c)$  shows the  $J_{SC}$  and FF in the tandem cell with various PbPc thicknesses. With increasing PbPc thickness,  $J_{SC}$  is firstly increasing from 3.1 mA/cm<sup>2</sup> to 3.98 mA/cm<sup>2</sup>, then decreasing from 3.98 mA/cm<sup>2</sup> to 2.61 mA/cm<sup>2</sup>. It demonstrated that when the PbPc's thickness is below 25 nm,  $J_{SC}$  in the subcell (PbPc/C<sub>60</sub>) is relatively low, which controlled the  $J_{SC}$  of tandem cell. When PbPc's thickness was further increased, the  $J_{SC}$  in the subcell (SubPc/C<sub>60</sub>) became lower due to the interference effect (Fig. 4(b)), which began to control the  $J<sub>SC</sub>$  of the tandem cell. The FF obtains the maximum value of 0.53 when PbPc's thickness is 25 nm, which also indicated a more balanced current in the tandem cell. Fig. 4(d) shows the optimal result of the optical simulation of the tandem cell according to transfer matrix method referred in the paper [\[34\].](#page-4-0) All the value of optical electric field  $|E|^2$ can reach 1 in both bottom cell ( $\lambda$  = 590 nm) and top cell  $(\lambda = 750 \text{ nm})$  which is influenced by interference effects. The  $|E|^2$  of single SubPc (18 nm)/C $_{60}$  (40 nm) cell was also shown in Fig. 4(d), it can be seen that  $|E|^2$  in the bottom cell is almost the same as the single SubPc/ $C_{60}$  cell, so it can be sure that there is a larger current in the bottom cell.

The illuminated J–V characteristics of the tandem cell and the two single cells under 100 mW/cm<sup>2</sup> AM 1.5G illumination are shown in Fig. 5 and their solar cell parameters are summarized in Table 1. It can be seen that the  $V_{OC}$  of the tandem cell is 1.52 V, which is the sum of the subcells (1.08 V and 0.46 V). From that we can see the correct operation of the series connected tandem architecture. The  $J_{SC}$  of the tandem cell is about 3.98 mA/cm<sup>2</sup> that approaches the  $J_{SC}$ 



Fig. 5. *J*–*V* characteristics of a tandem cell and reference single cells measured under standard AM 1.5G 100 mW cm-<sup>2</sup> illumination.





of single cells ( $J_{SC}$  = 4.80 mA/cm<sup>2</sup> for SubPc/C<sub>60</sub> cell and  $J_{SC}$  = 5.13 mA/cm<sup>2</sup> for PbPc/C<sub>60</sub> cell), which indicates the complementary optical absorption of the subcells and proper optical field distribution. At last, we get a PCE of the tandem cell 3.21% that is higher than the single cell (2.79% and 1.06%).

#### <span id="page-4-0"></span>4. Conclusion

In summary, we have presented an active interconnecting layer consisting of  $SnCl<sub>2</sub>PC/Al/F<sub>16</sub>CuPC/ZnPC$  to obtain an efficient organic tandem cell with complementary absorbing layers. The ICL provides a new function to enhance the light response of the top cell, so that nearly current matching between bottom cell and top cell is realized. Meanwhile, the ICL has high transparency and efficient charge collections to realize electric connection in series that the  $V_{\text{OC}}$  (1.52 V) of the tandem cell is the summation of the single cells (1.08 V and 0.46 V). Ultimately, through optimal utilization of the optical interference effect, we get a PCE of 3.21% for the tandem cell under 100 mW/cm<sup>2</sup>, which is higher than those of the single cells. This work provides new methods and thoughts to develop multi-functional ICLs to further improve the performance of the tandem cells.

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