Improved performance of polymer solar cells by using inorganic, organic, and doped cathode buffer layers*

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The interface between the active layer and the electrode is one of the most critical factors that could affect the device performance of polymer solar cells. In this work, based on the typical poly(3-hexylthiophene):[6,6]-phenyl C61-butyric acid methyl ester (P3HT:PCBM) polymer solar cell, we studied the effect of the cathode buffer layer (CBL) between the top metal electrode and the active layer on the device performance. Several inorganic and organic materials commonly used as the electron injection layer in an organic light-emitting diode (OLED) were employed as the CBL in the P3HT:PCBM polymer solar cells. Our results demonstrate that the inorganic and organic materials like Cs_2CO_3, bathophenanthroline (Bphen), and 8-hydroxyquinolatolithium (Liq) can be used as CBL to efficiently improve the device performance of the P3HT:PCBM polymer solar cells. The P3HT:PCBM devices employed various CBLs possess power conversion efficiencies (PCEs) of 3.0%–3.3%, which are ca. 50% improved compared to that of the device without CBL. Furthermore, by using the doped organic materials Bphen:Cs_2CO_3 and Bphen:Liq as the CBL, the PCE of the P3HT:PCBM device will be further improved to 3.5%, which is ca. 70% higher than that of the device without a CBL and ca. 10% increased compared with that of the devices with a neat inorganic or organic CBL.

Keywords: polymer solar cell, interface, cathode buffer layer, morphology

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1. Introduction

Organic solar cells (OSCs) have attracted increasing interest due to their advantages of easy processing, low-cost, high-performance, compatibility with flexible substrates, and use of abundant organic materials.[1–7] It has been regarded as one of the most potential green energy technologies to resolve the increasing energy problems worldwide.[8,9] Recently, rapid development has been achieved for the research of OSC, and the power conversion efficiency (PCE) of OSC has commonly reached 3%–8% in most laboratories,[10–15] while a few have obtained PCE over 10%.[16–18] The device performances of OSCs are mainly determined by the OSC materials, which include not only the donor and acceptor materials, but also the interface and electrode materials.

Conventional OSC devices are composed of a photoactive layer between an indium tin oxide (ITO) anode and a low work function metal cathode. The photoactive layer is formed by the combination of organic donor and acceptor materials, which absorbs solar light and converts photons into charge carriers. The interface material between the active layer and the electrodes plays a key role in charge extraction from the photoactive layer and then the collection to the respective anode and cathode.[19] When the top metal electrode is thermally evaporated on the photoactive layer, the reactive hot metal atoms can lead to chemical interaction at the interface and diffusion into the photoactive layer, which will result in excition quenching and/or a barrier to charge extraction. Inserting a cathode buffer layer (CBL) between the photoactive layer and the top metal cathode is a common way to mitigate the damage from the evaporation of metal onto the photoactive layer.[20] The use of a CBL with a low work function or shallow lowest unoccupied molecular orbital (LUMO) level can efficiently improve the device performance of OSC due to the enhancement of the internal electric field and the reduced carrier defects and traps at the active layer/Al interface.[4,5] Additionally, the CBL can act as both optical spacer and exciton blocking layer.[21] Up to now, the efficient CBL in OSC is still limited to several materials, such as LiF,[22] ZnO,[23] and BCP.[24–26] Thus, it is necessary to develop new materials as the CBL to improve the performance of OSC devices.

In this work, to explore more efficient CBL materials for high-performance OSCs, various inorganic and organic materials commonly used as the electron injection layer in an organic light-emitting diode (OLED), includ-

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ing LiF, Cs_2CO3, ZnS, 1,3,5-tris(2-N-phenylbenzimidazolyl) benzene (TPBi), bathophenanthroline (Bphen), and 8-hydroxyquinolatolithium (Liq), are employed as CBLs between the photoactive layer and the top metal electrode in the poly(3-hexylthiophene):[6,6]-phenyl C61-butyric acid methyl ester (P3HT:PCBM) polymer solar cells. Our results demonstrate that all of the devices with various CBLs exhibit much better device performance than those without CBL.

2. Experimental details

2.1. Materials

All of the materials used in this work were commercially purchased and used as received. The blend solution of P3HT:PCBM was prepared as follows: 20 mg P3HT and 20 mg PCBM were mixed and dissolved in 1 mL o-dichlorobenzene, then the blend solution was stirred for 24 h and filtered through a 0.2 μm filter before use.

2.2. Device fabrication and characterization

As shown in Fig. 1(a), the P3HT:PCBM polymer solar cells were fabricated with the device structure of ITO/PEDOT:PSS/P3HT:PCBM/CBL/Al. The patterned ITO glass substrates with a sheet resistance of 20 Ω/sq were cleaned in an ultrasonic bath with de-ionized water, acetone, and isopropanol consecutively, and then treated by ultraviolet-ozone for 30 min. The clean substrates were immediately spin-coated with a ∼ 30 nm thick layer of poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) (Clevios PVP.AI 4083) and subsequently heated in ambient air for 15 min at 130 °C to remove the residual water. The active layer P3HT:PCBM with a thickness of ∼ 200 nm was prepared in ambient air by spin-coating the blend P3HT:PCBM solution onto the PEDOT:PSS films at 800 rpm for 28 s. The wet films were slowly and completely dried in covered glass Petri dishes. Then, the P3HT:PCBM films were thermally annealed at 110 °C for 10 min. Finally, the films were transferred to a high-vacuum chamber (1×10^{-4} Pa), where the Al (100 nm) (reference device) or CBL/Al (100 nm) cathode was thermally evaporated. The device active area was 0.04 cm^2 defined by the overlap of anode and cathode.

The current density–voltage (J–V) characteristics were measured using a programmable source meter (Keithley 2400) under illumination of 100 mW/cm^2 from a solar simulator with AM 1.5G simulated solar spectrum. The external quantum efficiency (EQE) spectra of the devices were measured under standard measurement conditions on a 7-SCSpec solar cell measurement system manufactured by 7-STAR Co. All the measurements were performed in air under ambient conditions without device encapsulation.

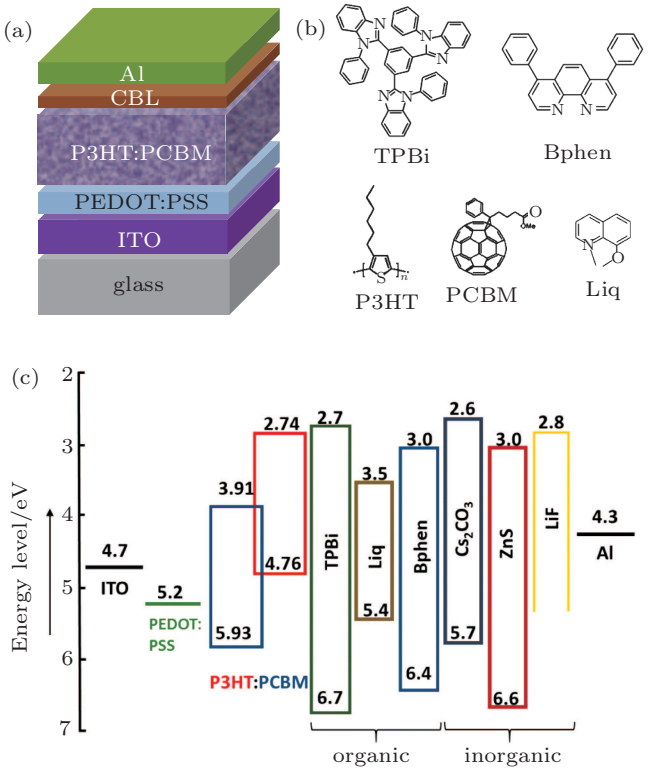


Fig. 1. (color online) (a) Device structure of P3HT:PCBM polymer solar cell. (b) Molecular structures of P3HT, PCBM, TPBi, Bphen, and Liq. (c) Energy-level diagram of materials under investigation.

2.3. Film morphology characterization

Atomic force microscopy (AFM) images were collected in air on a Veeco AFM using a tapping mode. The films for AFM measurement were prepared under the same conditions for fabricating the devices to enable accurate comparisons.

3. Results and discussion

To study the effect of the CBL on the device performance of P3HT:PCBM polymer solar cells, we fabricated and characterized the polymer solar cells based on the device structure of ITO/PEDOT:PSS/P3HT:PCBM/CBL/Al. In the device, the active layer is P3HT:PCBM, and PEDOT:PSS is adopted as the anode buffer layer. For comparison, the same ITO/PEDOT:PSS anode was employed in all of the devices. The P3HT:PCBM photoactive layer was fabricated by spin-coating the P3HT:PCBM solution followed by slow drying and thermal annealing.[27] A P3HT:PCBM device without any CBL was also fabricated as the reference. All of the CBL materials investigated in this work, including LiF (0.3 nm), Cs_2CO_3 (0.3 nm), ZnS (4 nm), TPBi (4 nm), Bphen (4 nm), Liq (1 nm), Bphen:10% Cs_2CO_3 (4 nm), and Bphen:10% Liq (4 nm), were thermally evaporated onto the P3HT:PCBM layer in a high-vacuum chamber. Molecular structures of P3HT, PCBM, TPBi, Bphen, and Liq are shown in Fig. 1(b), and the energy-level diagram of the materials used in this work is given in Fig. 1(c). The J–V characteristics and EQE of various CBL based P3HT:PCBM devices are shown in Figs. 2–4,

and the device performance parameters are summarized in Tables 1–3, which are average values of 16 individual devices. Note that the spin-coating, slow drying, and thermal annealing of the P3HT:PCBM photoactive layer were carried out in air under ambient conditions. We show here that the PCE of 3.58% can be obtained in these air-processed P3HT:PCBM devices, which are comparable to the P3HT:PCBM solar cells fabricated in controlled environments.[28]

3.1. Inorganic material as CBL

Three inorganic materials LiF, Cs_2CO_3, and ZnS were used as the CBL in the P3HT:PCBM device in this work to improve the device performance, and the relative device parameters are summarized in Fig. 2(a) and Table 1. LiF has been commonly used as CBL materials in OSCs and OLEDs.[21] The mechanism of LiF as the CBL and the function of the LiF/Al cathode can be summarized as follows. (i) The LiF layer can efficiently modify the work function of the cathode and thus enhance the electron injection.[29] (ii) The LiF layer can protect the active layer during metal deposition and prevent the reaction between the Al cathode and the carboxylic oxygen of PCBM.[30] (iii) The LiF layer can lead to charge transfer across the interface and form a dipole moment across the junction.[31] However, the insulating nature of LiF indicates that the LiF buffer layer must be very thin to ensure the ohmic contact. Another alkali metal compound Cs_2CO_3, which typically provides low work function contacts as LiF (Fig. 1(c)), has also been widely incorporated for electron injection and transport enhancement in organic devices.[32,33] The alkali cation Cs^+ can dope the adjacent electron transporting layer to form a low work function Al–O–Cs complex upon Al deposition, which facilitates electron extraction in polymer solar cells.[34] The inorganic material ZnS is also commonly used as the electron injection layer in OLED due to its lower work function and high electron mobility.[35] The introduction of ZnS as the CBL can lower the injection barrier and provide more effective electron transport, resulting in a relatively higher PCE.

Figure 2(a) shows the J–V characteristics of the P3HT:PCBM devices under illumination with an intensity of 100 mW/cm^2, while figure 2(b) demonstrates the EQE spectra of the devices without and with different inorganic CBLs. It can be seen that the short circuit current density (J_{sc}), open circuit voltage (V_{oc}), and fill factor (FF) of the devices with an inorganic CBL are significantly improved compared with those of the device without a CBL. As shown in Table 1, the P3HT:PCBM device without a CBL shows a PCE of 2.05% with J_{sc} of 8.69 mA·cm^{-2}, V_{oc} of 0.51 V, and FF of 47%. The incorporation of a 0.3 nm thick LiF layer between the P3HT:PCBM active layer and the Al electrode significantly improves V_{oc} from 0.51 V to 0.57 V and FF from 47% to 65%, thus resulting in an improved PCE of 3.21%. Similar to the LiF case, when applying a 0.3 nm thick Cs_2CO_3 layer as the CBL, an increased PCE of 3.24% will be obtained with enhanced J_{sc} of 8.81 mA·cm^{-2}, V_{oc} of 0.57 V, and FF of 65%. Finally, the insertion of a 4 nm thick ZnS CBL results in an improved PCE of 3.15% with J_{sc} of 8.80 mA·cm^{-2}, V_{oc} of 0.57, and FF of 63%.

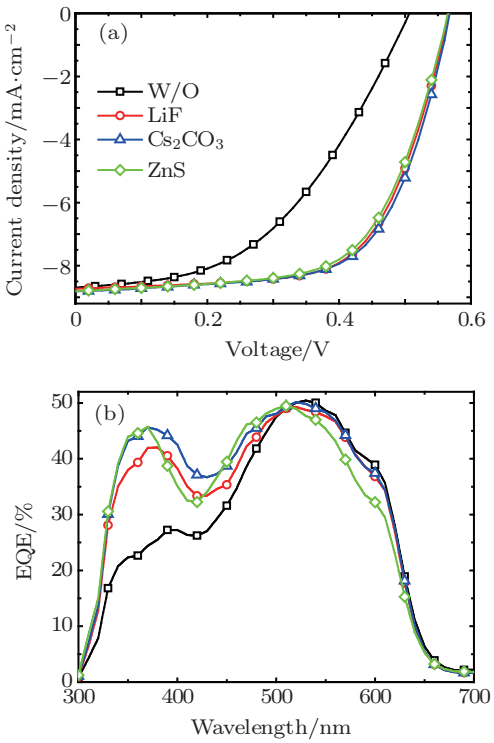


Fig. 2. (color online) Comparison of (a) the J–V characteristics under 100 mW/cm^2 illumination of AM1.5G solar spectrum, and (b) the EQE spectra of the P3HT:PCBM devices with various inorganic CBLs with that of the P3HT:PCBM device without a CBL.

Table 1. Device performance of the P3HT:PCBM devices with various inorganic CBLs. Every parameter in the table is the average of 16 individual devices. R_s is the series resistance of the P3HT:PCBM device.

CBL	V_{oc}/V	J_{sc}/mA·cm^{-2}	FF /%	PCE/%	R_s/Ω·cm^2
W/O	0.51	8.69	47	2.05	22.0
LiF	0.57	8.73	65	3.21	10.2
Cs_2CO_3	0.57	8.81	65	3.24	9.7
ZnS	0.57	8.80	63	3.15	10.9

It is widely believed that the V_{oc} of polymer solar cells depends mainly on the difference between the highest occupied molecular orbital (HOMO) of the donor and the LUMO of the acceptor.[3] Meanwhile, the difference between the work functions of anode and cathode has a minor effect on the V_{oc} of the device.[31] The significantly increased V_{oc} of the devices with LiF/Al, Cs_2CO_3/Al, and ZnS/Al cathodes in comparison with that of the device with Al cathode should be attributed to the lower work functions of LiF, Cs_2CO_3, and ZnS compared with that of Al. Meanwhile, the increased FF for the devices with LiF/Al, Cs_2CO_3/Al, and ZnS/Al cathodes could be explained

by the enhanced charge extraction and obviously decreased series resistance (R_s).[19,31] As shown in Table 1, the R_s of the P3HT:PCBM device decreases from 22.0 Ω·cm^2 for the device without CBL to 10.2 Ω·cm^2, 9.7 Ω·cm^2, and 10.9 Ω·cm^2 for the devices with LiF, Cs_2CO_3, and ZnS CBL layers, respectively. Additionally, the slightly increased J_{sc} of the devices with LiF, Cs_2CO_3, and ZnS CBL layers can be explained by the enhancement of the light harvest,[36,37] which is reflected by the improvement of the photoresponse in the range of 300–500 nm of the EQE spectra (Fig. 2(b)).

3.2. Organic material as CBL

BCP is one of the most common organic materials used as the CBL in OSC and OLED.[38] To explore other organic materials as efficient CBL, three organic materials usually used as the electron injection layer: TPBi, Bphen, and Liq, were employed as the CBL in the P3HT:PCBM device in this work. It has been reported that TPBi and Bphen have relatively higher electron mobilities of 3.3×10^{-5} cm^2·V^{-1}·s^{-1} and 5.0×10^{-4} cm^2·V^{-1}·s^{-1}, respectively, leading to good electron transporting properties.[39] This is why we chose Bphen and TPBi as the CBL in this work. Liq is an organic alkali metal complex, which has been considered as an excellent electron injection material similar to the inorganic alkali metal complex LiF and Cs_2CO_3. However, Liq has rarely been used as the CBL in OSC.

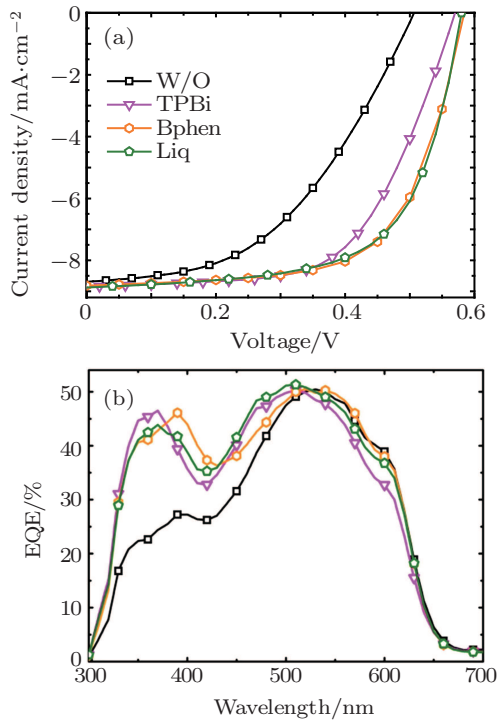


Fig. 3. (color online) Comparison of (a) the *J*–*V* characteristics under 100 mW/cm^2 illumination of AM1.5G solar spectrum, and (b) the EQE spectra of the P3HT:PCBM devices with various organic CBLs with that of the P3HT:PCBM device without a CBL.

Figure 3 shows the *J*–*V* characteristics and EQE spectra of the P3HT:PCBM devices with various organic CBLs compared with those without a CBL. Similar to the devices with an inorganic CBL, the organic CBL based devices also show higher J_{sc}, V_{oc}, and FF than the device without CBL (Table 2), and thus improved PCEs of 3.03%, 3.26% and 3.35% are obtained from the TPBi, Bphen, and Liq CBL based P3HT:PCBM devices, respectively. The inserting TPBi, Bphen, and Liq CBLs with LUMO levels in the range of −3.5 eV to −2.7 eV[40,41] (Fig. 1(c)) efficiently shallow the work function of Al, leading to increased V_{oc} compared with that of the device using a bare Al cathode. Similar to the inorganic CBL based P3HT:PCBM devices, the enhanced charge extraction and decreased R_s (Table 2) could explain the enhancement of the FF of the organic CBL based devices compared with the device without a CBL. In addition, the slightly increased J_{sc} of the device with the CBL layer can also be explained by the enhancement of the light harvest reflected by the improvement of the photoresponse in the range of 300–500 nm of the EQE spectra (Fig. 3(b)).

Table 2. Device performance of the P3HT:PCBM devices with various organic CBLs. Every parameter in the table is the average of 16 individual devices.

CBL	V_{oc}/V	J_{sc}/mA·cm^{-2}	FF /%	PCE/%	R_s/Ω·cm^2
W/O	0.51	8.69	47	2.05	22.0
TPBi	0.57	8.84	60	3.03	15.2
Bphen	0.58	8.82	64	3.26	9.2
Liq	0.58	8.88	65	3.35	9.1

The Liq based device demonstrates slightly higher PCE than the TPBi and Bphen based devices, which can be ascribed to the lower work function contacts provided by this organic alkali metal compound. As shown in the energy-level diagram in Fig. 1(b), the LUMO level of PCBM is −3.91 eV, while the LUMO levels of these three organic materials are in the order of TPBi (−2.7 eV) > Bphen (−3.0 eV) > Liq (−3.5 eV). As a result, the electron transport is more likely to be realized by an electron leaping between PCBM and Liq compared with the Bphen and TPBi cases, which explains the higher device performance of the Liq CBL based P3HT:PCBM solar cell.

3.3. Doped material as CBL

It has been reported that doping can increase the carrier mobility and lead to the generation of very thin space-charge layers at the contacts beneficial for efficient injection.[42–44] The doping of organic materials by metals, such as the doping of Bphen by ytterbium (Yb), has been successfully employed both as an efficient exciton blocker layer and an optical spacer in OSC devices.[45] To further improve the device performance of the P3HT:PCBM devices based on the above described inorganic and organic CBL materials, we used the doped Bphen:Cs_2CO_3 and Bphen:Liq as CBLs between the active layer and the Al electrode. As mentioned above, Bphen has been proved to have a high electron mobility, so we chose it as the matrix by doping with other materials to explore more

efficient doped CBL layers. To obtain the optimal device performance, we also optimized the doping ratio and the thickness of the Bphen:Cs_2CO_3 and Bphen:Liq layers. The Bphen matrix doped with 10% Cs_2CO_3 or 10% Liq with the film thickness of 4 nm was finally found as the most efficient doped CBL layer for high-performance devices.

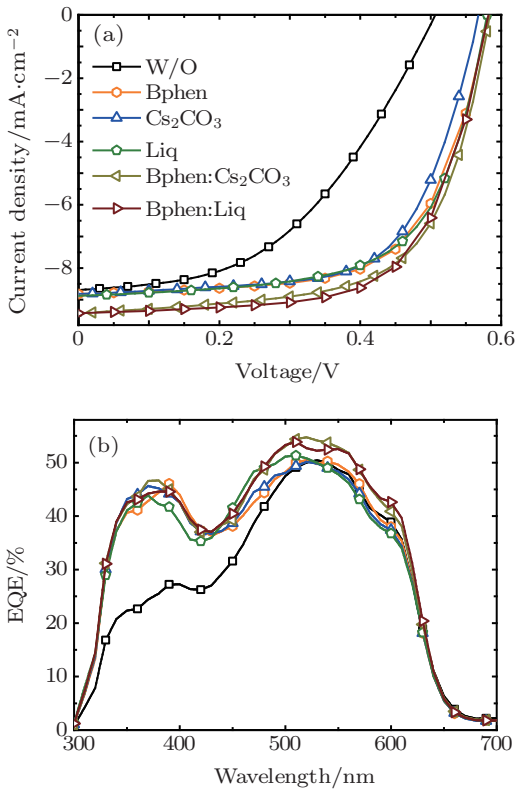


Fig. 4. (color online) Comparison of (a) the *J*–*V* characteristics under 100 mW/cm^2 illumination of AM1.5G solar spectrum, and (b) the EQE spectra of the P3HT:PCBM devices with various inorganic, organic, and doped CBLs with that of the P3HT:PCBM device without CBL.

Table 3. Device performance of the P3HT:PCBM devices with various inorganic, organic, and doped CBLs. Every parameter in the table is the average of 16 individual devices.

CBL	V_{oc}/V	J_{sc}/mA·cm^{-2}	FF /%	PCE/%	R_s/Ω·cm^2
W/O	0.51	8.69	47	2.05	22.0
Bphen	0.58	8.82	64	3.26	9.2
Cs_2CO_3	0.57	8.81	65	3.24	9.7
Liq	0.58	8.88	65	3.35	9.1
Bphen:Cs_2CO_3	0.58	9.43	64	3.54	7.3
Bphen:Liq	0.58	9.42	65	3.58	7.1

As depicted in Fig. 4(a) and Table 3, the P3HT:PCBM device with Bphen:10% Cs_2CO_3 (4 nm) as the CBL shows a PCE of 3.54% with J_{sc} of 9.43 mA·cm^{-2}, V_{oc} of 0.58 V, and FF of 64%, while the device using Bphen:10% Liq as CBL (4 nm) exhibits a PCE of 3.58% with J_{sc} of 9.42 mA·cm^{-2}, V_{oc} of 0.58 V, and FF of 65%. As a result, the doped CBL based device demonstrates ca. 70% higher PCE than the device without CBL, which is attributed to the significantly increased J_{sc}, V_{oc}, and FF. Similar to the above described inorganic and organic CBL based devices, the increased V_{oc} and FF in the doped CBL based devices should be explained by the lower work functions of the Bphen:Cs_2CO_3 and Bphen:Liq cathodes, and the enhanced charge extraction and decreased R_s shown in Table 3, respectively. The enhanced light harvest should be responsible for the increased J_{sc}, as reflected by the EQE spectra.

In comparison with the performance of the devices employing the neat organic or inorganic CBL (Bphen, Cs_2CO_3, and Liq), the doped CBL based devices demonstrate improved J_{sc} and comparable V_{oc} and FF, thus increased PCE. The enhancement of J_{sc} can directly be reflected by the photoresponse enhancement in the EQE spectra. To elucidate the improvement of J_{sc} for the device with doped CBL compared with the device without CBL or with neat inorganic and organic CBL, we compared the EQE spectra of the devices with Al, Bphen/Al, Cs_2CO_3/Al, Liq/Al, Bphen:Cs_2CO_3/Al, and Bphen:Liq/Al cathodes. As shown in the EQE spectra in Fig. 4(b), in comparison with the device with an Al cathode, the P3HT:PCBM devices with Bphen/Al, Cs_2CO_3/Al, and Liq/Al cathodes exhibit an enhanced photoresponse in the whole absorption band of 300–650 nm with a notable enhancement in the

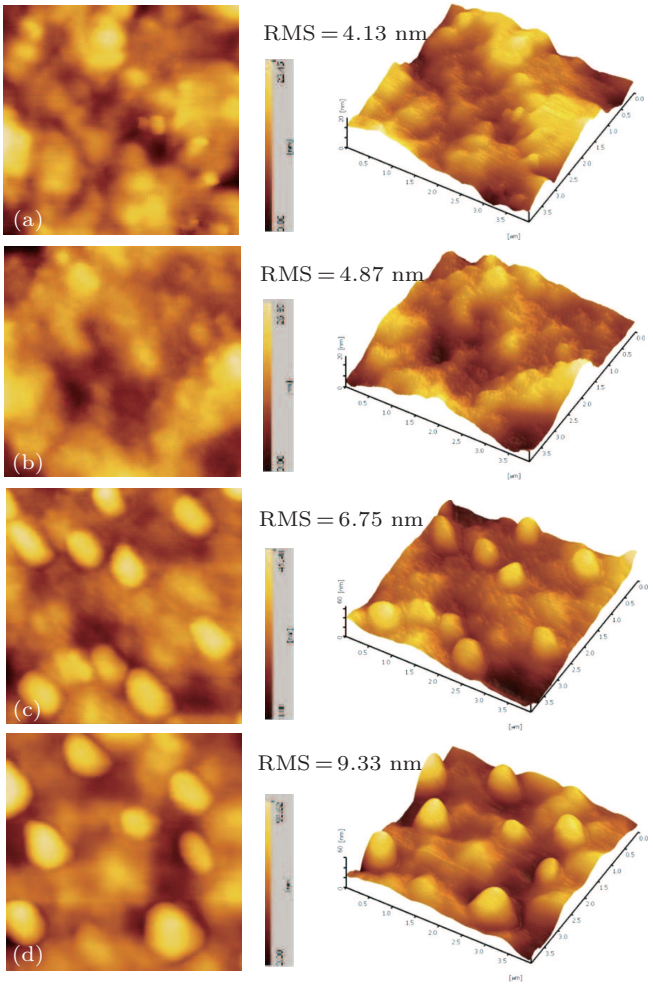


Fig. 5. (color online) Atomic force microscopy topographic and 3D images of P3HT:PCBM blend films on ITO/PEDOT:PSS substrate (a) without CBL, (b) with Cs_2CO_3 CBL, (c) with Bphen CBL, and (d) with Bphen:Cs_2CO_3 CBL. The scan size is 4 μm×4 μm.

wavelength range from 300 nm to 500 nm. Furthermore, the photoresponse of the Bphen:Cs_2CO_3/Al and Bphen:Liq/Al cathode based devices further increases compared with that of the devices with Bphen/Al, Cs_2CO_3/Al, and Liq/Al cathodes in the whole absorption band of 300–650 nm, which is inconsistent with the trend that the doped CBL based devices possess larger J_{sc} not only than the device without CBL, but also than the devices with neat organic and inorganic CBL.

To further understand the effect of these inorganic, organic, and doped CBLs on the device performance of P3HT:PCBM cells, the surface morphologies of P3HT:PCBM photoactive layers without and with various CBLs were also investigated by using tapping-mode AFM, as shown in Fig. 5. The root-mean-square (rms) roughness of the blend layer of P3HT:PCBM is 4.13 nm, while the P3HT:PCBM surfaces covered by Cs_2CO_3, Bphen, and Bphen:Cs_2CO_3 CBL become more rough with a rms of 4.87 nm, 6.75 nm, and 9.33 nm, respectively. The increased roughness of the active layer with the CBL could increase the contact area with the Al cathode deposited on it, which is beneficial to improving the charge collection and thus results in better device performance.[19]

4. Conclusion

We have studied the effect of a CBL between the active layer and the top metal electrode on the performance of P3HT:PCBM polymer solar cells. Several inorganic materials LiF, Cs_2CO_3, and ZnS, organic materials TPBi, Bphen, and Liq, as well as doped materials Bphen:Cs_2CO_3 and Bphen:Liq were employed as the CBL in the P3HT:PCBM polymer solar cells. Our results demonstrate that all of these inorganic and organic materials are efficient CBLs to significantly improve the device performance of the P3HT:PCBM cells. The P3HT:PCBM devices employed various CBLs possessing PCEs of 3.0%–3.3%, which are ca. 50% improved compared to that of the device without a CBL. Furthermore, by using the doped materials as the CBL, the PCE of the P3HT:PCBM device can be further improved up to 3.5%, which is ca. 70% higher than that of the device without a CBL and ca. 10% increased compared to that of the devices with neat inorganic and organic CBLs. This finding indicates that the doped Bphen:Cs_2CO_3 and Bphen:Liq CBLs have a great potential as efficient CBL for high-performance OSCs.

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