

## Article

# Enhanced Photovoltaic Properties of Perovskite Solar Cells by Employing Bathocuproine/Hydrophobic Polymer Films as Hole-Blocking/Electron-Transporting Interfacial Layers

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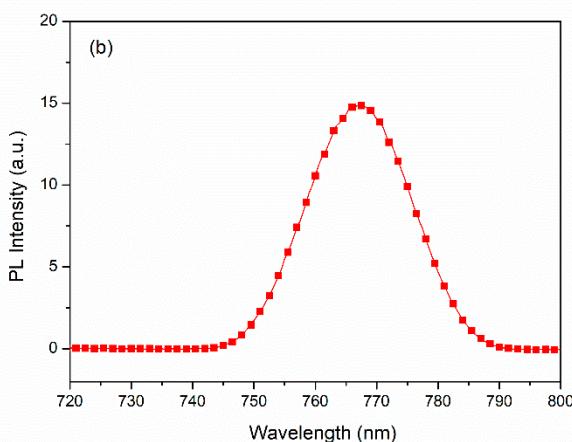
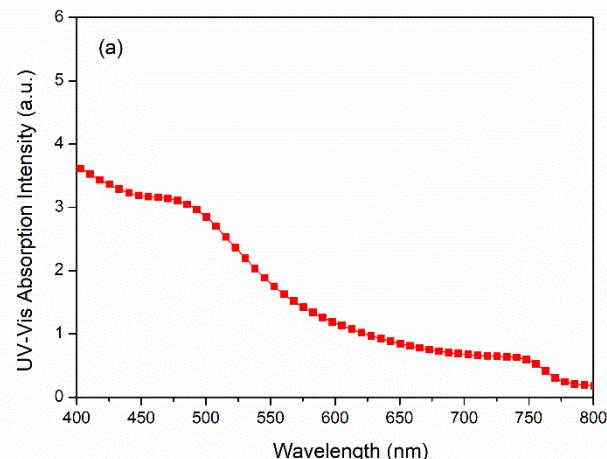
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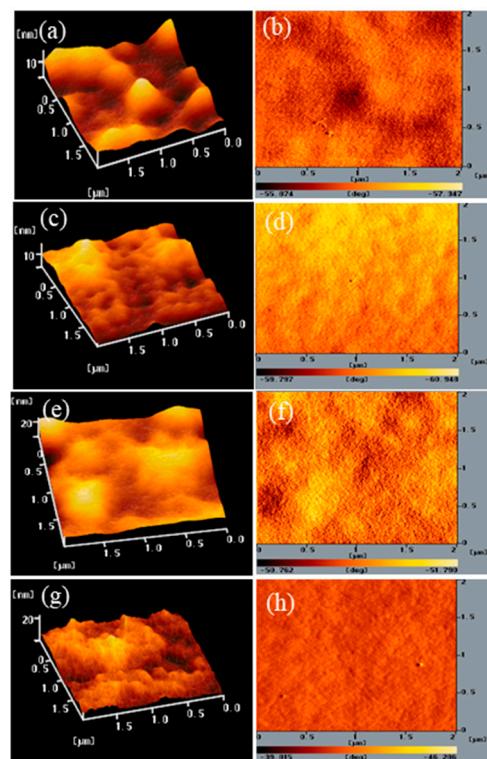
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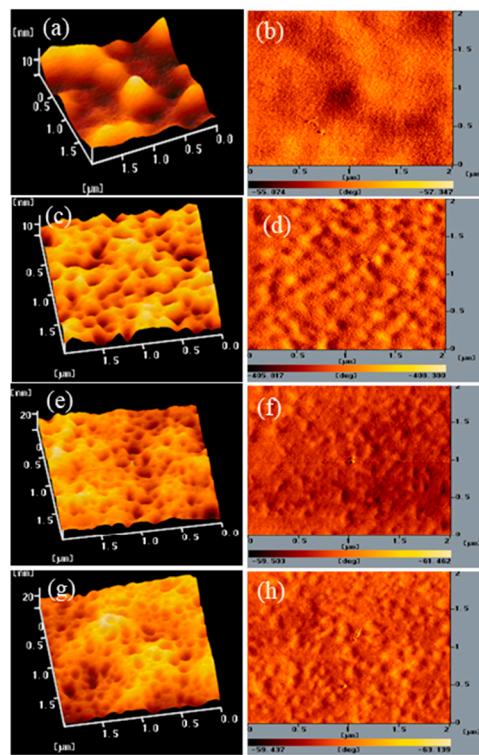
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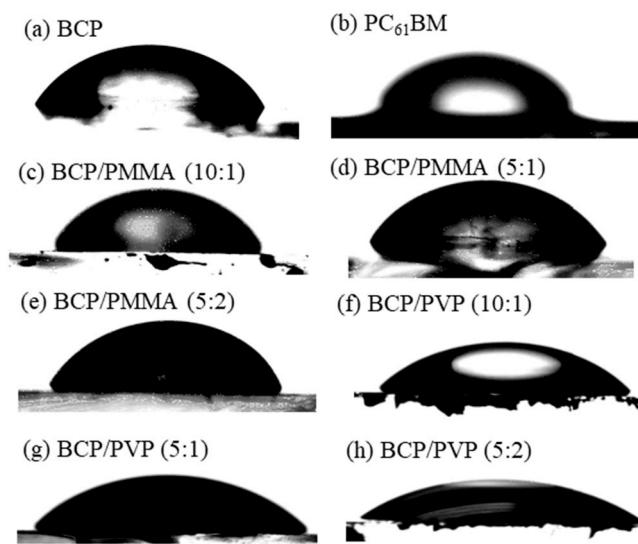
**Figure S1.** (a) UV-Vis absorption and (b) PL spectra of the  $\text{MAPbI}_3$  perovskite film.



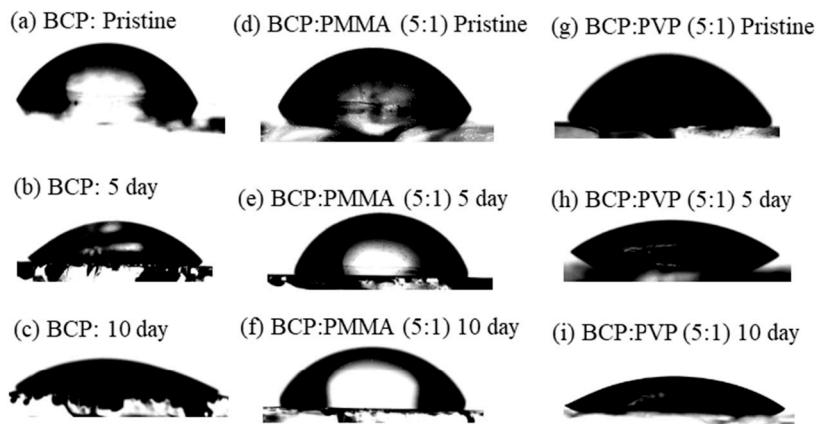
**Figure S2.** (a, c, e, g) Topographic and (b, d, f, h) phase AFM images of (a, b) BCP, (c, d) BCP/PMMA (10:1, *w/w*), (e, f) BCP/PMMA (5:1, *w/w*), and (g, h) BCP/PMMA (5:2, *w/w*) hole-blocking/electron-transporting interfacial layers, recorded after thermal treatment (80 °C, 5 min).



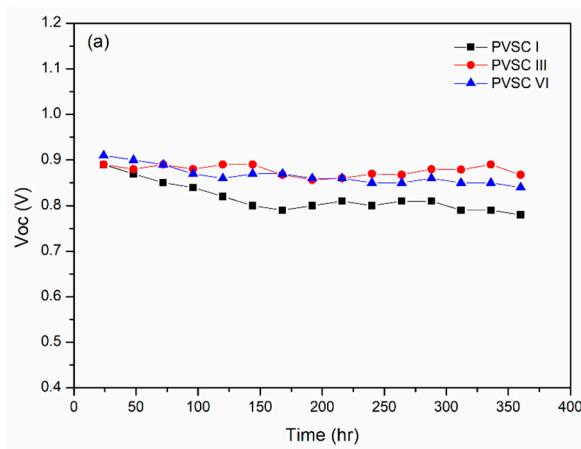
**Figure S3.** (a, c, e, g) Topographic and (b, d, f, h) phase AFM images of (a, b) BCP, (c, d) BCP/PVP (10:1, *w/w*), (e, f) BCP/PVP (5:1, *w/w*), and (g, h) BCP/PVP (5:2, *w/w*) hole-blocking/electron-transporting interfacial layers, recorded after thermal treatment (80 °C, 5 min).

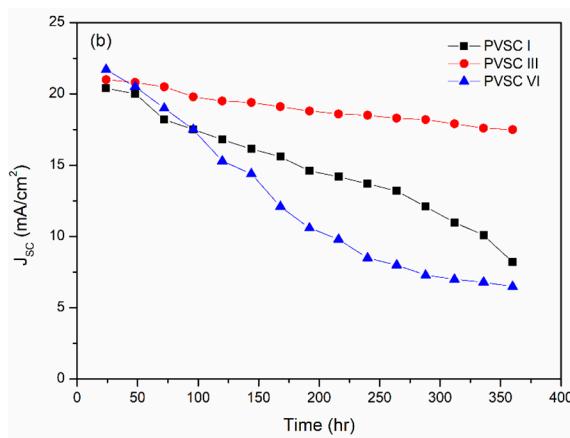


**Figure S4.** Photographs of water droplets on (a) BCP, (b) PC<sub>61</sub>BM, (c, d, e) BCP/PMMA (10:1, 5:1, and 5:2, *w/w*), and (f, g, h) BCP/PVP (10:1, 5:1, and 5:2, *w/w*) films.

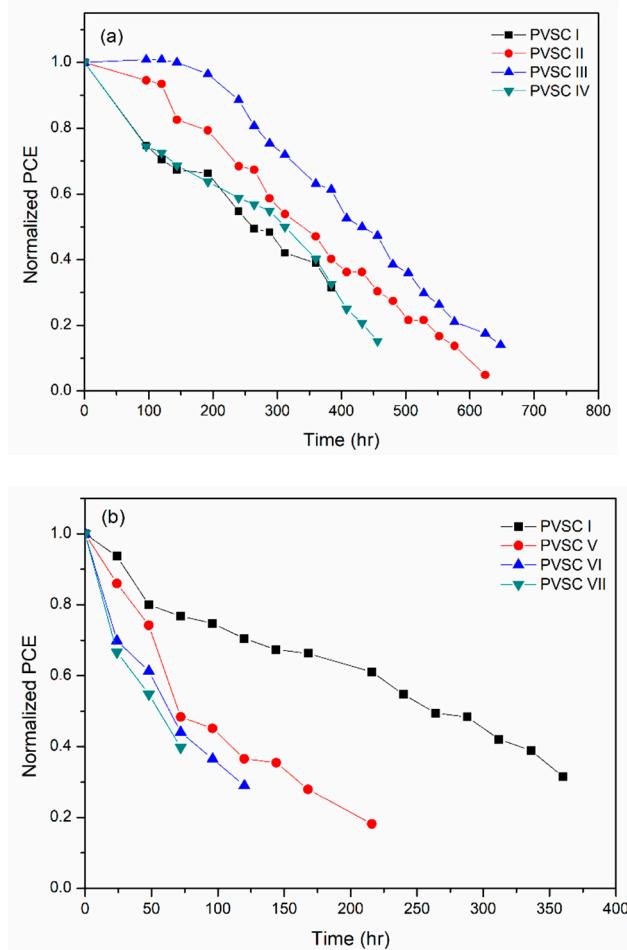


**Figure S5.** Photographs of water droplets on (a–c) BCP, (d–f) BCP/PMMA (5:1, *w/w*), and (g–i) BCP/PVP (5:1, *w/w*) films after storage at 30 °C and 35% relative humidity for 0, 5, and 10 days.





**Figure S6.** Time dependence of  $V_{oc}$  and  $J_{sc}$  of the PVSC I, PVSC III, and PVSC VI (measured at 30 °C and 35% relative humidity).



**Figure S7.** Storage-stability of PVSCs incorporating BCP, BCP/PMMA, and BCP/PVP (measured at 30 °C and 60% relative humidity).

**Table S1.** Crystal sizes of  $\text{MAPbI}_3$  film coated with BCP, BCP/PMMA, and BCP/PVP layers after storage at 30 °C and 35% relative humidity for 0, 5, and 10 days.

Sample	Interfacial layer	Time (days)	Crystal size (nm)
I-1	BCP	0	34.21
I-2	BCP	5	29.05
I-3	BCP	10	28.25

II	BCP/PMMA (10:1)	0	----
III-1	BCP/PMMA (5:1)	0	30.65
III-2	BCP/PMMA (5:1)	5	27.78
III-3	BCP/PMMA (5:1)	10	26.94
IV	BCP/PMMA (5:2)	0	----
V	BCP/PVP (10:1)	0	----
VI-1	BCP/PVP (5:1)	0	32.93
VI-2	BCP/PVP (5:1)	5	28.68
VI-3	BCP/PVP (5:1)	10	27.78
VII	BCP/PVP (5:2)	0	----

**Table S2.** Surface roughnesses and CAs of films of BCP, BCP/PMMA, and BCP/PVP layers after storage at 30 °C and 35% relative humidity for 0, 5, and 10 days.

Sample	Composition (w/w)	Time (days)	RMS (nm)	CA (°)
I-1	BCP	0	3.27	74.5
I-2	BCP	5	2.76	49.0
I-3	BCP	10	7.55	37.8
II	BCP/PMMA (10:1)	0	3.25	75.5
III-1	BCP/PMMA (5:1)	0	3.18	78.4
III-2	BCP/PMMA (5:1)	5	5.14	74.4
III-3	BCP/PMMA (5:1)	10	5.31	72.3
IV	BCP/PMMA (5:2)	0	3.29	81.8
V	BCP/PVP (10:1)	0	3.12	58.4
VI-1	BCP/PVP (5:1)	0	3.09	53.5
VI-2	BCP/PVP (5:1)	5	26.45	43.9
VI-3	BCP/PVP (5:1)	10	38.32	35.7
VII	BCP/PVP (5:2)	0	3.15	43.0

**Table S3.** PV performances of previously reported PVSCs, compared with those measured in this present study.

Device structure	PV performance	Reference
ITO/PEDOT/MAPbI <sub>3</sub> /PC <sub>61</sub> BM/BCP:PVP/Ag	$V_{OC}$ : 0.92 V; $J_{SC}$ : 21.72 mA cm <sup>-2</sup> ; FF: 0.62; PCE: 12.41%.	This study
ITO/PEDOT/MAPbI <sub>3</sub> :CDHC/PC <sub>61</sub> BM/Ag	$V_{OC}$ : 0.96 V; $J_{SC}$ : 17.73 mA cm <sup>-2</sup> ; FF: 0.61; PCE: 10.38%.	Cellulose, 2019, 26, 9229–9239.
ITO/PEDOT/MAPbI <sub>3</sub> /PC <sub>61</sub> BM/Al	$V_{OC}$ : 0.78 V; $J_{SC}$ : 13.2 mA cm <sup>-2</sup> ; FF: 0.60; PCE: 6.2%.	Nanoscale, 2014, 6, 11403–11410.
ITO/PEDOT/MAPbI <sub>3</sub> :NH <sub>4</sub> Cl/PC <sub>61</sub> BM/Al	$V_{OC}$ : 0.88 V; $J_{SC}$ : 14.08 mA cm <sup>-2</sup> ; FF: 0.80; PCE: 9.93%.	Nanoscale, 2014, 6, 9935–9938.
ITO/PEDOT/MAPbI <sub>3</sub> :PEOXA/PC <sub>61</sub> BM/Al	$V_{OC}$ : 1.04 V; $J_{SC}$ : 8.85 mA cm <sup>-2</sup> ; FF: 0.65; PCE: 6.16%.	RSC Adv., 2015, 5, 775–783.
ITO/PEDOT/MAPbI <sub>3</sub> /PC <sub>61</sub> BM/Al	$V_{OC}$ : 0.88 V; $J_{SC}$ : 14.16 mA cm <sup>-2</sup> ; FF: 0.60; PCE: 7.6%.	Solar Energy Mater. Solar Cells, 2016, 155, 166–175.
ITO/PEDOT/MAPbI <sub>3</sub> /PC <sub>61</sub> BM/Al	$V_{OC}$ : 0.87 V; $J_{SC}$ : 11.4 mA cm <sup>-2</sup> ; FF: 0.78; PCE: 7.79%.	ACS Appl. Mater. Interfaces, 2017, 9, 32957–32964.
ITO/PEDOT/MAPbI <sub>3</sub> /PC <sub>61</sub> BM/Ag	$V_{OC}$ : 0.75 V; $J_{SC}$ : 13.76 mA cm <sup>-2</sup> ; FF: 0.40; PCE: 4.13%.	J. Mater. Chem. A, 2017, 5, 12811–12821.

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ITO/PEDOT/MAPbI<sub>3</sub>/PC<sub>61</sub>BM/Ag       $V_{OC}$ : 0.95 V;  $J_{SC}$ : 16.55 mA cm<sup>-2</sup>; FF: 0.59; PCE: 9.29%. ACS Appl. Mater. Interfaces, 2017, 9, 32957–32964.

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