

Electronic Supplementary Information

**Homogeneous Bulk Heterojunction Domains with Continuous Charge Pathway via Hydrophobic Membrane Filtration for Stable Photo and Dark Current Operations**

*Hyunguk Park,<sup>‡a</sup> Jihyun Lim,<sup>‡b</sup> Byung Gi Kim,<sup>b</sup> Woongsik Jang,<sup>b</sup> and Dong Hwan Wang<sup>\*ab</sup>*

<sup>a</sup>School of Integrative Engineering, Chung-Ang University, 84 Heukseok-ro, Dongjak-gu, Seoul 06974, Republic of Korea

<sup>b</sup>Department of Intelligent Semiconductor Engineering, Chung-Ang University, 84 Heukseok-ro, Dongjak-gu, Seoul 06974, Republic of Korea

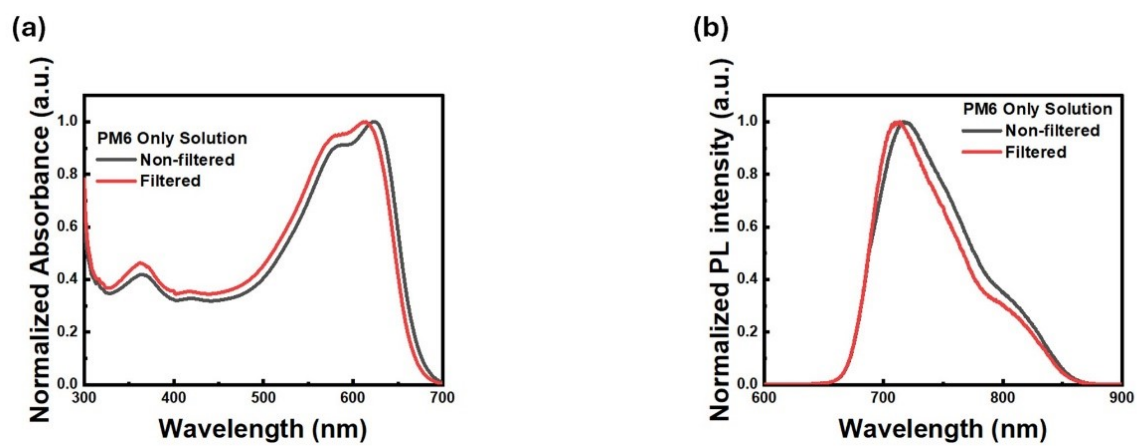
---

\* Corresponding authors, E-mail addresses: king0401@cau.ac.kr (Prof. D.H. Wang)

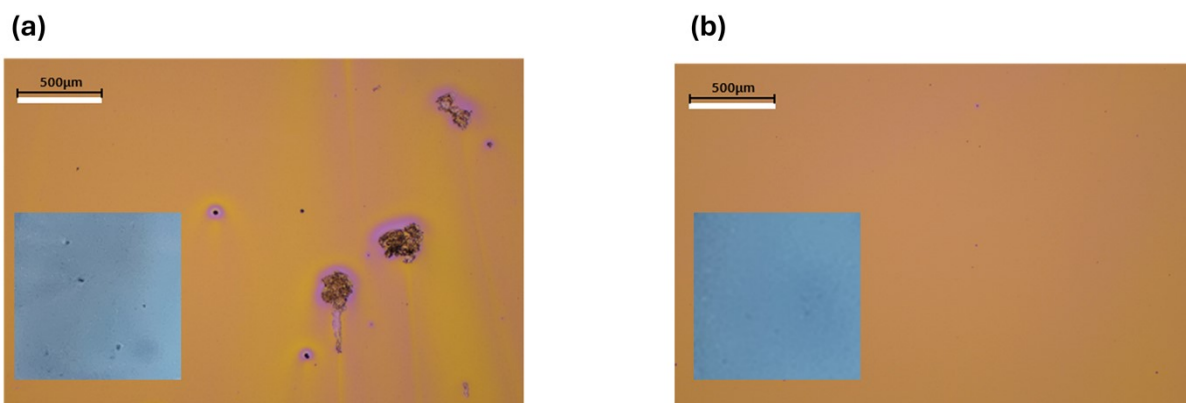
<sup>‡</sup> These authors contributed equally.



**Fig. S1.** Contact angle image of the PM6:Y6 BHJ film.

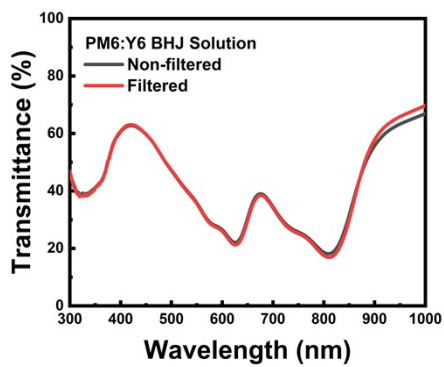


**Fig. S2.** Normalized (a) absorbance and (b) PL intensity spectra of the PM6 solutions with hydrophobic filtration.

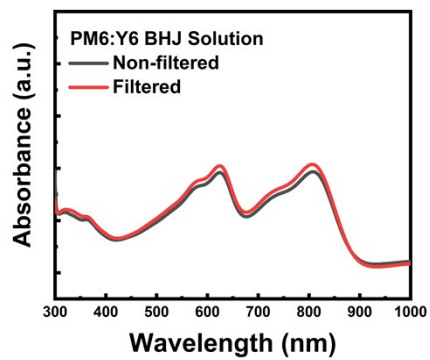


**Fig. S3.** Optical microscopy images of the (a) non-filtered and (b) filtered PM6:Y6 BHJ films (inset: image of the as-cast films).

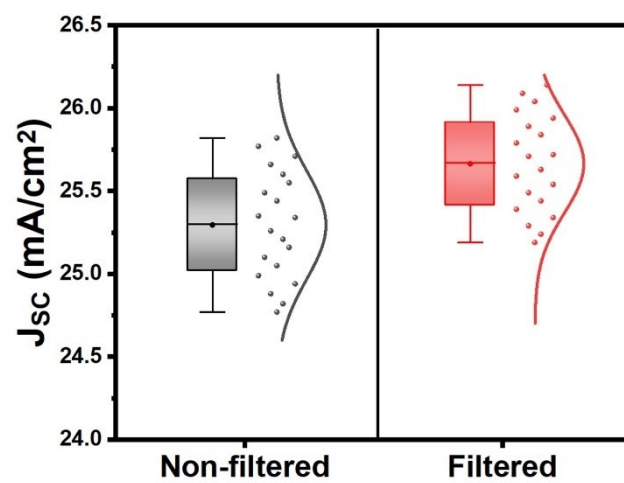
(a)



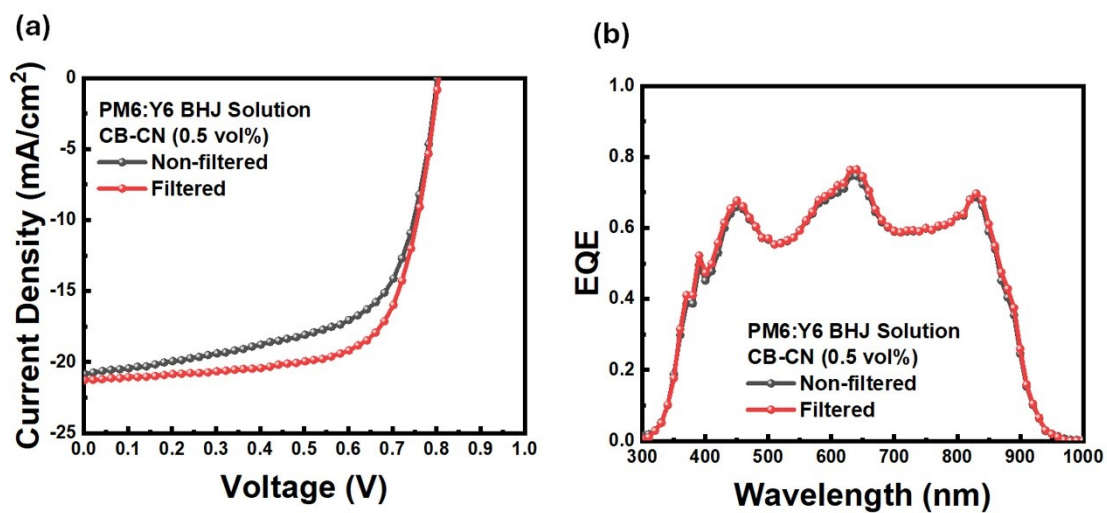
(b)



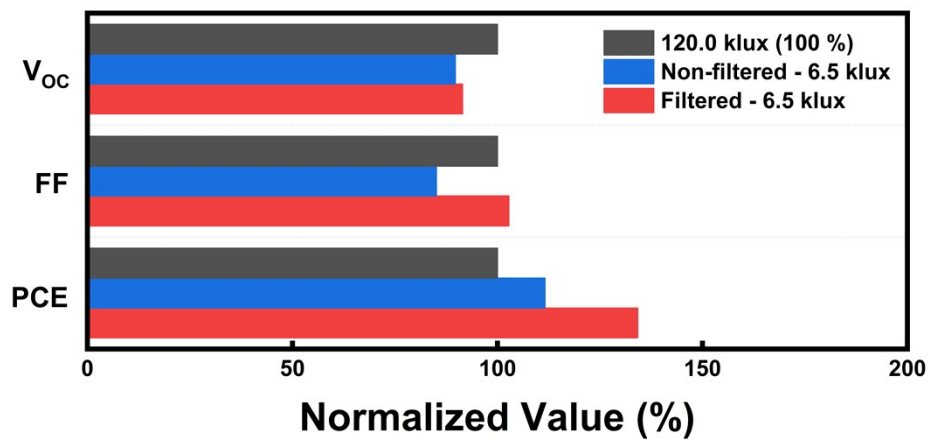
**Fig. S4.** (a) Transmittance and (b) absorbance spectra of the PM6:Y6 BHJ films with BHI hydrophobic filtration.



**Fig. S5.** Variation of the  $J_{SC}$  of OPV at 120.0 klux upon hydrophobic filtration.

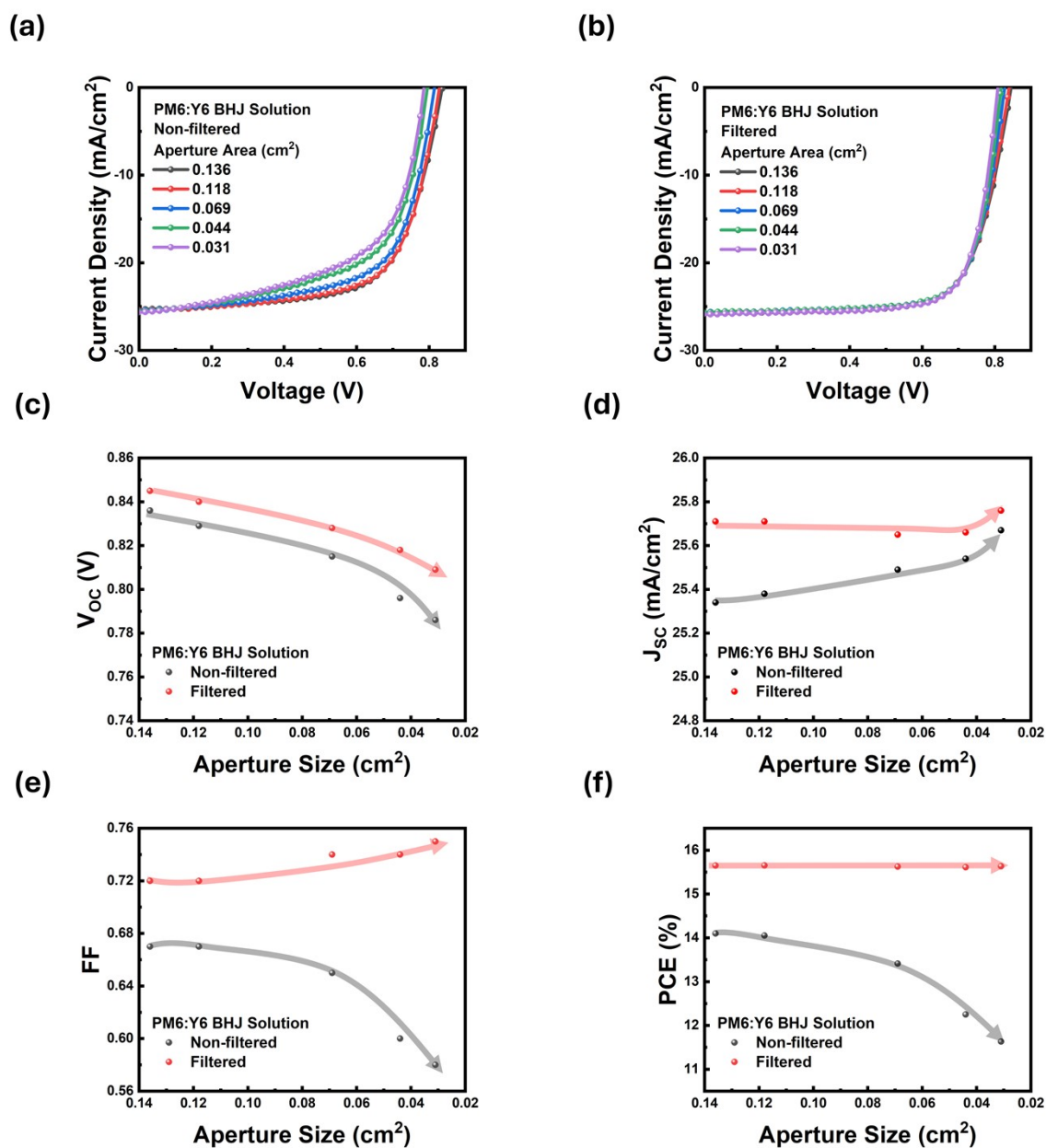


**Fig. S6.** J–V characteristics of CB-CN (0.5%) based OPV with filtration under 120.0 klux illumination.

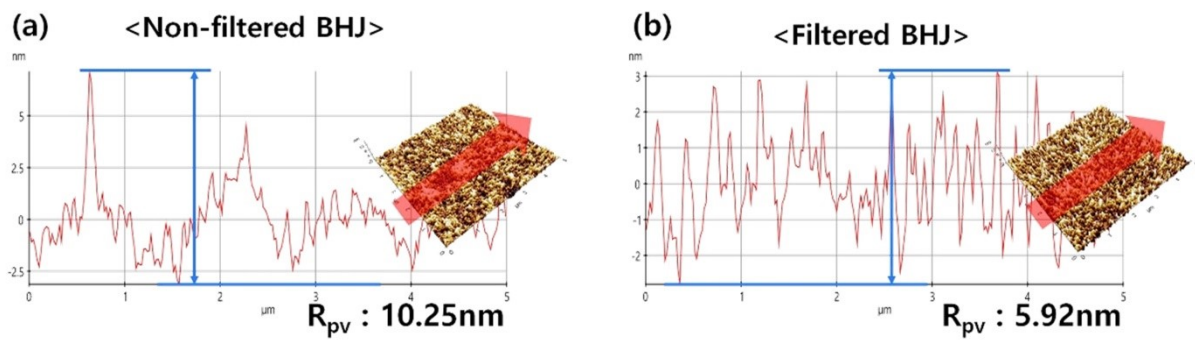


**Fig. S7.** Variation of the OPV parameters at 6.5 klux upon hydrophobic filtration:  $V_{oc}$ , FF and PCE values normalized by 120.0 klux devices.





**Fig. S8.** Photovoltaic characteristics of the non-filtered and filtered devices with a photoactive layer area of (a) 0.031, (b) 0.044, (c) 0.069, and (d) 0.118  $\text{cm}^2$ . Variation of the (e)  $V_{OC}$ , (f)  $J_{SC}$ , (g) FF, and (h) PCE of the OPVs with different photoactive areas.



**Fig. S9.** Line profile and peak-to-valley value from AFM Height image of (a) non-filtered and (b) filtered BHJ film. (inset: direction of line profile)

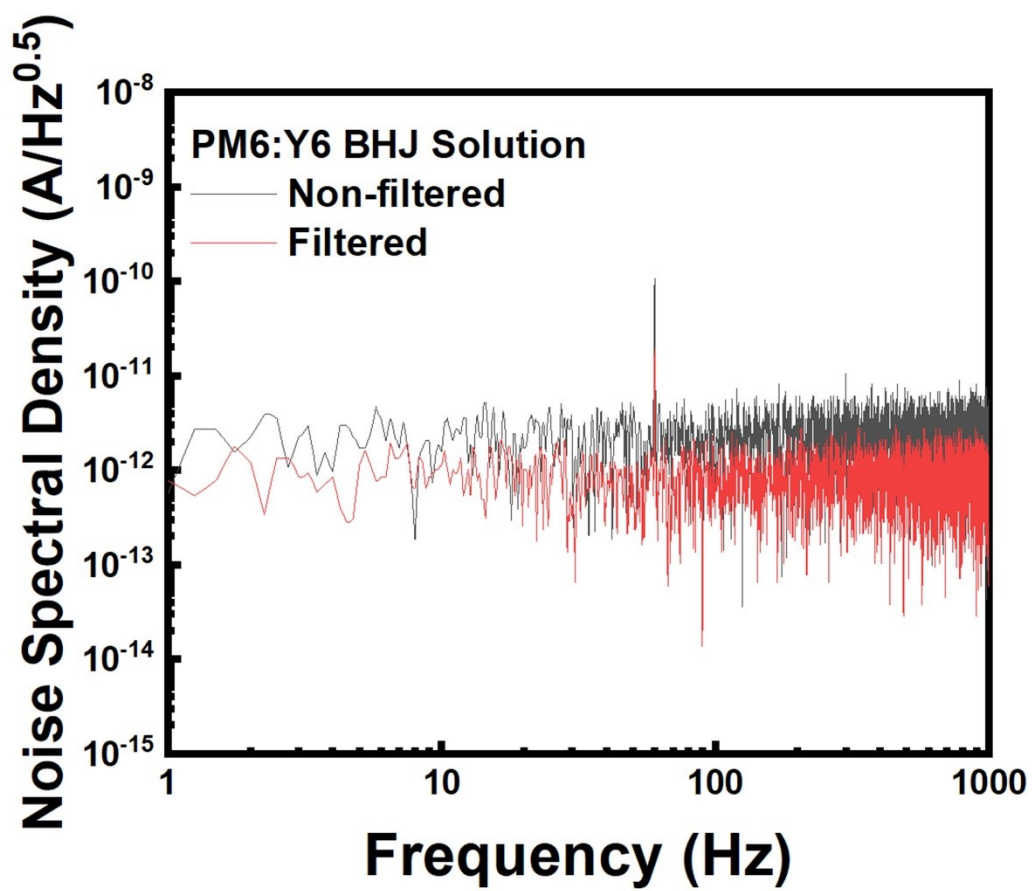
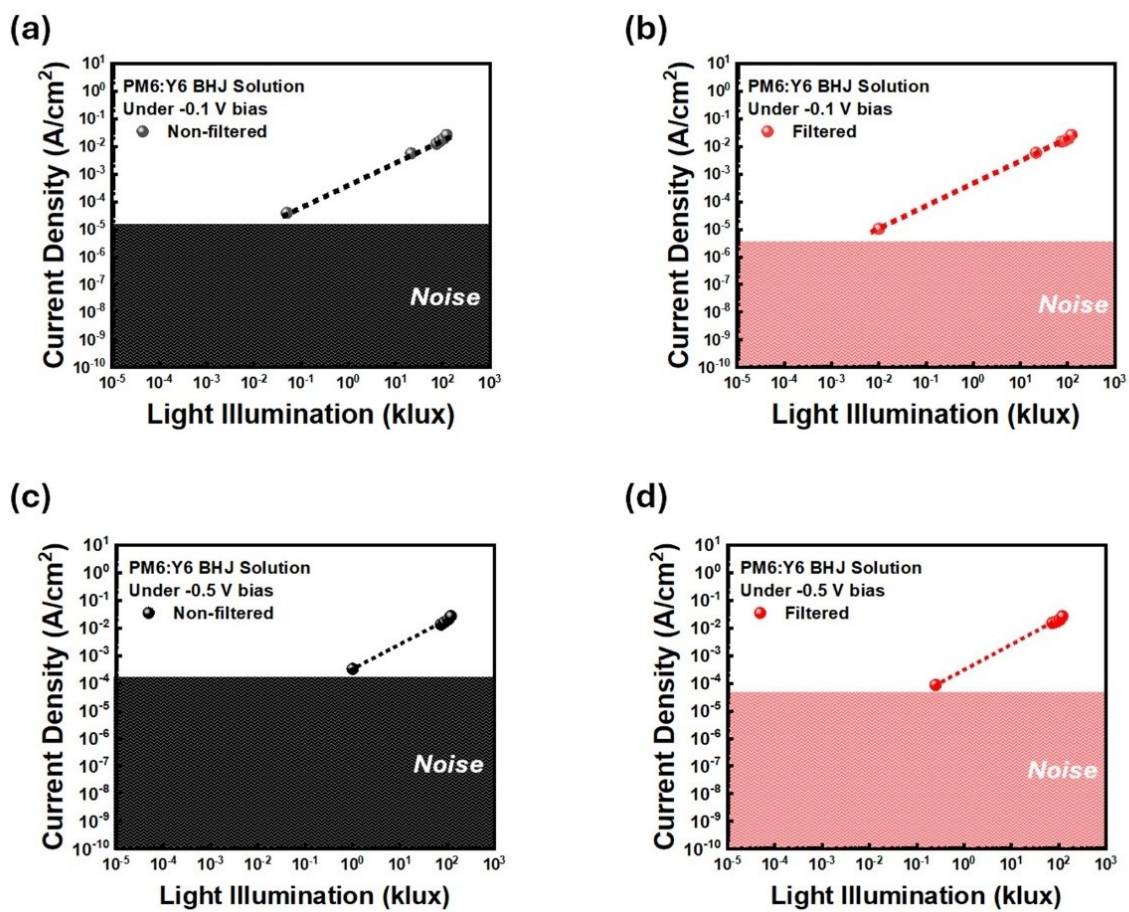
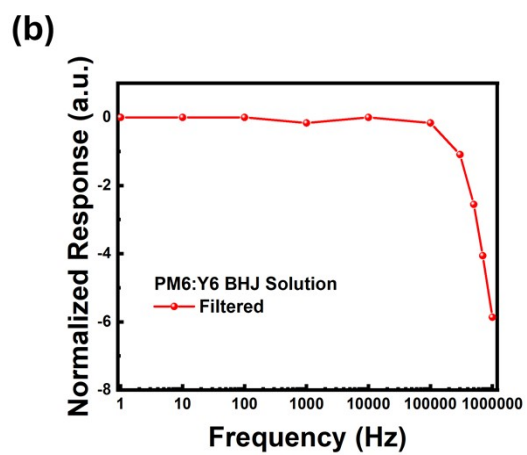
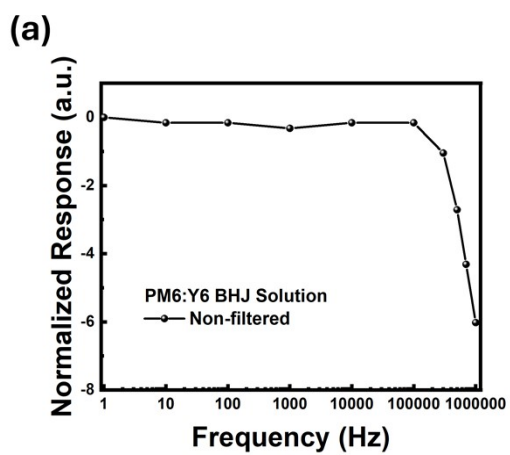


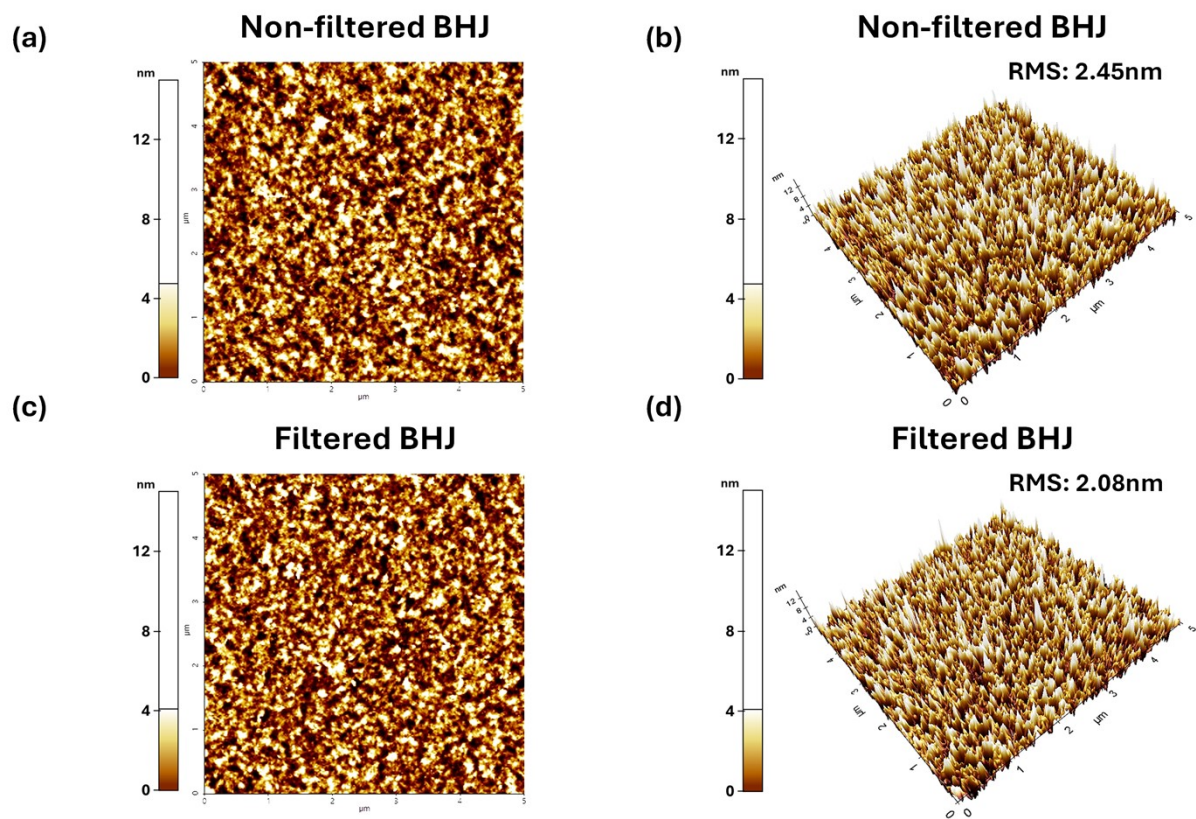
Fig. S10. The noise spectral density of the non-filtered and filtered OPD devices.



**Fig. S11** LDR of the OPD devices under  $-0.1$  V and  $-0.5$  V bias under various light illumination conditions for the (a), (c) non-filtered and (b), (d) filtered devices.



**Fig. S12.** Normalized photo-response at various frequencies of the (a) non-filtered and (b) filtered OPD devices.



**Fig. S13.** 2D and 3D AFM height images of 480 h-aged (a), (b) non-filtered and (c), (d) filtered BHJ films.

**Table S1.** Membrane filter chemical compatibility chart of various solvents for each membrane (C: compatible, IC: incompatible, LC: limited compatible)

<b>Solvent</b>	<b>Membrane filter</b>			
	<b>PTFE</b>	<b>PES</b>	<b>CA</b>	<b>PP</b>
<b>Dimethylsulfoxide</b>	C	IC	IC	C
<b>Methanol</b>	C	C	C	C
<b>Ethyl Ether</b>	C	C	C	LC
<b>Acetone</b>	C	IC	IC	C
<b>Chlorobenzene</b>	C	LC	C	C
<b>Chloroform</b>	C	IC	IC	LC

**Table S2.** Photovoltaic parameters of CB-CN (0.5%) based OPV with filtration under 120.0

<b>PM6:Y6 BHJ</b>	<b>V<sub>OC</sub></b>	<b>J<sub>SC</sub></b>	<b>J<sub>SC,EQE</sub></b>	<b>FF</b>	<b>PCE</b>
<b>Solution (CB-CN)</b>	<b>(V)</b>	<b>(mA/cm<sup>2</sup>)</b>	<b>(mA/cm<sup>2</sup>)</b>		<b>(%)</b>
<b>Non-filtered</b>	0.802	20.86	20.17	0.63	10.45
<b>Filtered</b>	0.805	21.27	20.47	0.69	11.86

klux illumination.



**Table S3.** Photovoltaic characteristics of the OPVs under 6.5 klux illumination with BHJ hydrophobic filtration.

<b>PM6:Y6 Solution</b>	<b>V<sub>oc</sub> (V)</b>	<b>J<sub>sc</sub> (mA/cm<sup>2</sup>)</b>	<b>FF</b>	<b>PCE (%)</b>
<b>Non-filtered</b>	0.750	4.56	0.57	15.73
<b>Filtered</b>	0.774	4.56	0.74	20.96

**Table S4.** Series and shunt resistance of the OPVs under 120.0 and 6.5 klux illumination with BHJ hydrophobic filtration.

<b>PM6:Y6</b>	<b>Light Intensity</b>	<b>R<sub>s</sub></b>	<b>R<sub>sh</sub></b>
<b>Solution</b>	<b>(klux)</b>	<b>(Ω/cm<sup>2</sup>)</b>	<b>(kΩ/cm<sup>2</sup>)</b>
<b>Non-filtered</b>	120.0	4.35	1.16
	6.5	17.50	2.90
<b>Filtered</b>	120.0	4.00	3.71
	6.5	13.46	10.15

**Table S5.** Photovoltaic parameters of OPVs under 120.0 klux illumination for different photoactive layer areas.

<b>PM6:Y6 Solution</b>	<b>Active Area (cm<sup>2</sup>)</b>	<b>V<sub>oc</sub> (V)</b>	<b>J<sub>sc</sub> (mA/cm<sup>2</sup>)</b>	<b>FF</b>	<b>PCE (%)</b>
<b>Non-filtered</b>	0.031	0.786	25.07	0.58	11.36
	0.044	0.796	25.10	0.60	12.04
	0.069	0.815	25.05	0.65	13.18
	0.118	0.829	25.16	0.67	13.93
<b>Filtered</b>	0.031	0.809	25.44	0.75	15.44
	0.044	0.818	25.46	0.74	15.49
	0.069	0.828	25.51	0.74	15.54
	0.118	0.840	25.54	0.72	15.55

**Table S6.** Responsivity (R) and specific detectivity ( $D^*$ ) values under self-powered conditions from voltage-dependent curves of OPDs with hydrophobic-filtration-processed BHJ.

<b>PM6:Y6</b>	<b>Light illumination</b>	<b>R @0 V</b>	<b><math>D^*</math> @0 V</b>
<b>Solution</b>	<b>(klux)</b>	<b>(A/W)</b>	<b>(Jones)</b>
<b>Non-filtered</b>	120	0.25	$3.02 \times 10^{12}$
	6.5	0.37	$4.43 \times 10^{12}$
<b>Filtered</b>	120	0.26	$1.04 \times 10^{13}$
	6.5	0.37	$1.51 \times 10^{13}$

**Table S7.** Calculated trap-filled limited voltage ( $V_{\text{TFL}}$ ) and trap density ( $N_t$ ) of the optoelectronic devices with hydrophobic filtration.

<b>PM6:Y6</b>	<b><math>V_{\text{TFL}}</math></b>	<b><math>N_t</math></b>
<b>Solution</b>	<b>(V)</b>	<b>(#/cm<sup>3</sup>)</b>
<b>Non-filtered</b>	0.132	$3.62 \times 10^{15}$
<b>Filtered</b>	0.117	$3.21 \times 10^{15}$

**Table S8.** -3 dB frequency of the OPD devices with hydrophobic filtration.

<b>PM6:Y6</b>	<b>-3 dB frequency</b>
<b>Solution</b>	<b>(kHz)</b>
<b>Non-filtered</b>	537
<b>Filtered</b>	545