## **Supplementary Information**

## Performance and Stability Enhancement of Perovskite Photodetectors by Additive and Interface Engineering Using a Dual-Functional PPS Zwitterion

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Fig. S1: Detailed fabrication process



Fig. S2: Cross-sectional SEM image of device without PPS inerlayer



Fig. S3: Top-view SEM images of  $SnO_2$  (a) without PPS (b) with PPS



**Fig S4:** Large area too-view SEM images of perovskite films at (a) Control 0% (b) 0.2% PPS (c) 0.4% PPS (c) 0.4% PPS (d) 0.6% PPS



Fig S5: Ratio of (110) peak intensity of perovskite with various PPS concentration.



Fig S6: Tauc plot of (a) CsMAPbI<sub>3</sub> (b) 0.2% PPS (c) 0.4% PPS (d) 0.6% PPS



**Fig S7:** (a) XPS spectra of S 2p of SnO<sub>2</sub> and SnO<sub>2</sub>/PPS layers. (b) FTIR spectra of PPS and PPS zwitterion incorporated perovskite.



Fig S8: Deconvoluted C 1s and (d) deconvoluted O 1s spectra of Perovskite.



Fig S9: Cross-sectional SEM Images with the thickness (a) control and (b) 0.4% PPS incorporated perovskite films



Fig S10: (a) PL spectra of complete devices (b) TRPL spectra of complete devices



**Fig S11:** Perovskite only devices at different concentrations (a) dark current (b) photocurrent. PPS incorporated Perovskite devices with  $SnO_2$  layer (c) dark current (d) photocurrent



Fig S12: Different interlayer concentration of PPS (a) dark current (b) photocurrent





W/O PPS - 20 Days



W/O PPS - Fresh

With PPS - 20 Days

Fig S13: Sample stability images with and without PPS for over 30 days.

With PPS - Fresh



W/O PPS – 30 Days





With PPS - 30 Days





With PPS - 4 hrs, T=130° C



With PPS - 24 hrs, T=130° C



Fig S14: Thermal stability of samples with and without PPS

	FWHM	Crystallite Size (nm)
Control	0.1359	65.5
0.2%	0.1374	64.80
0.4%	0.1385	64.28
0.6%	0.1404	63.32

Table S1: FWHM and Crystallite size of (110) peak of perovskite with different concentration of PPS.

**Table S2:** FWHM of control and with PPS incorporated perovskite films.

Peak	FWHM of Control	FWHM of Psk:PPS (0.4%)
(110)	0.1359	0.1374
(220)	0.1705	0.1682
(310)	0.1913	0.1872

Table S3: Atomic concentration of the elements.

Atom	Atomic % in PSk	Atomic% in PSK:PPS
С	28.09	24.54
0	6.15	4.46
Ι	53.01	57.8
Pb	12.75	13.2

**Table S4:** Fitted parameters of TRPL decay in figure 5b.

Film	А	$ au_1$ (ns)	В	$\tau_{2}(ns)$	$ au_{avg}$ (ns)
Psk	0.78663	2.92794	0.56691	11.95355	9.664089
Psk:PPS	0.48183	23.87487	0.14917	78.20074	51.22724

Table S5: Fitted parameters of TRPL decay in figure S10(b)

Film	Α	$ au_1$ (ns)	В	$\tau_2$ (ns)	$\tau_{avg}$ (ns)
SnO2/Psk:PPS	141.41	164	1772.6	26.6	71.89951
SnO2/PPS(0.2%)/Psk:PPS	269.8	138	3059.3	35.5	61.6683

	I <sub>L</sub> (A)	I <sub>D</sub> (A)	Respo nsivity A/W	On/off	Detectivit y (J)	Rise/Dec ay time (sec)
SnO <sub>2</sub> /Psk	$4.08 \times 10^{-7}$	$1.32 \times 10^{-10}$	3.26	3.0×10 <sup>3</sup>	5.62×10 <sup>12</sup>	1.00/1.12
SnO <sub>2</sub> /Psk:PPS(0.4%)	$6.58 \times 10^{-7}$	8.12×10 <sup>-11</sup>	5.27	8.1×10 <sup>3</sup>	1.15×10 <sup>13</sup>	0.98/0.73
SnO <sub>2</sub> /PPS(0.2%)/Psk:PPS(0. 4%)	7.41×10 <sup>-7</sup>	6.05×10 <sup>-11</sup>	5.93	1.2×10 <sup>4</sup>	1.51×10 <sup>13</sup>	0.74/0.09

 Table S6: SnO<sub>2</sub>/Psk, SnO<sub>2</sub>/Psk:PPS(0.4%), and SnO<sub>2</sub>/PPS(0.2%)/Psk:PPS(0.4%) Photodetector device results obtained from I-V.

**Table S7:** The photodetector device results obtained by performing I-V measurements at various concentrations of PPS incorporation in the perovskite layer and on bare SnO<sub>2</sub>.

	I <sub>L</sub> (A)	I <sub>D</sub> (A)	Responsivity A/W	On/off	Detectivity (J)
SnO <sub>2</sub> /Psk	4.08×10 <sup>-7</sup>	$1.32 \times 10^{-10}$	3.26	3.0×10 <sup>3</sup>	5.62×10 <sup>12</sup>
SnO <sub>2</sub> /Psk:PPS(0.2%)	5.06×10 <sup>-7</sup>	9.69×10 <sup>-11</sup>	4.04	5.2×10 <sup>3</sup>	8.12×10 <sup>12</sup>
SnO <sub>2</sub> /Psk:PPS(0.4%)	$6.58 \times 10^{-7}$	8.12×10 <sup>-11</sup>	5.27	8.1×10 <sup>3</sup>	1.15×10 <sup>13</sup>
SnO <sub>2</sub> /Psk:PPS(0.6%)	4.64×10 <sup>-7</sup>	9.15×10 <sup>-11</sup>	3.71	5.0×10 <sup>3</sup>	7.66×10 <sup>12</sup>

**Table S8:** The photodetector device results obtained by performing I-V measurements at various concentrations of PPS interlayer with pristine perovskite.

	I <sub>L</sub> (A)	I <sub>D</sub> (A)	Responsivity A/W	On/off	Detectivity (J)
SnO <sub>2</sub> /Psk	$4.08 \times 10^{-7}$	$1.32 \times 10^{-10}$	3.26	3.0×10 <sup>3</sup>	$5.62 \times 10^{12}$
SnO <sub>2</sub> /PPS(0.1%)/Psk	$4.55 \times 10^{-7}$	$9.42 \times 10^{-11}$	3.64	$4.8 \times 10^{3}$	$7.40 \times 10^{12}$
SnO <sub>2</sub> /PPS(0.2%)/Psk	$4.64 \times 10^{-7}$	$8.67 \times 10^{-11}$	3.71	5.3×10 <sup>3</sup>	$7.88 \times 10^{12}$
SnO <sub>2</sub> /PPS(0.4%)/Psk	$4.49 \times 10^{-7}$	$1.08 \times 10^{-10}$	3.59	4.1×10 <sup>3</sup>	$6.82 \times 10^{12}$

**Table S9:** The photodetector device results obtained by performing I-V measurements at various concentrations of PPS interlayer with PPS incorporated (0.4%) perovskite.

	I <sub>L</sub> (A)	I <sub>D</sub> (A)	Responsi vity A/W	On/off	Detectivit y (J)
SnO <sub>2</sub> /Psk	$4.08 \times 10^{-7}$	$1.32 \times 10^{-10}$	3.26	3.0×10 <sup>3</sup>	$5.62 \times 10^{12}$
SnO <sub>2</sub> /Psk:PPS(0.4%)	$6.58 \times 10^{-7}$	$8.12 \times 10^{-11}$	5.27	8.1×10 <sup>3</sup>	$1.15 \times 10^{13}$
SnO <sub>2</sub> /PPS(0.1%)/Psk:PPS(0.4%)	$7.01 \times 10^{-7}$	$7.17 \times 10^{-11}$	5.60	9.7×10 <sup>3</sup>	$1.31 \times 10^{13}$
SnO <sub>2</sub> /PPS(0.2%)/Psk:PPS(0.4%)	$7.41 \times 10^{-7}$	$6.05 \times 10^{-11}$	5.93	$1.2 \times 10^{4}$	$1.51 \times 10^{13}$
SnO <sub>2</sub> /PPS(0.4%)/Psk:PPS(0.4%)	$5.31 \times 10^{-7}$	$9.71 \times 10^{-11}$	4.24	5.4×10 <sup>3</sup>	$8.52 \times 10^{12}$