

## Ionic liquid as modulate interface for high-efficient and stable perovskite solar cells

Xiang Chen,<sup>a</sup> Lixin Song,<sup>\*a</sup> Ningxia Gu,<sup>a</sup> Pengyun Zhang,<sup>a</sup> Lei Ning,<sup>a</sup> Pingfan Du,<sup>a</sup> Fengfeng Chen,<sup>b</sup> and Jie Xiong<sup>\*a, b</sup>

*a* College of Textile Science and Engineering, *b* School of Materials Science & Engineering,  
Zhejiang Sci-Tech University, Hangzhou, 310018, China. E-mail: [jxiong@zstu.edu.cn](mailto:jxiong@zstu.edu.cn),  
[lsxong@zstu.edu.cn](mailto:lsxong@zstu.edu.cn)

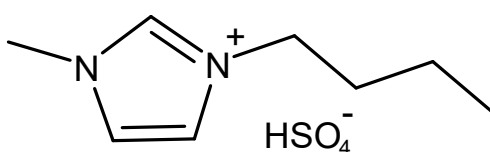


Fig.S1 Chemical structure of BMIMHSO<sub>4</sub> molecule.

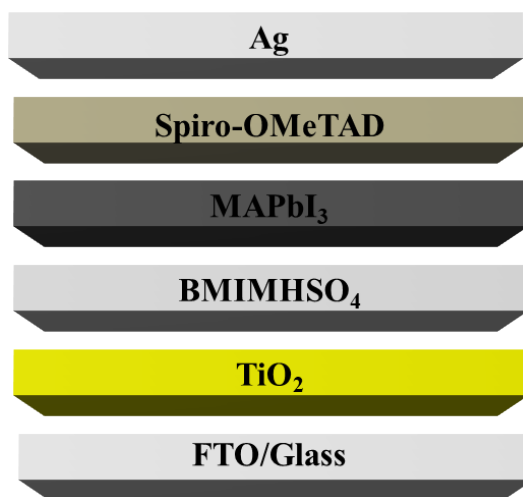


Fig.S2 Device structure of the PSCs.

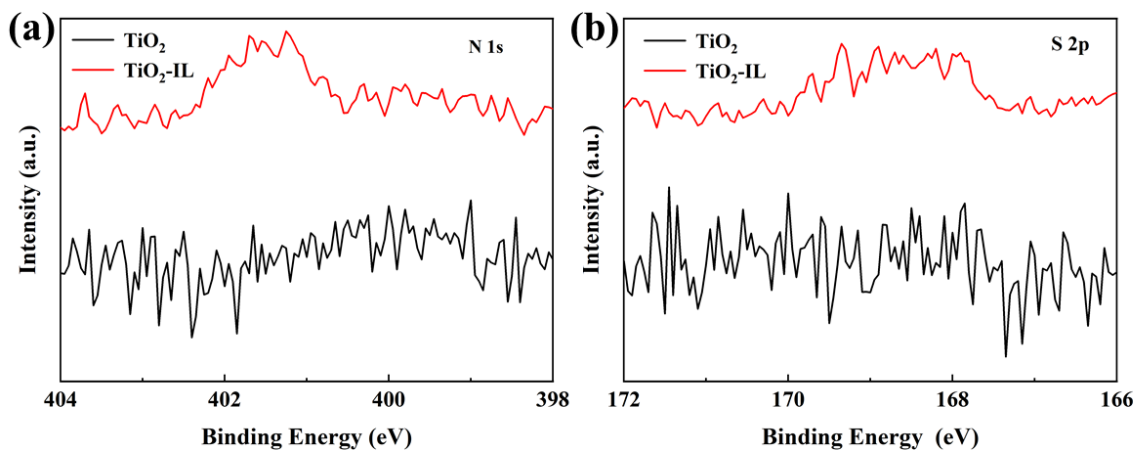


Fig.S3 The high-resolution XPS spectra of (a) N and (b) S element.

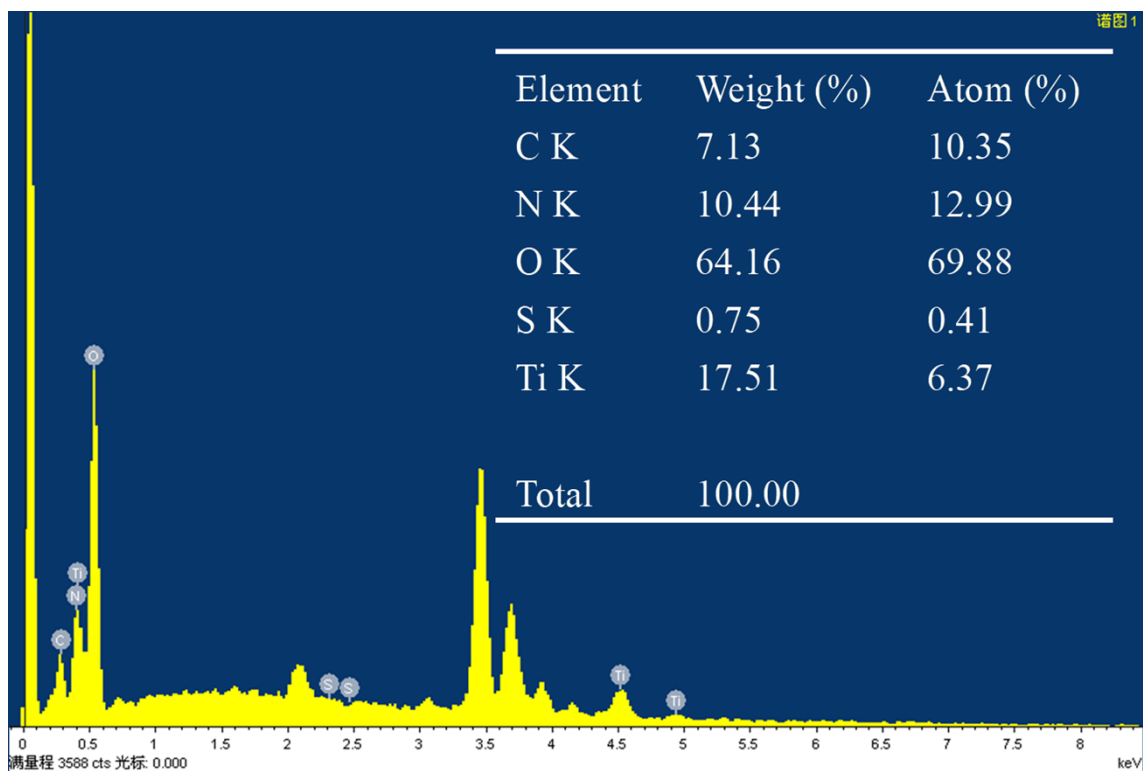


Fig.S4 The EDS measurement of TiO<sub>2</sub>-IL sample with the top-right showing the detail element analysis.

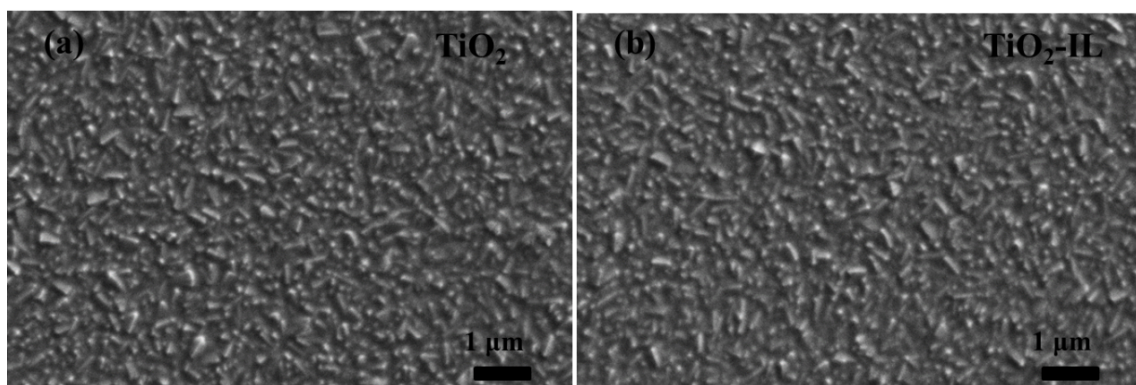


Fig.S5 The SEM images of (a) TiO<sub>2</sub> and (b) TiO<sub>2</sub>-IL film.

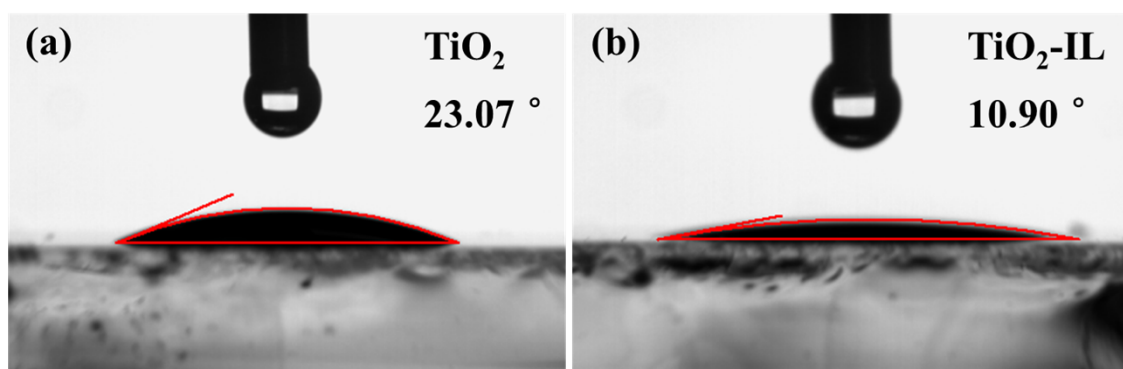


Fig.S6 Contact angle performance using DMSO solvent of (a) TiO<sub>2</sub> and (b) TiO<sub>2</sub> film.

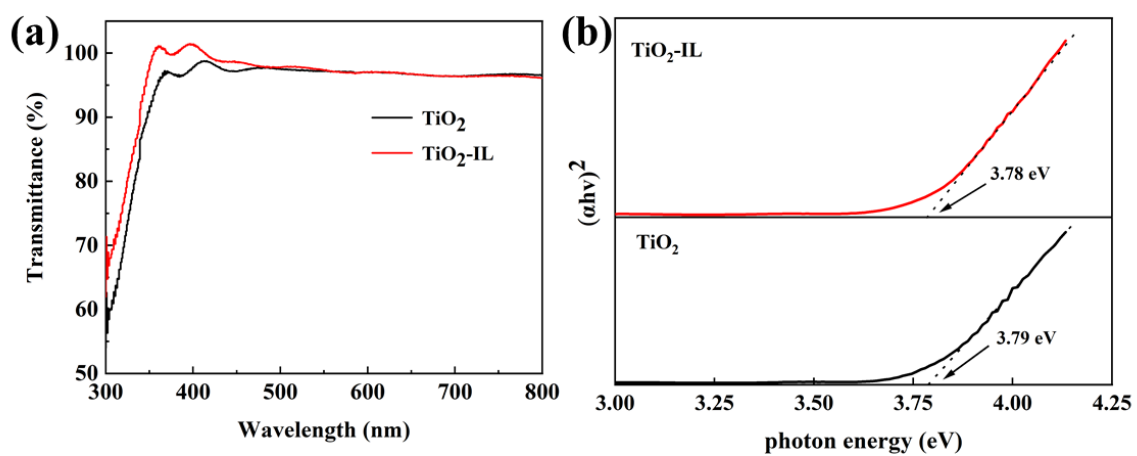


Fig.S7 (a)Transmission spectra and (b) Tauc plot of absorption spectra of TiO<sub>2</sub> and TiO<sub>2</sub>-IL film.

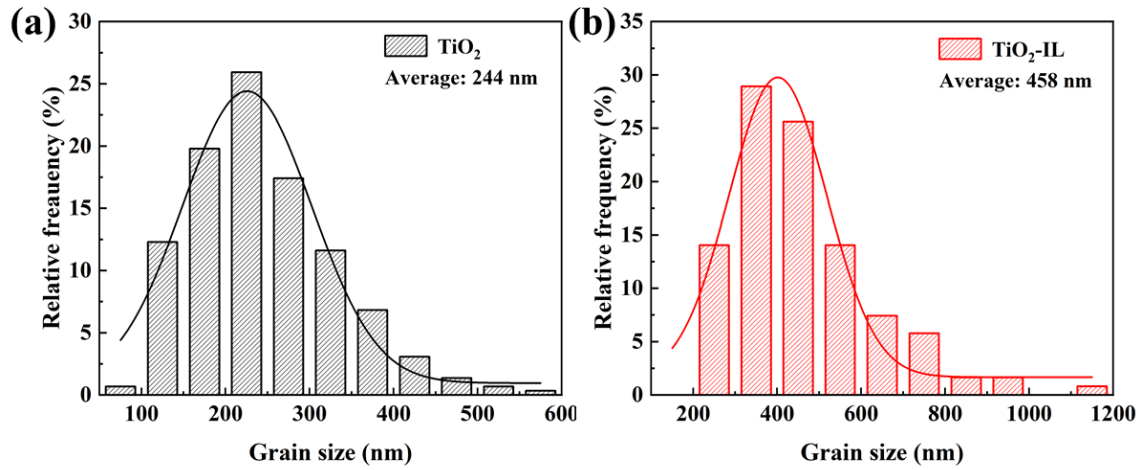


Fig.S8 The grain size distribution histograms of (a) TiO<sub>2</sub>-based and (b) TiO<sub>2</sub>-IL-based perovskite film.

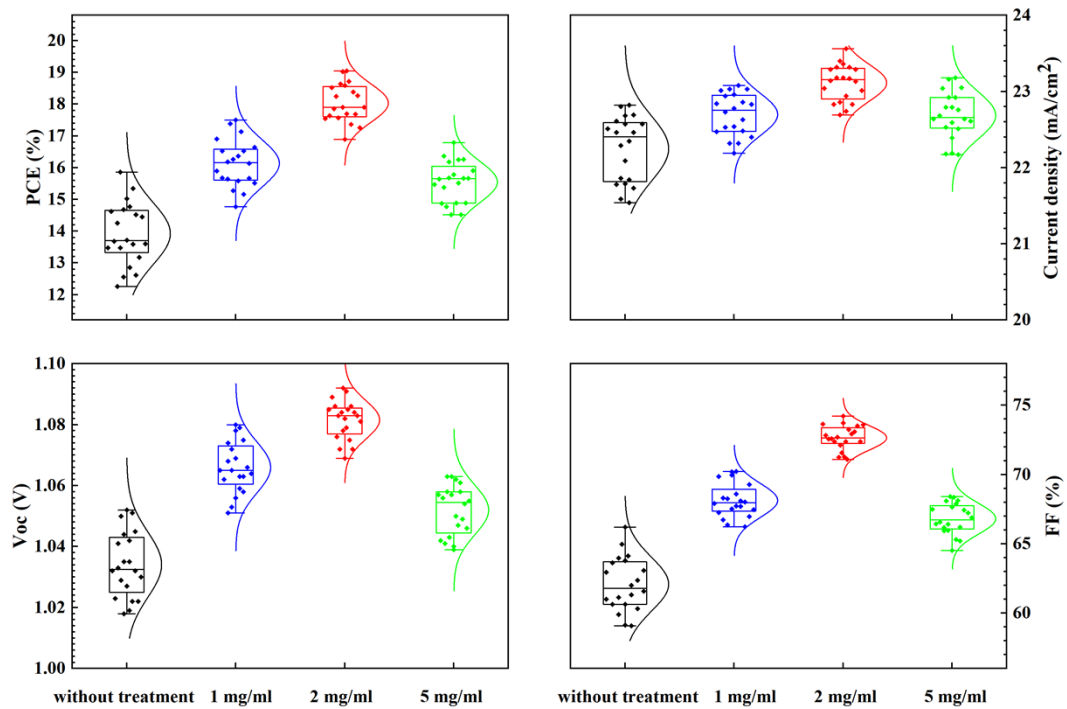


Fig.S9 Statistics on photovoltaic parameters for studied PSCs at various IL concentrations. J-V curves collected at 1 sun irradiation are utilized to derive parameters. A box is used to illustrate the standard error. Twenty devices with a cell active area of 0.06 cm<sup>2</sup> were fabricated for each PSC structure.

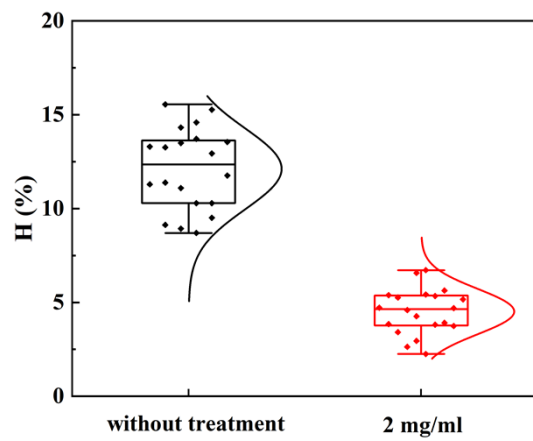


Fig.S10 Hysteresis index of twenty devices.

Table S1 The electron mobility and conductivity parameters based on two ETL.

Samples	Mobility ( $\text{cm}^2 \text{V}^{-1} \text{S}^{-1}$ )	Conductivity ( $\text{mS cm}^{-1}$ )
TiO <sub>2</sub>	$1.43 \times 10^{-7}$	$1.27 \times 10^{-3}$
TiO <sub>2</sub> -IL	$4.88 \times 10^{-7}$	$1.93 \times 10^{-3}$

Table S2 Photovoltaic parameters of PSCs based on TiO<sub>2</sub>-IL with different concentrations.

Samples	Jsc ( $\text{mA cm}^{-2}$ )	Voc (V)	FF (%)	PCE (%)
Without treatment	22.82	1.05	66.20	15.92
1 mg/ml	23.08	1.08	70.23	17.56
2 mg/ml	23.56	1.09	74.21	19.13
5 mg/ml	23.18	1.06	68.36	16.83

Table S3 Photovoltaic parameters measured at different scan directions of the champion PSCs based on the 0 mg/ml and 2 mg/ml IL concentration devices.

Samples	Scan direction	Jsc ( $\text{mA cm}^{-2}$ )	Voc (V)	FF (%)	PCE (%)	H (%)
Without treatment	Reverse	22.82	1.05	66.20	15.92	8.70
	Forward	22.55	1.04	61.76	14.53	
2 mg/ml	Reverse	23.56	1.09	74.21	19.13	2.40
	Forward	23.45	1.09	72.86	18.67	

Table S4. The fitting parameters of TRPL of perovskite film for growing on TiO<sub>2</sub> and TiO<sub>2</sub>-IL substrates.

Samples	A <sub>1</sub>	τ <sub>1</sub> (ns)	A <sub>2</sub>	τ <sub>2</sub> (ns)	τ <sub>avg</sub> (ns)
TiO <sub>2</sub>	1.12	6.53	0.25	212.79	187.86
TiO <sub>2</sub> -IL	1.89	2.64	0.57	23.55	17.88

Table S5 the electron trap density was calculated by SCLC measures of the deposited on TiO<sub>2</sub> film and TiO<sub>2</sub>-IL film devices.

Samples	V <sub>TFL</sub> (V)	N <sub>trap</sub> (cm <sup>-3</sup> )
TiO <sub>2</sub>	0.683	1.20 × 10 <sup>14</sup>
TiO <sub>2</sub> -IL	0.475	0.83 × 10 <sup>14</sup>

Table S6 EIS parameters of PSCs based on TiO<sub>2</sub> and TiO<sub>2</sub>-IL devices.

Samples	R <sub>s</sub> (Ω)	R <sub>rc</sub> (Ω)
TiO <sub>2</sub>	30.48	12388.32
TiO <sub>2</sub> -IL	22.31	17704.03

Table S7 summarized the stability research about BMIMHSO<sub>4</sub> IL with other ILs.

Ionic liquid	Perovskite	PCE (%)	Decrease in stability (%) after hours	Ref

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MAAc	CsPbIBr <sub>2</sub>	8.85	Air( room temperature), remains 82% of initial PCE after 30 days.	1
BMIMPF <sub>6</sub>	CsPbI <sub>2</sub> Br	13.19	Air( 20 °C, ~20% RH), remains 91% of initial PCE after 60 days.	2
FBABF <sub>4</sub>	(FAPbI <sub>3</sub> ) <sub>1-x</sub> (MAPbBr <sub>3</sub> ) <sub>x</sub>	23.07	Air( 35±5% RH), remains 85% of initial PCE after 3000h.	3
N(CH <sub>3</sub> ) <sub>4</sub> OH (TMAH)	FA <sub>0.75</sub> MA <sub>0.25</sub> PbI <sub>2.5</sub> Br <sub>0.5</sub>	20.28	Desiccator( ~15% RH), remains 97% of initial PCE after 360h.	4
BMIMBF <sub>4</sub>	MAPbI <sub>3</sub>	19.62	Air( AM 1.5G illumination), remains 85% of initial PCE after 240 min.	5
BMIMBF <sub>4</sub>	FA <sub>0.83</sub> MA <sub>0.17</sub> Pb(I <sub>0.83</sub> Br <sub>0.17</sub> ) <sub>3</sub>	20.80	Air( 85 °C, dark, under 45% RH), remains less than 50% of initial PCE after 800h.	6
EMIMI	MAPbI <sub>3</sub>	14.59	Dry air( RH<5%), remains 93% of initial PCE after 360h; air ( 30-40% RH), remains 51% of initial PCE after 360h.	7
[EMIM]PF <sub>6</sub>	MAPbI <sub>3</sub>	13.50	Air, remains 93.5% of initial PCE after 45 days.	8
MAAc	MAPbI <sub>3</sub>	21.08	Light, N <sub>2</sub> -filled glovebox, remains 86% of initial PCE after 400h.	9
BMIMHSO <sub>4</sub>	MAPbI <sub>3</sub>	19.13	Air( ~40% RH, 25±5°C, dark), remains 90% of initial PCE after 600h.	This work

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