

## Supporting Information

# Aqueous Solution Processable Metal Oxides For High-Performance Organic and Perovskite Solar Cells

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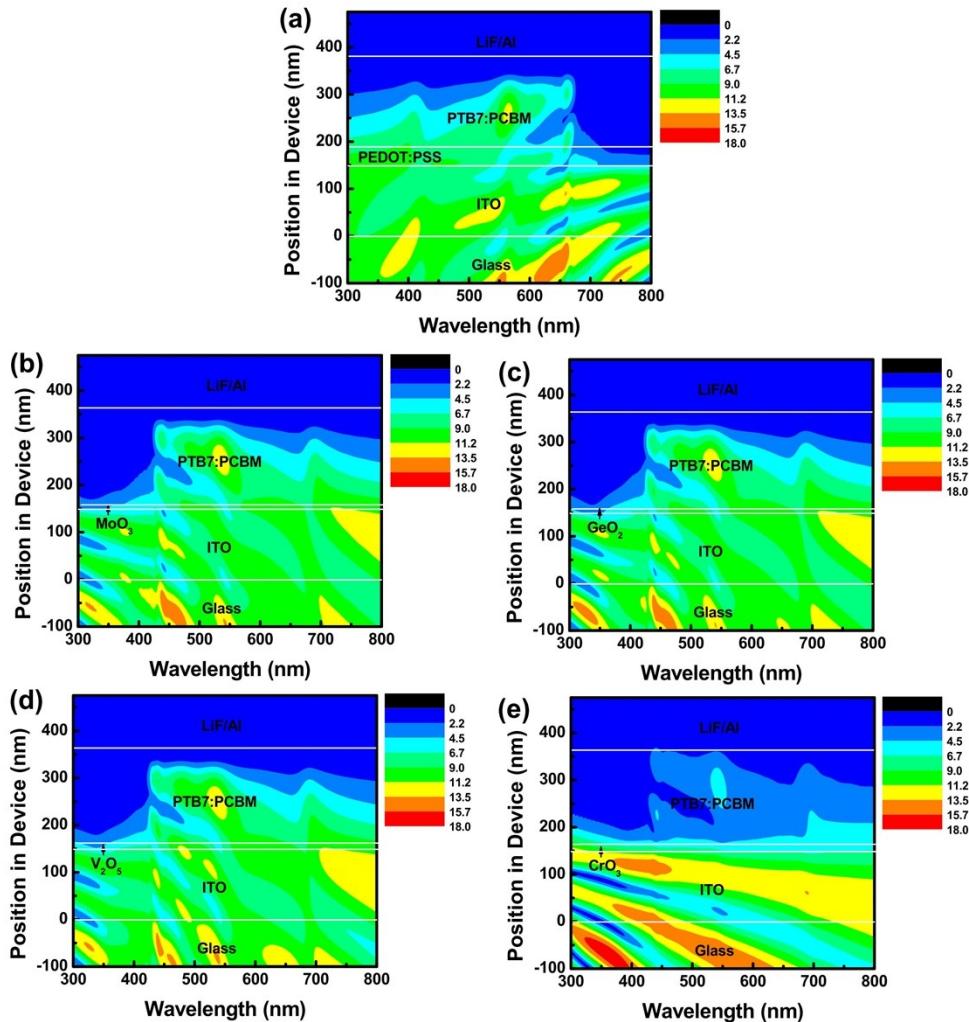
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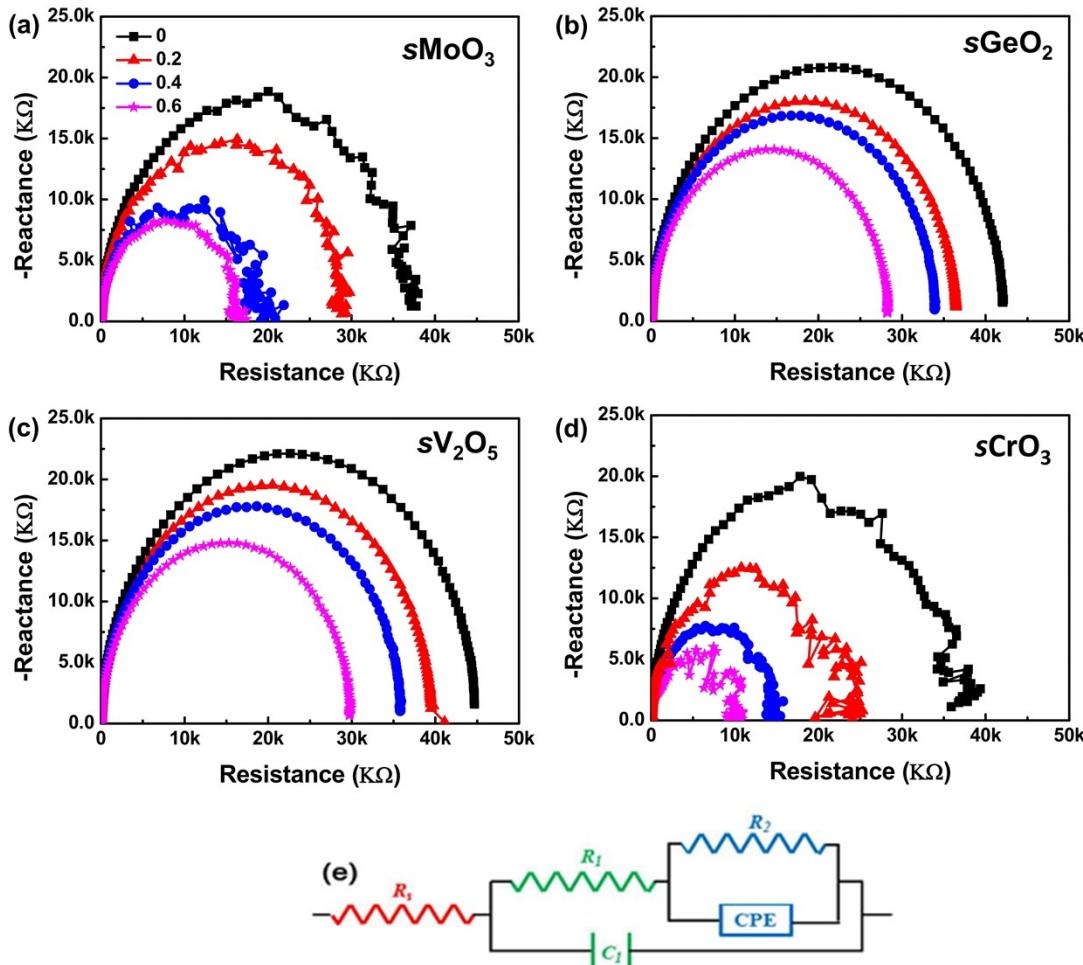


**Figure S1.** Optical field ( $|E|^2$ ) distributions in PTB7:PC<sub>71</sub>BM based bulk heterojunction solar cells incorporating with differnt hole transporting layers of (a) PEDOT:PSS, (b) sMoO<sub>3</sub>, (c) sGeO<sub>2</sub>, (d) sV<sub>2</sub>O<sub>5</sub> and (e) sCrO<sub>3</sub>. Calculation were carried out under the consideration of the whole visible light range (300~800 nm).

## Impedance Spectra Evaluations of the Anode Interfaces

### Device structure:

ITO/TMOs ( $s\text{MoO}_3$ ,  $s\text{GeO}_2$ ,  $s\text{V}_2\text{O}_5$ , or  $s\text{CrO}_3$ )/PTB7:PC<sub>71</sub>BM/MoO<sub>3</sub>/Al

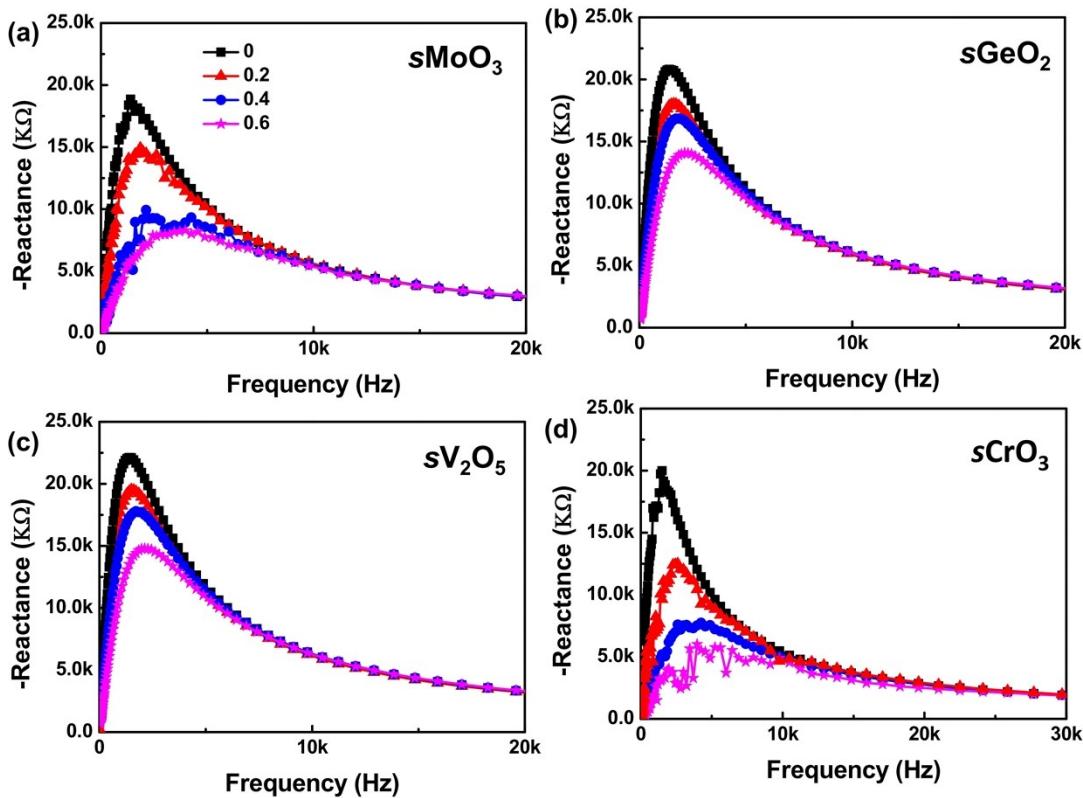


**Figure S2.** Impedance response in PTB7:PC<sub>71</sub>BM based hole-dominant devices incorporating with different aqueous solution processed hole transporting layers (a)  $s\text{MoO}_3$ , (b)  $s\text{GeO}_2$ , (c)  $s\text{V}_2\text{O}_5$  and (d)  $s\text{CrO}_3$ . (e) Equivalent circuit used to fit the data in (a)- (d).  $R_s$  represents the resistive losses in the ITO and sTMOs.

## Impedance Spectra Evaluations of the Anode Interfaces

### Device structure:

ITO/TMOs ( $s\text{MoO}_3$ ,  $s\text{GeO}_2$ ,  $s\text{V}_2\text{O}_5$ , or  $s\text{CrO}_3$ )/PTB7:PC<sub>71</sub>BM/MoO<sub>3</sub>/Al

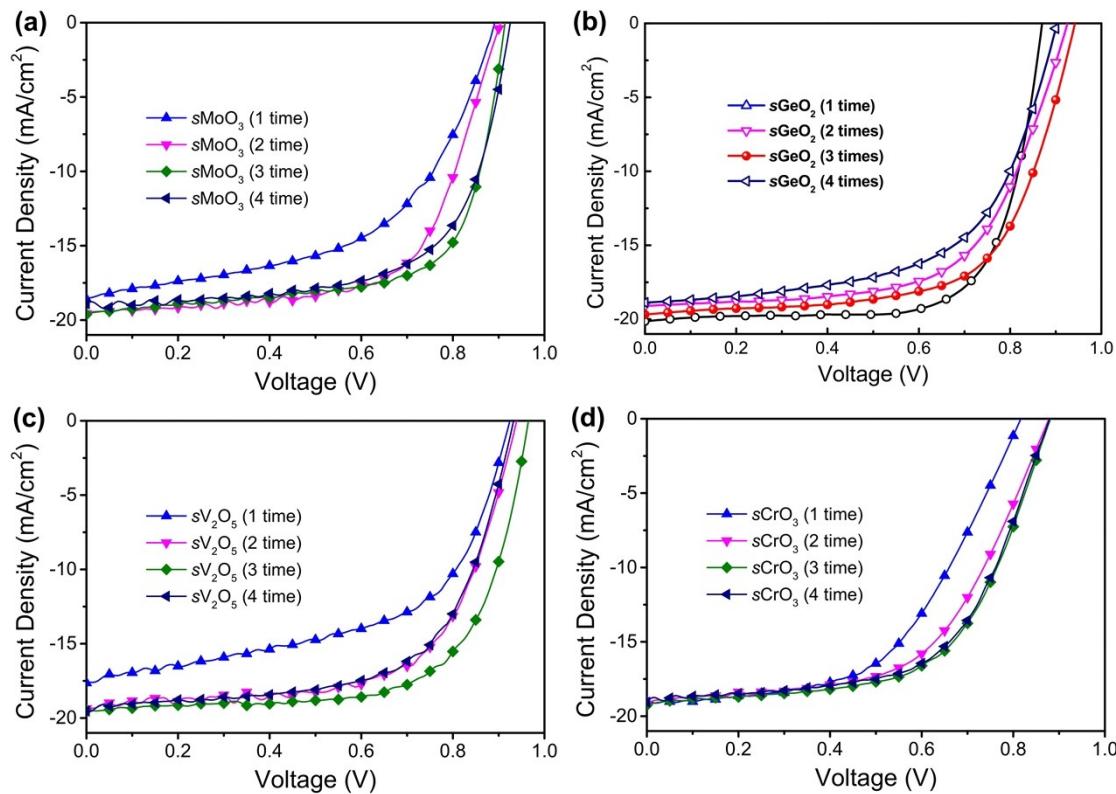


**Figure S3.** Reactance-frequency relations in PTB7:PC<sub>71</sub>BM based hole-dominant devices incorporating with different aqueous solution processed hole transporting layers (a)  $s\text{MoO}_3$ , (b)  $s\text{GeO}_2$ , (c)  $s\text{V}_2\text{O}_5$  and (d)  $s\text{CrO}_3$ .

# Device Optimization of *s*TMOs Based Perovskite Solar Cells

Device structure:

ITO/ TMOs (*s*MoO<sub>3</sub>, *s*GeO<sub>2</sub>, *s*V<sub>2</sub>O<sub>5</sub>, or *s*CrO<sub>3</sub>)/CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3-x</sub>Cl<sub>x</sub>/PC<sub>61</sub>BM/Ag



**Figure S4.** *J-V* curves of perovskite solar cells using different *s*TMO as the HTLs under AM 1.5G illumination of 100 mW cm<sup>2</sup>.

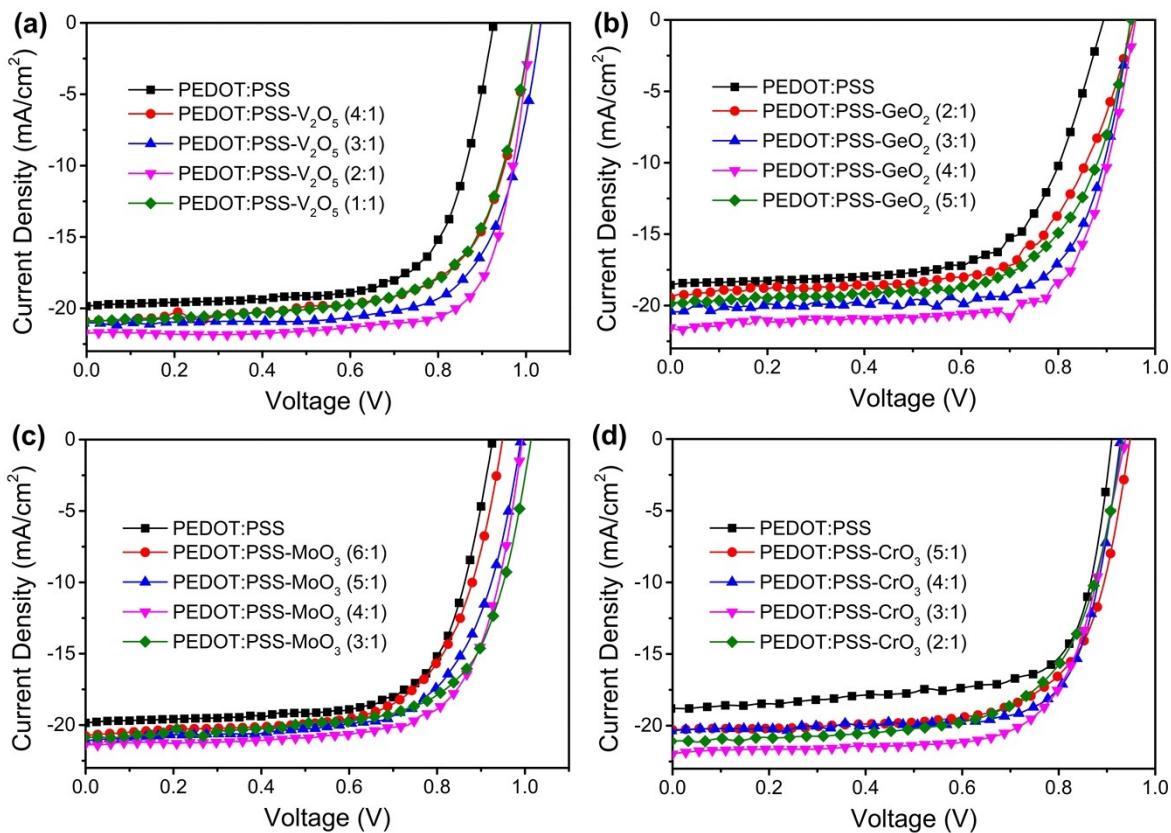
**Table S1.** Cell parameters of perovskite solar cells using different *s*TMO as HTLs

HTL	$J_{SC}$ (mA/cm <sup>2</sup> )	$V_{oc}$ (V)	FF	PCE (%)
<i>s</i> MoO <sub>3</sub> (1 time)	18.58	0.89	0.53	8.79
<i>s</i> MoO <sub>3</sub> (2 time)	19.56	0.90	0.64	11.36
<i>s</i> MoO <sub>3</sub> (3 time)	19.61	0.91	0.68	12.23
<i>s</i> MoO <sub>3</sub> (4 time)	18.90	0.92	0.66	11.47
<i>s</i> GeO <sub>2</sub> (1 time)	18.63	0.88	0.55	8.95
<i>s</i> GeO <sub>2</sub> (2 time)	19.27	0.92	0.62	11.02
<i>s</i> GeO <sub>2</sub> (3 time)	19.62	0.94	0.66	12.12
<i>s</i> GeO <sub>2</sub> (4 time)	18.95	0.90	0.60	10.37
<i>s</i> V <sub>2</sub> O <sub>5</sub> (1 time)	17.63	0.92	0.56	9.10
<i>s</i> V <sub>2</sub> O <sub>5</sub> (2 time)	19.36	0.94	0.65	11.71
<i>s</i> V <sub>2</sub> O <sub>5</sub> (3 time)	19.46	0.96	0.68	12.75
<i>s</i> V <sub>2</sub> O <sub>5</sub> (4 time)	19.53	0.93	0.63	11.46
<i>s</i> CrO <sub>3</sub> (1 time)	19.04	0.81	0.54	8.33
<i>s</i> CrO <sub>3</sub> (2 time)	19.17	0.87	0.57	9.48
<i>s</i> CrO <sub>3</sub> (3 time)	19.26	0.88	0.60	10.15
<i>s</i> CrO <sub>3</sub> (4 time)	19.06	0.88	0.60	9.98

## Device Optimization of PEDOT:PSS-TMOs Based Perovskite Solar Cells

Device structure:

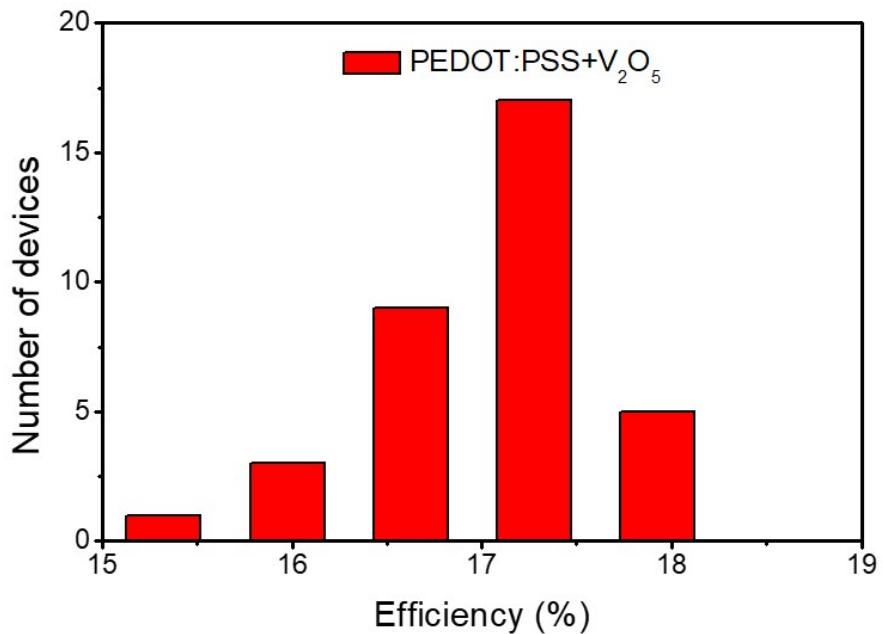
ITO/ PEDOT:PSS-TMOs ( $s\text{MoO}_3$ ,  $s\text{GeO}_2$ ,  $s\text{V}_2\text{O}_5$ , or  $s\text{CrO}_3$ )/ $\text{CH}_3\text{NH}_3\text{PbI}_{3-x}\text{Cl}_x/\text{PC}_{61}\text{BM}/\text{Ag}$



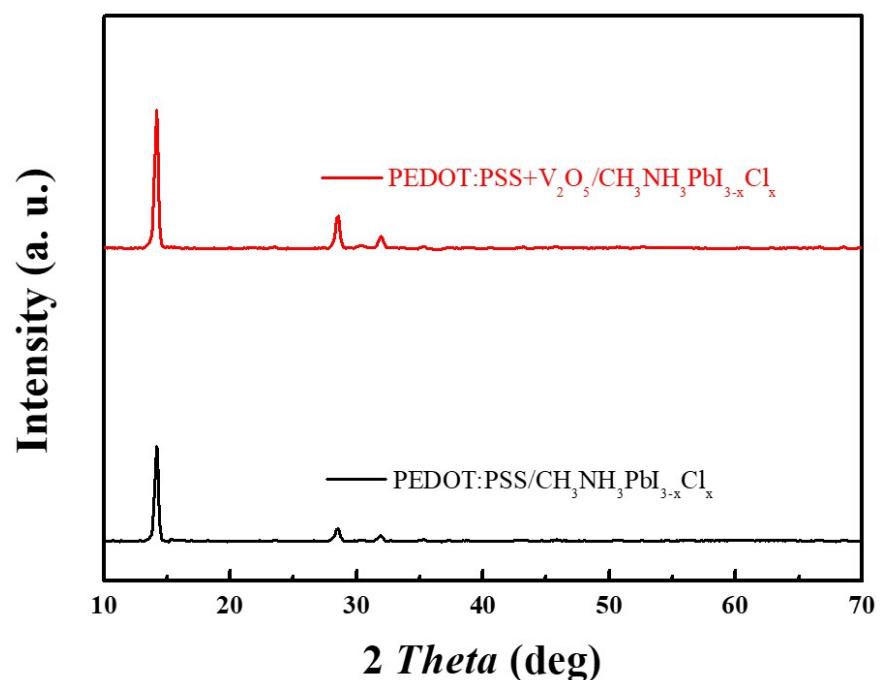
**Figure S5.**  $J$ - $V$  curves of perovskite solar cells using different PEDOT:PSS-TMO as the HTLs under AM 1.5G illumination of 100 mW cm<sup>-2</sup>.

**Table S2.** Cell parameters of perovskite solar cells using PEDOT:PSS-TMOs as HTLs

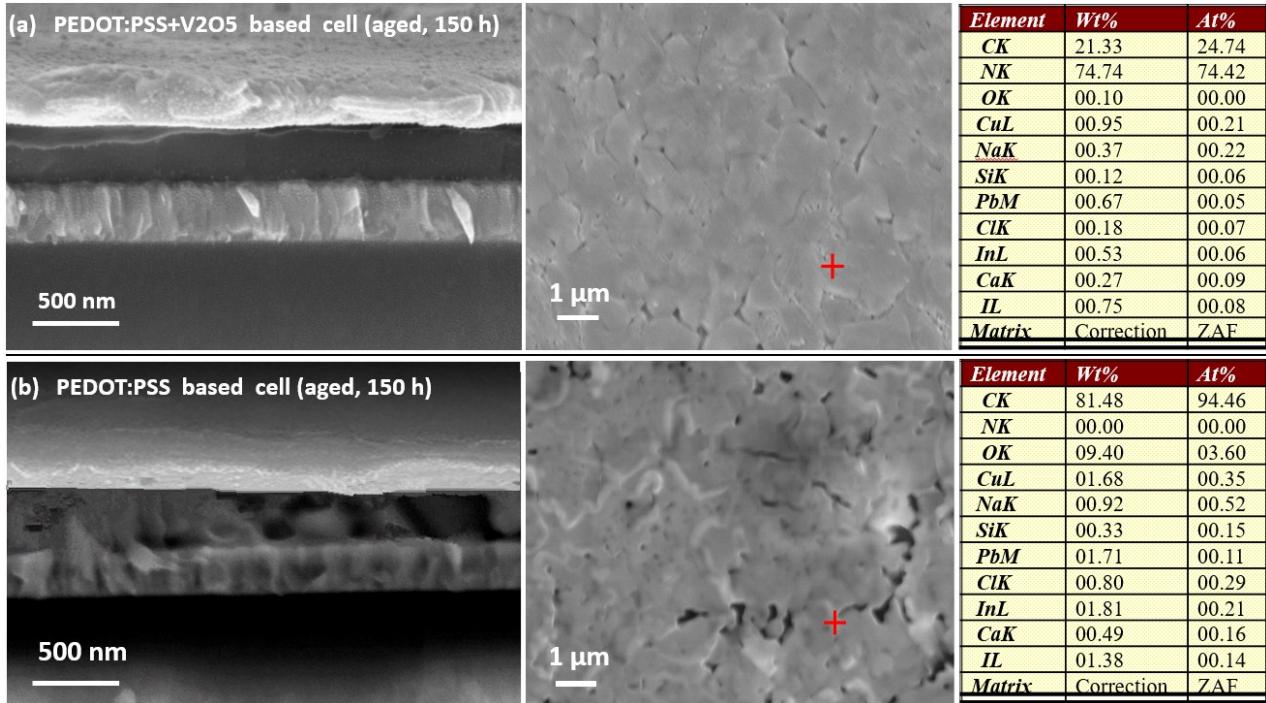
HTL	$J_{SC}$ (mA/cm <sup>2</sup> )	$V_{oc}$ (V)	FF	PCE (%)
PEDOT:PSS	20.64	0.86	0.68	12.08
PEDOT:PSS-MoO <sub>3</sub> (6:1)	21.25	0.94	0.67	13.05
PEDOT:PSS-MoO <sub>3</sub> (5:1)	21.11	0.99	0.67	14.03
PEDOT:PSS-MoO <sub>3</sub> (4:1)	21.38	0.99	0.72	15.10
PEDOT:PSS-MoO <sub>3</sub> (3:1)	20.86	1.01	0.68	14.32
PEDOT:PSS	20.54	0.93	0.74	14.13
PEDOT:PSS-V <sub>2</sub> O <sub>5</sub> (4:1)	20.86	1.01	0.68	14.32
PEDOT:PSS-V <sub>2</sub> O <sub>5</sub> (3:1)	21.02	1.02	0.72	15.56
PEDOT:PSS-V <sub>2</sub> O <sub>5</sub> (2:1)	22.38	1.02	0.79	18.03
PEDOT:PSS-V <sub>2</sub> O <sub>5</sub> (1:1)	20.90	1.01	0.67	14.48
PEDOT:PSS	18.57	0.89	0.67	10.97
PEDOT:PSS-GeO <sub>2</sub> (5:1)	19.84	0.94	0.67	12.50
PEDOT:PSS-GeO <sub>2</sub> (4:1)	21.55	0.96	0.74	15.15
PEDOT:PSS-GeO <sub>2</sub> (3:1)	20.39	0.95	0.72	13.86
PEDOT:PSS-GeO <sub>2</sub> (2:1)	19.44	0.96	0.65	12.10
PEDOT:PSS	18.79	0.91	72	12.37
PEDOT:PSS-CrO <sub>3</sub> (5:1)	20.29	0.94	70	13.33
PEDOT:PSS-CrO <sub>3</sub> (4:1)	20.32	0.93	75	14.09
PEDOT:PSS-CrO <sub>3</sub> (3:1)	21.97	0.94	71	14.50
PEDOT:PSS-CrO <sub>3</sub> (2:1)	21.07	0.93	67	13.13



**Figure S6.** A histogram of PCEs measured from 35 PEDOT:PSS- $V_2O_5$  HTL based perovskite solar cells.



**Figure S7.** XRD patters of polycrystalline  $\text{CH}_3\text{NH}_3\text{PbI}_{3-x}\text{Cl}_x$  films deposited on PEDOT:PSS and PEDOT:PSS+V<sub>2</sub>O<sub>5</sub> underlayers.



**Figure S8.** Cross-sectional SEM and EDX analysis of (a) PEDOT:PSS+V<sub>2</sub>O<sub>5</sub> based and (b) PEDOT:PSS based perovskite solar cells (aged, 150h).