Top-contacts-interface Engineering for High-performance Perovskite Solar

Cell with Reducing Lead Leakage

Huan Bi^{1,2}, Gaoyi Han¹, Mengna Guo¹, Chao Ding², Shuzi Hayase², Hanjun Zou⁴, Qing Shen²*, Yao Guo³*, and Wenjing Hou¹*

¹Institute of Molecular Science, Key Laboratory of Materials for Energy Conversion and Storage of Shanxi Province, Shanxi University, Taiyuan 030006, P. R. China

²Faculty of Informatics and Engineering, The University of Electro-Communications, 1-5-1 Chofugaoka, Chofu, Tokyo 182-8585, Japan.

³School of Materials Science and Engineering, Henan Joint International Research Laboratory

of Nanocomposite Sensing Materials, Anyang Institute of Technology, Anyang 455000, P. R. China

⁴Analytical and Testing Center, Chongqing University, Chongqing 401331, P. R. China

Corresponding Authors (Q. Shen, Y. Guo, W.J. Hou)

E-mail: shen@pc.uec.ac.jp

E-mail: guoyao@ayit.edu.cn

E-mail: houwenjing@sxu.edu.cn



Figure S1. Chemical structures of the emtricitabine (FTC) used in this work. Here, in order to make the following calculation clearer, O on carbonyl is named O1. Elements marked with black dashed lines are used for calculation



Figure S2. A) Pb, B) F, C) I, and D) full spectra EDS mapping of the perovskite film with FTC modification.



Figure S3. FTIR spectra of FTC, perovskite, and perovskite treated by FTC.



Figure S4. XPS spectra of FTC, perovskite, and FTC-modified perovskite film.



Figure S5. FTIR spectra of PVSK and FTC-treated PVSK. In which, the peak appeared in 3262 cm⁻¹ is assigned to stretching vibration of N-H.



Figure S6. The 3D visual charge density difference of a) control and b) target film (O2-site).



Figure S7. The 3D visual charge density difference of a) control and b) target film (N-site).



Figure S8. The density of state of the FTC with the site of O2.



Figure S9. The density of state of the control film.



Figure S10. The density of state of the FTC modified perovskite film with the site of O2.



Figure S11. The DOS of I 5*s*, 5*p*, and 5*d* of the target and control film.



Figure S12. Bandgaps of the perovskite films without and with modification calculated from UV-Vis spectra.



Figure S13. Dark *I*–*V* curves of the device ITO/PEDOT:PSS/PVSK/Spiro-OMeTAD/Au based on (g) pristine and (h) FTC-modified perovskite films.



Figure S14. V_{OC} depended on light intensity curves for the control and target devices.



Figure S15. The cut-off energy (E_{cut} -off) and Fermi edge (E_F , edge) of the control device.



Figure S16. The cut-off energy (E_{cut} -off) and Fermi edge (E_F , edge) of the target device.



Figure S17. Statistics of A) V_{OC} , B) J_{SC} , C) FF, and D) PCE of PSCs based on perovskite modified by different mass concentrations of FTC.



Figure S18. A) Normalized J_{SC} , B) Normalized V_{OC} , and C) Normalized FF as a function of time for the unencapsulated devices based on perovskite film without and with FTC modification exposed to the humidity of 5% RH at room temperature.



Figure S19. A) Normalized J_{SC} , B) Normalized V_{OC} , and C) Normalized FF as a function of time for the unencapsulated devices based on perovskite film without and with FTC modification aged under the humidity of 40%~45% RH at room temperature.



Figure S20. Chemical structures of the lamivudine (LMD) used in this work.



Figure S21. Normalized PCE as a function of time for the unencapsulated devices based on LMD-, FTC-modified perovskite and bare perovskite aged under the humidity of 65%~75% RH at room temperature.



Figure S22. The Pb concentration was measured by ICP-MS with the structure of ITO/PbI₂ and ITO/PbI₂/FTC.

	Glass/PVSK	Glass/PVSK/FTC
τ_1 (ns)	13.63	15.95
%	93	88
τ_2 (ns)	34.32	63.74
%	7	12
$\tau_{\rm ave}$ (ns)	16.93	32.82

Table S1 Fitting results from TRPL kinetics in Figure 4B.

	Glass/PVSK/HTL	Glass/PVSK/FTC/HTL
τ_1 (ns)	4.05	2.24
%	92	97
τ_2 (ns)	12.19	5.68
%	8	3
$\tau_{\rm ave} ({\rm ns})$	5.74	2.49

Table S2 Fitting results from TRPL dynamics in Figure 5B.

FTC (mg/mL)		$J_{\rm SC}$ (mA/cm ²)	V _{OC} (V)	FF	PCE (%)
0	Champion	24.52	1.092	0.778	20.83
	Average	24.49±0.15	1.091 ± 0.004	0.775 ± 0.008	$\begin{array}{c} 20.71 \pm \\ 0.12 \end{array}$
	Champion	24.66	1.098	0.791	21.42
0.25	Average	24.59±0.13	1.096 ± 0.002	0.793 ± 0.006	$\begin{array}{c} 21.37 \pm \\ 0.17 \end{array}$
0.5	Champion	24.99	1.107	0.804	22.24
	Average	24.84 ± 0.14	1.104 ± 0.004	0.797 ± 0.008	$\begin{array}{c} 21.81 \pm \\ 0.41 \end{array}$
0.75	Champion	24.62	1.099	78.93	21.36
	Average	24.71±0.11	1.096±0.003	0.786 ± 0.008	21.29± 0.11
1	Champion	24.69	1.092	0.784	21.13
	Average	24.58±0.14	1.091 ± 0.003	0.784 ± 0.007	$\begin{array}{c} 21.02 \pm \\ 0.10 \end{array}$

Table S3 Champion, average and standard deviation photovoltaic parameters of the PSCs modified different mass concentrations of FTC.