Supporting Information

Strengthened Perovskite/Fullerene Interface Enhances the Inverted Planar Perovskite Solar Cells Regarding Efficiency and Stability via Tetrafluoroterephthalic Acid Interlayer

Minhua Zou[1], Xuefeng Xia[1], Yihua Jiang[1], Jiayi Peng[1], Zhenrong Jia[2], Xiaofeng Wang[1], Fan Li*[1]

[1]Department of Materials Science and Engineering, Nanchang University, 999 Xuefu Avenue, Nanchang 330031, China

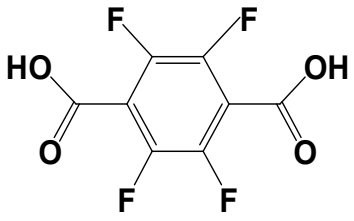
[2] National Laboratory for Molecular Sciences, CAS Key Laboratory of Organic Solids, Institute of Chemistry, Chinese Academy of Sciences, Beijing 100190, China

*Corresponding author. E-mail address: lfan@ncu.edu.cn (F. Li)

Characterization

The X-ray diffraction (XRD) spectrum of the $MAPbI_3$ and $MAPbI_3$/TFTPA films were characterized by an X-ray diffractometer (Bruker D8 DISCOVER) at a scanning rate of 1° min^{-1} in the Bragg-Brentano mode, using Cu K_α radiation (1.540598 Å). Spectra were acquired with a linear silicon strip "LynxEye" detector from 2θ = 4°-45° at a step width of 0.02° in a source slit width of 1 mm. The surface morphological observations of $MAPbI_3$, $MAPbI_3$/TFTPA and $MAPbI_3$/TFTPA/$PC_{61}BM$ films were made by field-emission scanning electron microscopy (SEM, QuanTA-200F) with gold vapor deposition and atomic force microscopy (AFM, Digital Instrument Nanoscope 31). Ultraviolet-visible (UV-vis) absorption were measured by a UV-vis spectrophotometer (PerkinElmer LAMBDA 750). Fourier Transform infrared spectroscopy (FT-IR) was recorded using a Nicolet iS50. Contact-angle measurements were conducted on Kruss DSA-100, the photographs were taken 1 s after water dripping. Elemental analysis was characterized by X-ray photoelectron spectroscopy (XPS, Thermo Fisher Scientific). Ultraviolet photoelectron spectroscopy (UPS) was measured with XPS measurements using an AXIS-ULTRA DLD

spectrometer (Kratos Analytical Ltd.). For UPS measurement, the photon energy of the He I lamp was 21.21 eV and the spectra were collected in a pass energy of 5 eV. Time-resolved Fluorescence for photoluminescence (TRPL) decays were measured with a Horiba FluoreCube spectrofluorometer system by using a UV diode laser (NanoLED λ=465nm) for excitation. Steady Fluorescence measurements for photoluminescence (PL) of the samples were carried on a Horiba FluoroMax-4 luminescence spectrofluorophotometer. The excitation wavelength was 465 nm and emission wavelength was 767 nm for $MAPbI_3$ films. The *J-V* curves measured by Keithley model 2400 source measuring unit with 0.01 V s^{-1} scan rate, the decay time is 10 ms and without pretreatments under the simulated AM 1.5 G one illumination of 100 mW cm^{-2}. A Newport Air Mass 1.5 Global (AM1.5G) full-spectrum solar simulator was applied as the light source. The EQE spectrum was obtained using a solar cell spectral response measurement system (Enlitech, Taiwan). Electron-only devices (FTO/TiO_2/$MAPbI_3$/$PC_{61}BM$/Ag or FTO/TiO_2/$MAPbI_3$/TFTPA/$PC_{61}BM$/Ag) were used to measure the dark J-V by an Agilent (B2901A) precision source for the SCLC analysis. The mobility was extracted by fitting the J-V curves by the Mott-Gurney equation. The trap state density was determined by the trap-filled limit voltage using equation. Capacitance-voltage (C-V) measurements were characterized using an AutoLab potentiostat (model PGSTAT30) equipped with a frequency analyzer module at normal atmosphere (25 °C), in which a small voltage perturbation (20 mV rms) was applied at 10 kHz at a scan rate of 0.02 V s^{-1}. The electrochemical impedance spectroscopy (EIS) measured by Electrochemical workstation (VersaSTAT3, AMETEK) with the frequency range from 1 Hz to 1 MHz in the dark at room temperature. The applied voltage is close to the V_{oc} of each device and the AC amplitude is 10 mV.



Scheme S1. Chemical structure of TFTPA molecule.

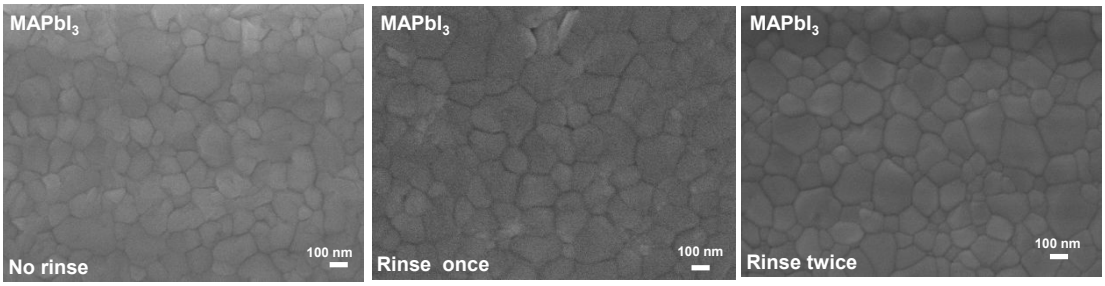


Figure S1. Top-view SEM images of the $MAPbI_3$ films rinsed with ethyl acetate at different times.

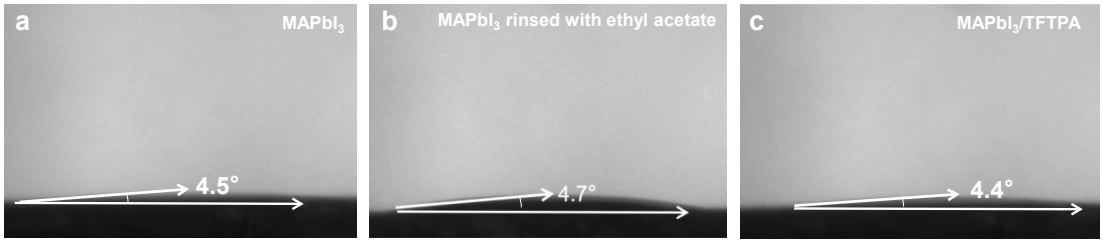


Figure S2. Contact angles with $PC_{61}BM$ drops on (a) the $MAPbI_3$ film, (b) the $MAPbI_3$ film rinsed with ethyl acetate and (c) the $MAPbI_3$/TFTPA film.

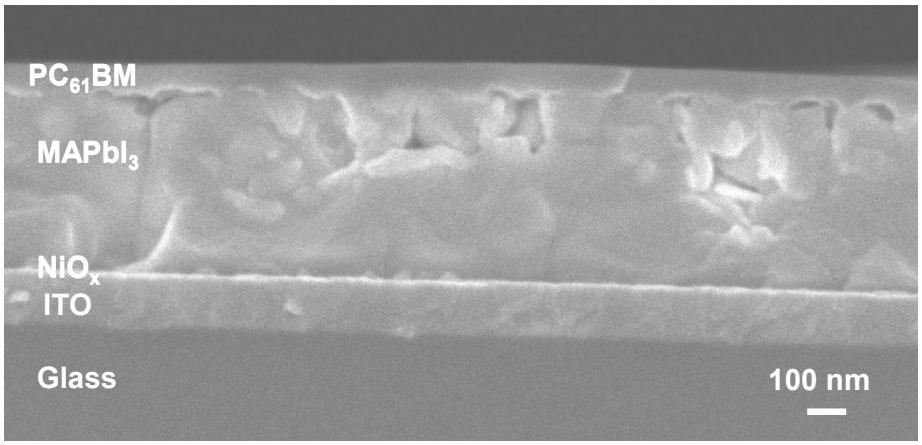


Figure S3. Cross-sectional SEM image of Glass/ITO/NiO_x/$MAPbI_3$/$PC_{61}BM$.

Table S1. Performance of PSCs fabricated with TFTPA and without TFTPA under standard AM 1.5 illumination (100 mW cm^{-2}) at active layer area (0.10 cm^2).[a]

	V_{oc} (V)	J_{sc} (mA cm^{-2})	FF	PCE (%)
Control device	1.05 ± 0.04	18.1 ± 1.2	0.727 ± 0.031	13.8 ± 2.3
TFTPA device	1.12 ± 0.01	20.4 ± 0.7	0.786 ± 0.021	18.0 ± 1.4

[a]The statistics is determined from 30 devices.

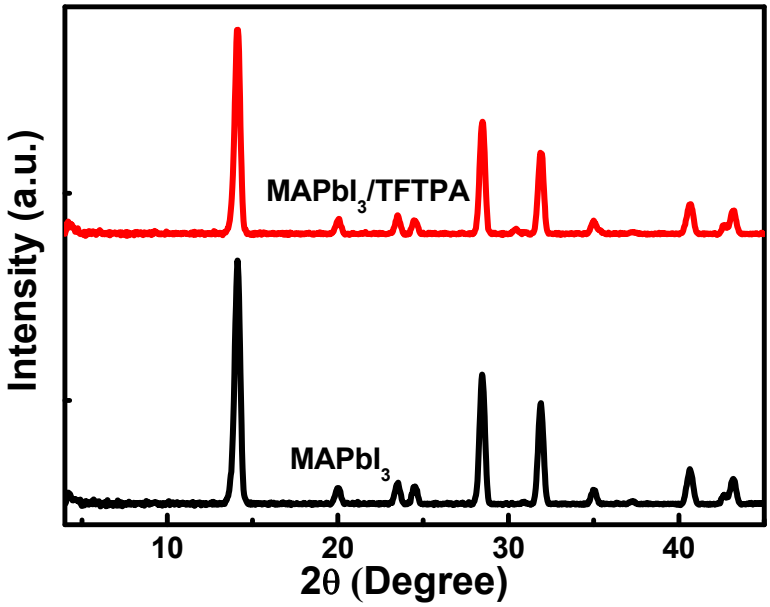


Figure S4. XRD patterns of the $MAPbI_3$ film and $MAPbI_3$/TFTPA film.

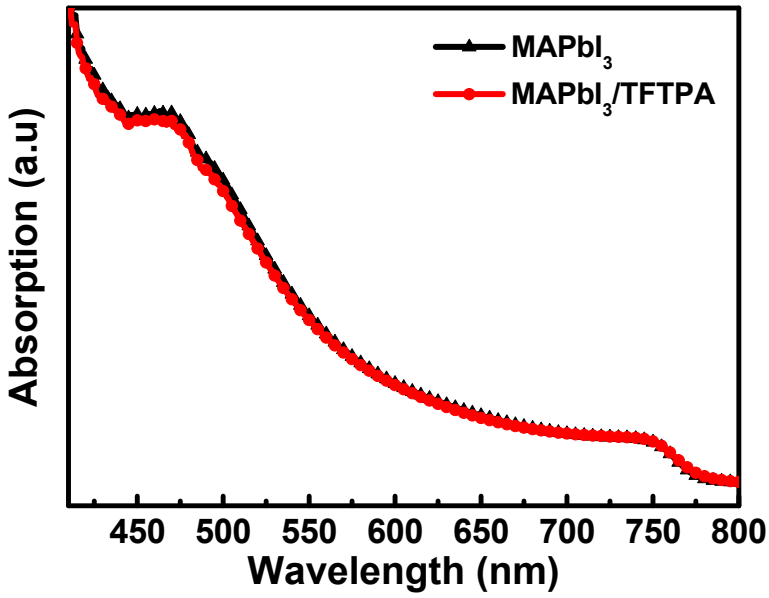


Figure S5. UV–vis absorption spectra of the $MAPbI_3$ film and $MAPbI_3$/TFTPA film.

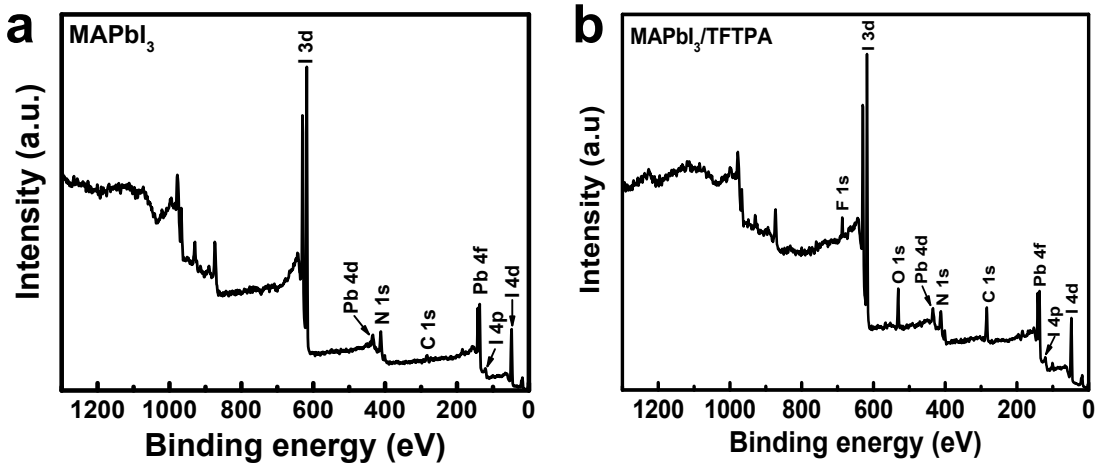


Figure S6. XPS spectra of (a) the $MAPbI_3$ film and (b) $MAPbI_3$/TFTPA film.

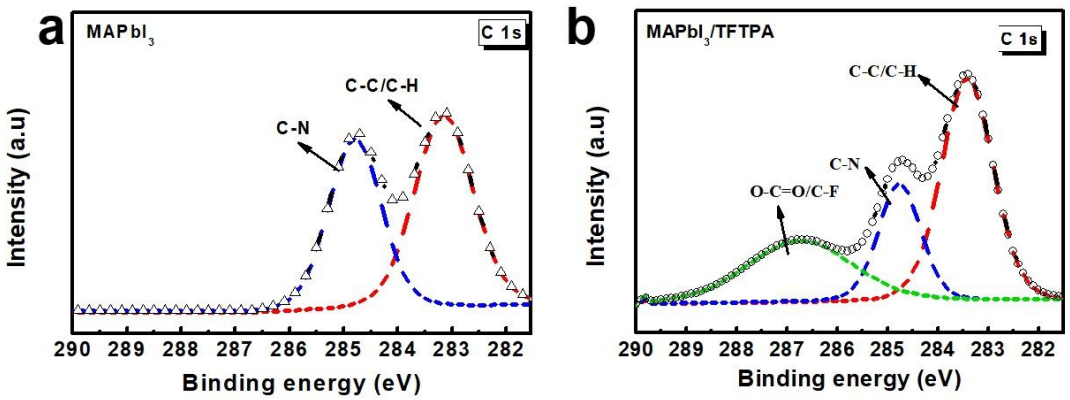


Figure S7. High-resolution C 1s XPS spectra of (a) the $MAPbI_3$ film and (b) $MAPbI_3$/TFTPA film.

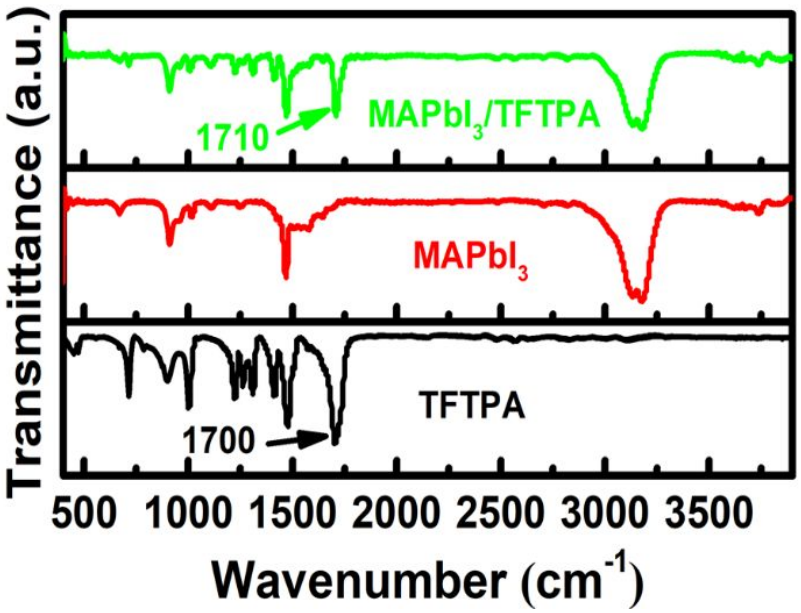


Figure S8. FTIR spectra of TFTPA, $MAPbI_3$ film and $MAPbI_3$/TFTPA film.

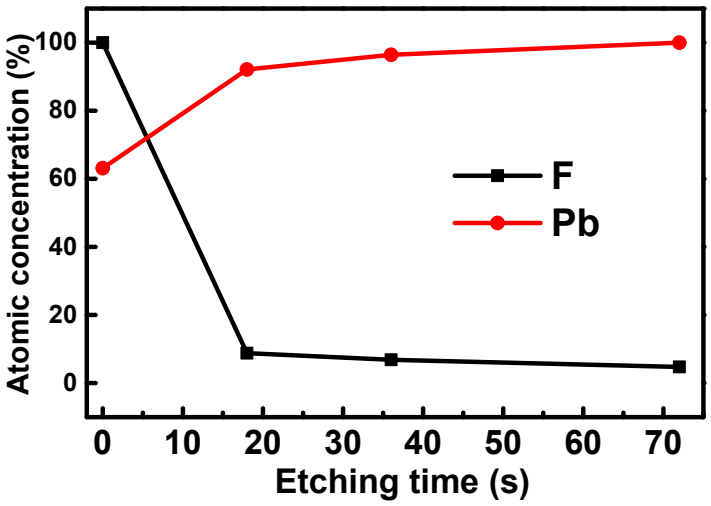


Figure S9. Vertical atomic concentration distribution of F and Pb in the $MAPbI_3$/TFTPA film.

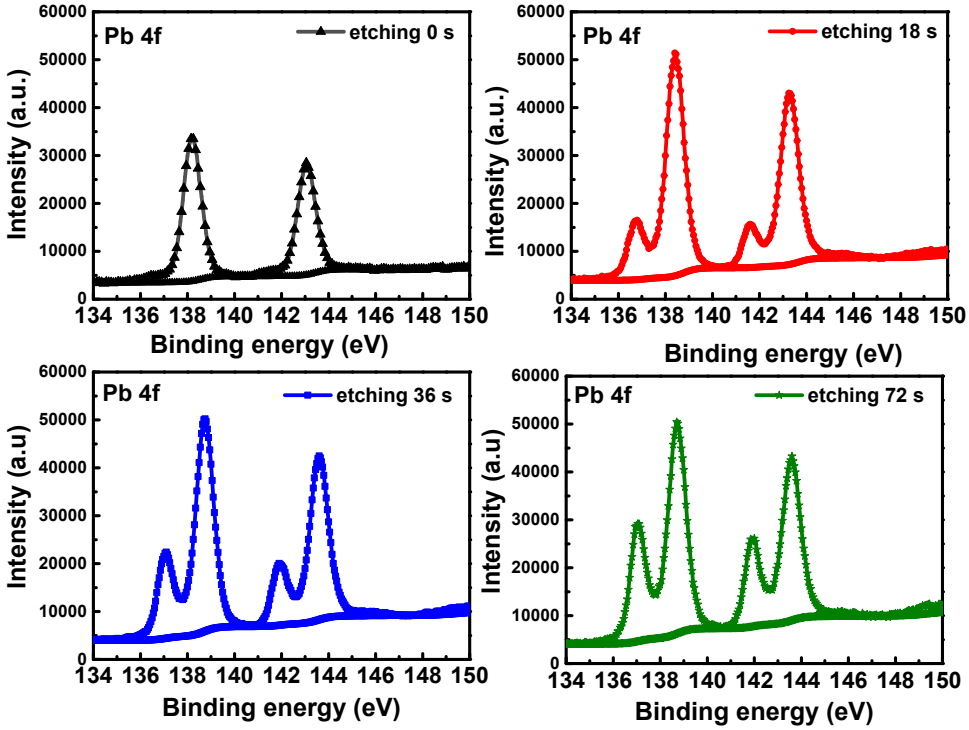


Figure S10. High-resolution Pb 4f XPS spectra of the $MAPbI_3$/TFTPA film after etching different time.

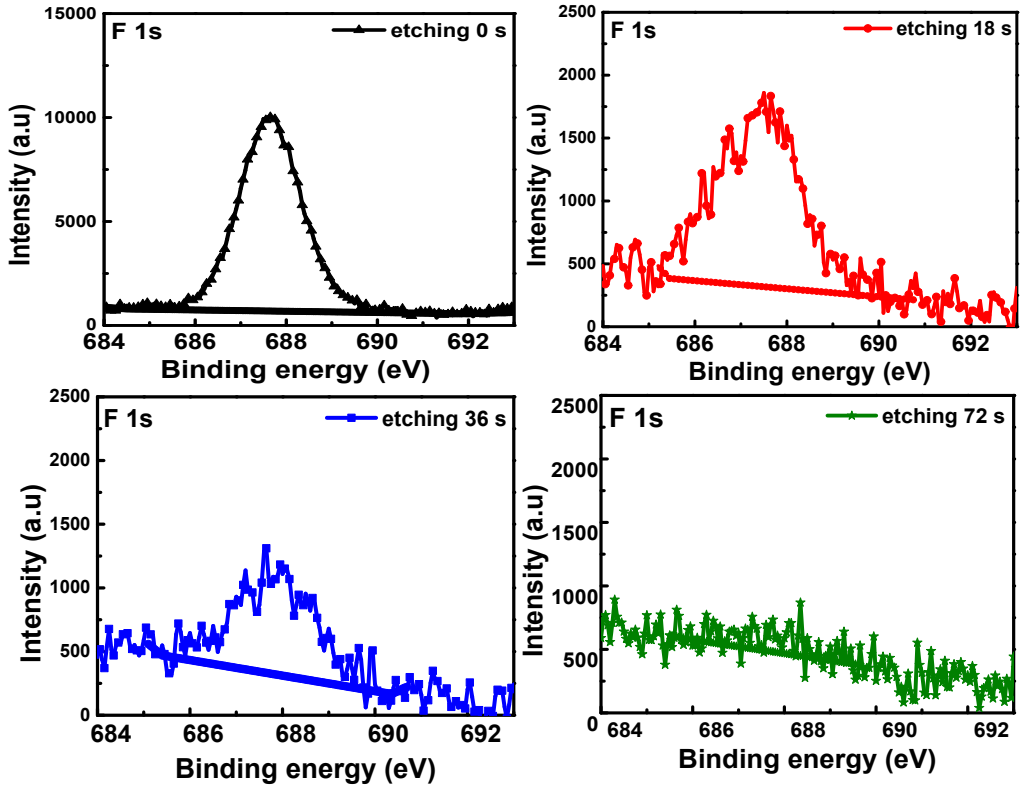


Figure S11. High-resolution F 1s XPS spectra of the $MAPbI_3$/TFTPA film after etching different time.

Table S2. Time-resolved photoluminescence (TRPL) decay fitting parameters of the $MAPbI_3$, $MAPbI_3$/TFTPA, $MAPbI_3/PC_{61}BM$ and $MAPbI_3$/TFTPA/$PC_{61}BM$ films.

Sample	τ_1 (ns)	τ_2 (ns)	A_1 (%)	A_2 (%)	τ_{ave} (ns)
$MAPbI_3$	0.84	3.36	8.77	91.23	3.14
$MAPbI_3$/TFTPA	1.58	6.31	7.74	92.26	5.83
$MAPbI_3/PC_{61}BM$	2.07	2.62	43.28	56.72	2.38
$MAPbI_3$/TFTPA/$PC_{61}BM$	0.62	2.47	49.71	50.29	1.55

Table S3. Energy levels of the pristine $MAPbI_3$ and $MAPbI_3$/TFTPA films.

Substrate	E_{cutoff} (eV)	E_{onset} (eV)	HOMO (eV)[a]	LUMO (eV)[b]
$MAPbI_3$	16.58	0.78	5.42	3.83
$MAPbI_3$/TFTPA	16.50	0.76	5.48	3.89

[a]The HOMO level is acquired from the UPS spectrum.

[b]The LUMO level is acquired by adding the bandgap obtained from the UV-vis absorption spectrum to the HOMO level.

Table S4. EIS parameters for the PSCs fabricated with TFTPA and without TFTPA.

Devices	R_s (Ω)	R_{tr} (Ω))	R_{rec} (Ω)	CPE_{tr} -T (F)	CPE_{rec} -T (F)
w/o TFTPA	1.35	38.42	824.56	1.31×10^{-9}	6.08×10^{-9}
With TFTPA	1.07	27.56	1180.38	1.07×10^{-9}	1.79×10^{-8}