A Novel Alkylsilyl-fused Copolymer-based Non-fullerene Solar Cells with over 12% Efficiency

Guodong Xu [a,b], Lie Chen*[a,b] Hui Lei [a,b], Zhihui Liao [a,b], Nan Yi [a,b], Jinliang Liu [a,b], Yiwang Chen*[a,b]

[a]College of Chemistry, Nanchang University, 999 Xuefu Avenue, Nanchang 330031, China

[b]Jiangxi Provincial Key Laboratory of New Energy Chemistry/Institute of Polymers, Nanchang University, 999 Xuefu Avenue, Nanchang 330031, China

Corresponding author. Tel.: +86 791 83968703; fax: +86 791 83969561. E-mail: ywchen@ncu.edu.cn (Y. Chen), chenlie@ncu.edu.cn (L. Chen).

Supporting information

Materials: tetrakis(triphenylphosphine)palladium(0)($Pd(PPh_3)_4$),and other materials were purchase from Alfa or Aldrich, Some solvents were distilled before use (tetrahydrofuran (THF) from sodium), IT-4F were purchased form Derthon. molybdenum oxide (MoO_3) and Ag (99.998%) were purchased from Nano-C and Rieke Metals, Inc. Indium tin oxide (ITO) was purchased from Delta Technologies Limited.

Device Fabrication

(1)The inverted device structure was ITO/ZnO/PBDS-TZ:IT-4F/MoO_3/Ag. A glass substrate with a pre-patterned ITO (sheet resistance=15 Ω sq^{-1}) was ultrasonicated in acetone, detergent, deionized water and isopropanol in turn, and then modified by air plasma treatment for 1 min. After spin-coating ZnO with 4000rmp, 1min. and baked on a hot plate at 205 ^oC for about 60 min. PTZ1-Si:IT-4F was dissolved in chlorobenzene with 0.25% (vol %)1,8-diiodoctane(DIO),(total concentration is 16 mg/mL) with 1000 rpm for 1 min for active materials. And then with various temperature TA treatment. Then the molybdenum trioxide (7 nm) and argentum cathode (90 nm) was successively thermal evaporation over interface layer under a high vacuum chamber (7×10^{-4} Pa) to accomplish the device fabrication.

(2) The conventional device structure was ITO/PEDOT:PSS/PBDS-TZ:IT-4F /PNDIO/Al. A glass substrate with a pre-patterned ITO (sheet resistance =15 Ω sq^{-1})

was first ultrasonicated in acetone, detergent, deionized water and isopropanol in turn, and then modified by air plasma treatment for 3 min. After filtration through a 0.45 μm filter, PEDOT:PSS (Bay PVPAI4083, Bayer AG) was spin-coated at 5000 rpm for 60 s to form a thickness of 30 nm thin layer on the cleaned ITO substrate, and baked on a hot plate at 140 ^{o}C for about 20 min. The PBDS-TZ:IT-4F (1:1 w/w) was dissolved in chlorobenzene with 0.25% (vol %)1,8-diiodoctane (DIO), mixed in solution with total concentration of 16 mg mL^{-1}, spin-coated onto the interfacial layer at 1000 rpm for 1 min. The PNDIO solution (1 mg/mL) was spin-coated on the active layer at 3000 rmp for 60 s, then the aluminium (100 nm) cathode was thermal evaporation over interface layer under a high vacuum chamber (7×10^{-4} Pa) to accomplish the device fabrication.

The effective device area of one cell was 0.04cm^2. The I–V characterization of the devices were measured by a Keithley 2400 Source Meter under simulated solar light (100 mW/cm^2, AM 1.5 G, Abet Solar Simulator Sun2000). Electron-only devices were fabricated similar to the PSCs with a structure of ITO/ZnO/PBDS-TZ:IT-4F / PDINO/Al. And the hole-only devices fabricated similar to the PSCs with a structure of ITO/PEDOT:PSS/ PBDS-TZ:IT-4F /MoO_3/Al. The EQE values measuring system (Oriel Cornerstone 260 1/4 m monochromator equipped with Oriel 70613NS QTH lamp), the monochromatic light from a xenon lamp. A solar simulator was used as the light source, and the light intensity was monitored by using a standard Si solar cell. All characterization processes were conducted in the air.

CV measurements were carried out on a CHI660 potentiostat/galvanostat electrochemical work station at a scan rate of 20 mVs^{-1}, with a platinum wire as counter electrode and a silver/silver chloride (Ag/AgCl) as reference electrode in a 0.1 mol L^{-1} solution of tetrabutylammonium perchlorate (n-Bu_4NClO_4) in anhydrous and nitrogen-saturated acetonitrile (CH_3CN). A platinum plate coated on a thin film of the studied the two small molecule electrolyte was used as the work electrode. The HOMO and LUMO energy level were calculated by the empirical formula:

E_{HOMO}= [E_{ox}+4.8-$E_{Fc/Fc+}$] (eV J^{-1}), E_{LUMO}- E_{HOMO} =E_g.

where $E_{Fc/Fc+}$ is the redox potential of ferrocene/ferrocenium ($E_{Fc/Fc+}$) couple in the electrochemical measurement system, and the evergy level of $E_{Fc/Fc+}$ was taken as 4.8eV below vacuum. The E_{ox} is the analyte oxidation onset.

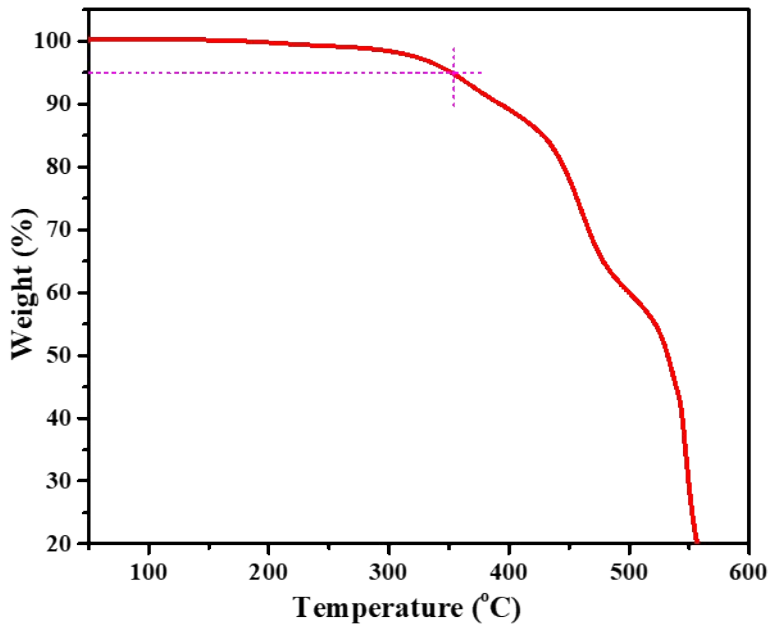

Figure S1. Thermogravimetric analysis (TGA) spectra of PBDS-TZ under nitrogen at a heating rate of 10 °C/min.

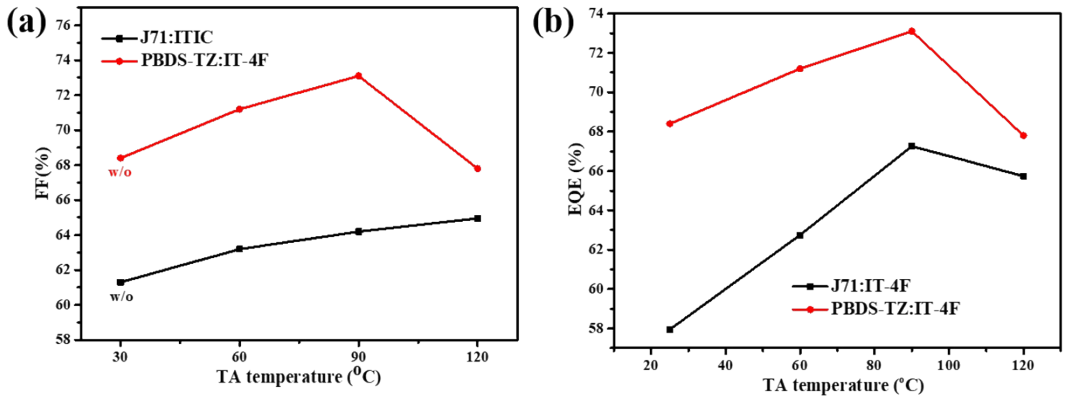

Figure S2. (a) The fill factor (FF) of inverted OSCs device based on J71:ITIC and PBDS-TZ:IT-4F with various TA temperature. (b) The fill factor (FF) of inverted OSCs device based on J71:IT-4F and PBDS-TZ:IT-4F with various TA temperature.

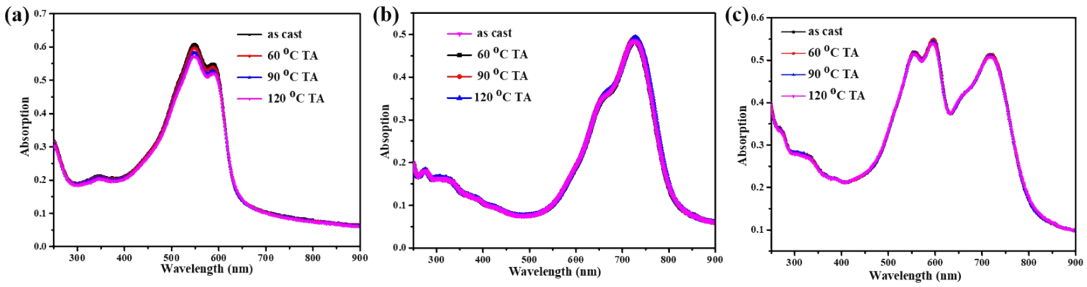

Figure S3. Absorption spectra of films with various temperature thermal annealing treatment: (a) PBDS-TZ film. (b) IT-4F film. (c) PBDS-TZ:IT-4F blend film.

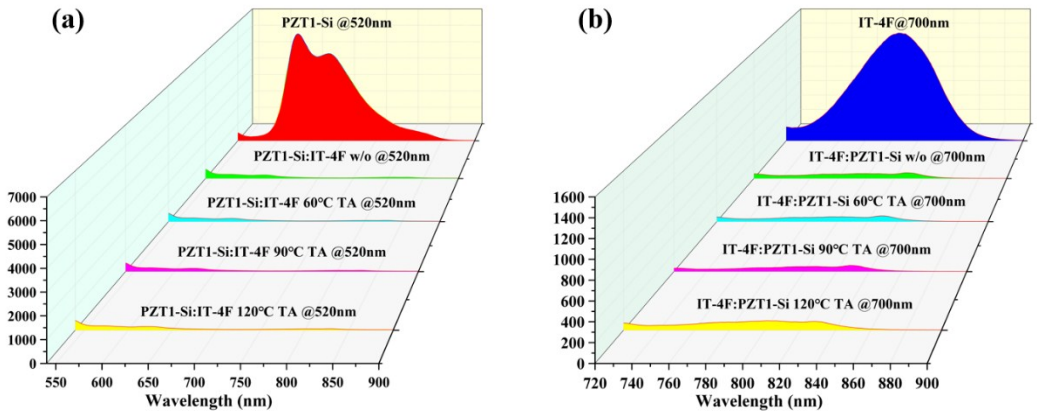

Figure S4. Photoluminescence spectra with various temperature of the a) PBDS-TZ:IT-4F blend film (excited at 520 nm); b) PBDS-TZ:IT-4F blend film (excited at 700 nm).

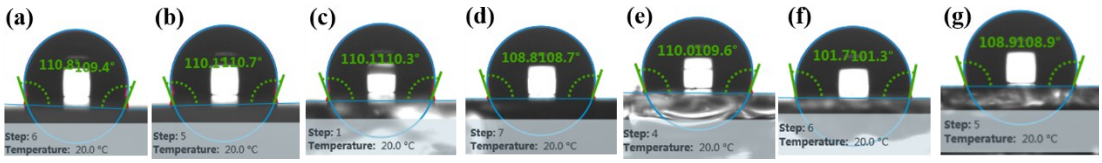

Figure S6. Photographs of water droplets on the surface of various film. a) PBDS-TZ film, b) PBDS-TZ:IT-4F blend film without thermal annealing treatment, c) PBDS-TZ :IT-4F blend film with 60 ^{o}C thermal annealing treatment.(d) PBDS-TZ :IT-4F blend film with 90 ^{o}C thermal annealing treatment. (e) PBDS-TZ:IT-4F blend film with 120 ^{o}C thermal annealing treatment.(f) IT-4F film.(g) PZT1-Si :IT-4F blend film without DIO.

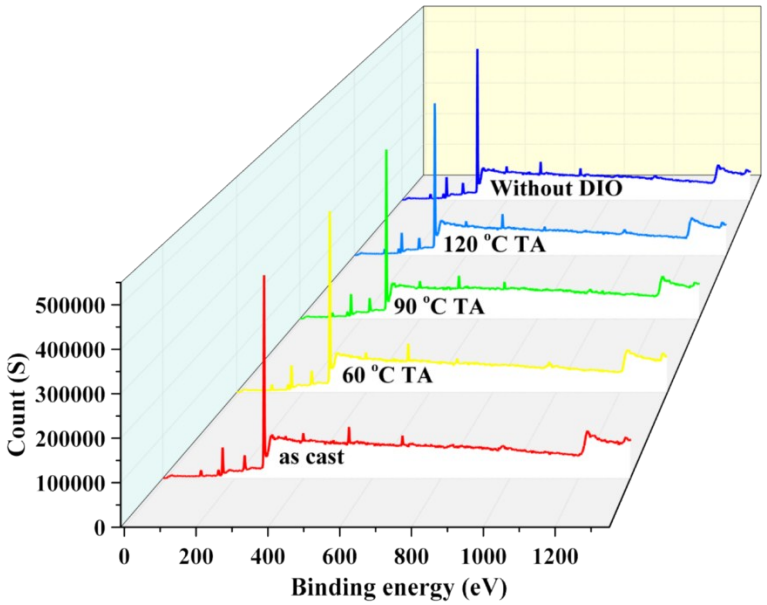

Figure S7. X-ray photoelectron spectroscopy (XPS) spectra of surface on blend films.

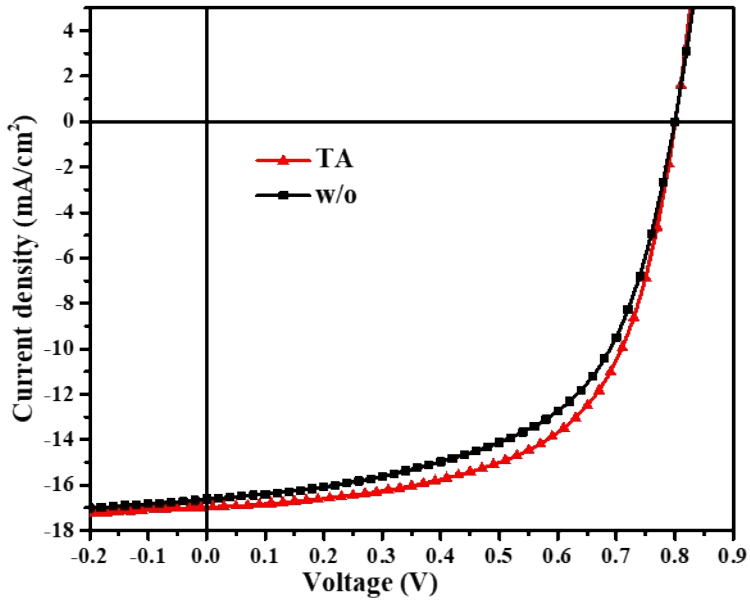

Figure S8. *J-V* curves of the PSCs devices ITO/PEDOT:PSS/PBDS-TZ:IT-4F /PDINO/Al without or with 90 °C TA treatment.

Table S1. *J-V* curves of the OSCs devices ITO/ZnO/PBDS-TZ:IT-4F/MoO_3/Ag.

DIO (%)	TA (^oC)	D/A ratio	V_{oc} (V)	J_{sc} (mA/cm^2)	FF (%)	PCE (%)
—	—	1:1	0.800±0.007	19.30±0.35	33.80±0.37	5.42[b](5.20[a]±0.25)
0.5	—	1:1	0.787±0.002	18.45±0.37	66.70±0.35	9.69[b](9.57[a]±0.23)
0.5	60	1:1	0.781±0.004	19.10±0.42	68.62±0.31	10.24[b](9.97[a]±0.37)
0.5	90	1:1	0.773±0.003	20.17±0.46	69.60±0.35	10.85[b](10.7[a]±0.25)
0.5	120	1:1	0.778±0.004	19.20±0.32	67.30±0.34	10.10[b](9.93[a]±0.21)
0.25	90	1.5:1	0.734±0.003	17.38±0.31	63.48±0.34	8.11[b](7.89[a]±0.27)
0.25	90	1:1.5	0.764±0.003	15.24±0.38	60.91±0.38	7.09[b](6.97[a]±0.25)

[a]The averages for photovoltaic parameters of each device are given in parentheses with mean variation obtained from 10 devices, and the ± refer to the standard deviation.
[b]Best device PCE.

Table S2. Information of hole and electron mobility of PBDS-TZ:IT-4F.

	μ_h ($cm^2V^{-1}s^{-1}$)	μ_e ($cm^2V^{-1}s^{-1}$)	μ_e/μ_h
Without	4.53×10^{-4}	2.40×10^{-4}	1.89
90 ^oC TA	5.01×10^{-4}	2.71×10^{-4}	1.85

Table S3. Information of the top surface measured by Contact angle and X-ray photoelectron spectroscopy.

	θ_{water} [°]	Surface energy γ_s [mJ m^{-2}]	Si [%]	F [%]	S [%]	C [%]	N [%]
PZTI-Si	110.11	17.03	—	—	—	—	—
IT-4F	101.50	19.93	—	—	—	—	—
blend film w/o	110.20	16.97	3.66	1.26	6.84	82.10	2.73
blend film 60 °C TA	109.94	17.70	3.28	1.20	6.64	82.53	2.36
blend film 90 °C TA	108.77	17.80	3.31	1.24	6.67	82.67	3.14
blend film 120 °C TA	109.80	17.21	3.19	0.94	6.26	83.9	2.62
blend film without DIO	108.90	17.75	3.16	1.25	6.80	82.20	2.77

Table S4. The atomic content of blend films with various etch times.

	Etch time (s)	Si (%)	S (%)	C (%)	N (%)	O (%)	F (%)
Without TA	0	3.66	6.84	82.10	2.73	3.40	1.26
	100	2.86	4.95	87.21	1.65	1.54	1.76
	200	2.77	4.82	87.30	1.63	1.68	1.77
	300	2.79	4.90	87.15	1.72	1.66	1.76
	400	2.77	4.98	87.09	1.65	1.81	1.70
TA	0	3.31	6.67	82.27	3.14	3.38	1.24
	100	2.284	4.90	86.84	1.70	2.57	1.71
	200	2.68	4.89	85.60	1.94	3.18	1.70
	300	2.72	4.99	85.29	1.89	3.39	1.71
	400	2.66	5.08	84.64	2.41	3.38	1.83

Table S5. *J-V* curves of the PSCs devices ITO/PEDOT:PSS/PBDS-TZ:IT-4F /PDINO/Al without or with 90 °C TA treatment.

	V_{oc} (V)	J_{sc} (mA/cm^2)	FF (%)	PCE (%)
Without	0.802±0.003	16.8±0.35	59.0±0.43	8.00[b] (7.83[a]±0.25)
90 °C TA	0.800±0.005	17.1±0.42	60.5±0.47	8.34[b] (8.07[a]±0.32)

[a]The averages for photovoltaic parameters of each device are given in parentheses with mean variation obtained from 10 devices, and the ± refer to the standard deviation.
[b]Best device PCE.

Table S6. *J-V* curves of the PSCs devices ITO/ZnO/J71:IT-4F /MoO_3/Ag without or with TA treatment.

TA treatment	V_{oc} [V]	J_{sc} [mA cm^{-2}]	FF [%]	PCE [%]
w/o	0.764±0.003	18.69±0.35	56.98±0.45	8.23[b] (8.09[a]±0.15)
60 °C	0.759±0.003	18.78±0.36	62.74±0.35	9.06 [b] (8.78[a]±0.23)
90 °C	0.756±0.003	18.24±0.33	67.26±0.37	9.38 [b] (9.18[a]±0.22)
120 °C	0.734±0.003	17.86±0.37	65.74±0.42	8.73 [b] (8.57[a]±0.17)

[a]The averages for photovoltaic parameters of each device are given in parentheses with mean variation obtained from 10 devices, and the ± refer to the standard deviation.
[b]Best device PCE.