Electronic Supporting Information

Achieving Highly Efficient All-Polymer Solar Cells by Green-Solvent-

Processing under Ambient Atmosphere †

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Experimental Section

Materials

The polymer L14 was synthesized according to procedures reported in the literature.¹ CS₂, 1,2,4-TMB, *o*-XY, 1-MN and 1-PN are purchased from Sigma-Aldrich without further purification.

Methods.

UV-vis absorption spectra of polymer solution and films were recorded on a Shimadzu UV-3600 UV-VIS-NIR spectrophotometer. The cyclic voltammetry was used to determine the polymer energy levels in thin-film under argon atmosphere using a CHI760 Voltammetry Workstation with a saturated solution of 0.1 M tetra(*n*-butyl) ammonium hexafluorophosphate (Bu₄NPF₆) in acetonitrile (CH₃CN) as the supporting electrolyte. The energy levels were calculated using the following formula: $E_{LUMO} = -e(E_{red}^{onset} + 4.80) eV$; $E_{HOMO} = -e(E_{ox}^{onset} + 4.80) eV$.

Atomic force microscopy (AFM) measurements of two polymer blend films were conducted using a Dimension Icon Scanning Probe Microscope (Asylum Research, MFP-3D-Stand Alone) in the tapping mode. TEM micrographs were obtained on a Hitachi HT7700 microscope operating at 100 kV and equipped with an AMF-5016 charge-coupled device camera. Two-dimensional grazing incident wide-angle X-ray scattering (2D-GIWAXS) measurements were carried out at the PLS-II 9A U-SAXS beamline of Pohang Accelerator Laboratory, Korea.

Fabrication and Characterization of all-PSCs.

All-PSCs with a conventional structure of glass/ITO/PEDOT:PSS/binary and ternarybased active layers/PDINO are fabricated. Pre-patterned ITO-coated glass with a sheet resistance of $10 \Omega \text{ sq}^{-1}$ is used as the substrate, which is cleaned by sequential sonication in H₂O containing detergent, deionized H₂O, acetone, and isopropanol followed by drying in a vacuum oven and then UV-ozone (BZS250GF-TC, HWOTECH, Shenzhen) treatment for 15 min. PEDOT:PSS (Clevios P VP A1 4083) was spin-coated onto the UV-ozone-treated ITO substrates at 3500 rpm for 40 s then annealed at 150 °C for 15 min in air, forming ~35 nm film. The PEDOT:PSS-coated ITO substrates were transferred into an N2-filled glove box for subsequent steps. For CS2 processed all-PSCs, the total concentration of PM4 and L14 was fixed at 16 mg mL⁻¹, and the blend films were obtained by spin coating solutions of the polymers in CS₂ containing 1phenylnaphthalene (1-PN, 0.5 vol %). The solutions were stirred at room temperature for 6 h and heated at 35 °C for about 1 h before use. For 1,2,4-TMB processed all-PSCs, the total concentration of PM4 and L14 was fixed at 14 mg mL⁻¹. The solutions were stirred at room temperature for 6 h and heated at 80 °C for about 2 h before use. The blend films were obtained by spin coating 60 °C heated solutions of the polymers in 1,2,4-TMB containing 1-methoxynaphthalene (1-MN, 0.5 vol %). For o-XY processed all-PSCs, the total concentration of PM4 and L14 was fixed at 12 mg mL⁻¹. The solutions were stirred at room temperature for 6 h and heated at 80 °C for about 2 h before use to reach complete dissolution. The blend films were obtained by 60°C heated spin-coating technique of polymers dissolved in o-XY containing 0.5 vol % 1-MN. In addition, control devices based on PM6:L14 blend processed with CF solvent were also fabricated for comparison. The active layer with an optimal thickness of 100~110 nm is spin-coated onto PEDOT:PSS layer, followed by thermal annealing at 90°C for 5 min. Then PDINO was dissolved in methanol at the concentration of 1.0 mg mL⁻¹ and spin-coated at 3000 rpm for the 40s on the organic active layers. 110 nm Al is sequentially deposited on the active layer via thermal evaporation (*ca.* 1×10^{-5} Pa). All current-voltage (J-V) characteristics of the devices were measured under simulated AM1.5G irradiation (100 mW/cm²) using a Xe lamp-based SS-F5-3A Solar Simulator

(Enli Technology, Inc.). A Xe lamp equipped with an AM1.5G filter was used as the white light source. The light intensity was controlled with an NREL-calibrated Si solar cell with a KG-5 filter. The external quantum efficiency (EQE) was measured by a QE-R3011 measurement system (Enli Technology, Inc.).

SCLC Mobility Measurement.

Hole and electron mobilities were measured using the space-charge-limited-current (SCLC) method. The structure of ITO/PEDOT:PSS/active layer/MoO₃/Ag was used for hole-only devices and the structure of ITO/ZnO/active layer/Ca/Al was used for electron-only devices, respectively. For CS₂ processed all-PSCs, the total concentration of PM4 and L14 was fixed at 16 mg mL⁻¹, and the blend films were obtained by spin coating solutions of the polymers in CS₂ containing phenylnaphthalene (PN, 0.5 vol %). The solutions were stirred at room temperature for 6 h and heated at 35 °C for about 1 h before use. For 1,2,4-TMB processed all-PSCs, the total concentration of PM4 and L14 was fixed at 14 mg mL⁻¹. The solutions were stirred at room temperature for 6 h and heated at 80 °C for about 2 h before use. The blend films were obtained by spin coating 60 °C heated solutions of the polymers in 1,2,4-TMB containing 1methoxynaphthalene (1-MN, 0.5 vol %). For o-XY processed all-PSCs, the total concentration of PM4 and L14 was fixed at 12 mg mL⁻¹. The solutions were stirred at room temperature for 6 h and heated at 80 °C for about 2 h before use to reach complete dissolution. The blend films were obtained by 60°C heated spin-coating techniques of polymers dissolved in o-XY containing 0.5 vol % 1-MN. The SCLC mobilities were calculated by the MOTT-Gurney equation:

$$J = \frac{9}{8} \varepsilon_{\gamma} \varepsilon_0 \mu \frac{V^2}{L^3}$$

Where *J* is the current density, ε_r is the relative dielectric constant of active layer material, usually 2- 4 for organic/polymer semiconductors, herein we use a relative

dielectric constant of 3 for polymer. ε_0 is the permittivity of space, μ is the mobility of hole or electron and *d* is the active layer thickness, *V* is the internal voltage in the device, and $V = V_{appl} - V_{bi}$, where V_{appl} is the voltage applied to the device, and V_{bi} is the built-in voltage resulting from the relative work function difference between the two electrodes (in the hole-only and the electron-only devices, the V_{bi} values can be neglected).

Donor: Acceptor	V _{oc} (V)	$J_{\rm sc}$ (mA cm ⁻²)	FF (%)	PCE (%)	Ref.
PTzBI-Si:N2200	0.86	15.8	73	10.0	2
PM6:PN1	1.00	15.2	69	10.5	3
PBDB-T:BSS10	0.86	18.55	64	10.1	4
PM6:PFBDT-IDTIC	0.96	15.27	68	10.3	5
PTzBI-Si:N2200	0.85	17.2	77.9	11.5	6
PBDTTT-E-T: DCNBT-TPIC	0.70	22.52	64.8	10.22	7
PM6:PZ1	0.96	17.1	68.2	11.2	8
PM6:PF3-DTCO	0.943	15.75	68.2	10.13	9
PM6:PF2-DTSi	0.99	16.48	66.1	10.77	10
PBDB-T:PJ1	0.90	22.6	71	14.4	11
CD1:PBN-12	1.17	13.39	64	10.07	12
PM6:PYT	0.93	21.78	66.33	13.44	13
PBDB-T:PTPBT-ET _{0.3}	0.899	21.33	65.3	12.53	14
PM6:L14	0.96	20.6	72.1	14.3	15
PBDB-T:PF5-Y5	0.946	20.65	74.0	14.45	16
PM6:PY-IT	0.933	22.30	72.3	15.05	17
PTzBI-oF:PFA1	0.87	23.96	72.67	15.11	18
PBDB-T:PJ1	0.90	22.7	75.3	15.4	19
PBDB-T:PZT- γ	0.896	24.7	71.3	15.8	20
JD40:PJ1	0.91	23.2	75	15.8	21
PBDB-T:PYT	0.891	23.03	73.98	15.17	22
PM6:PY-IT	0.937	21.90	73.6	15.11	23
PM6:L14 (o-XY)	0.953	22.10	74.10	15.62	This Work

Table S1. Summary of photovoltaic performance parameters of binary ASPCsprocessed with halogenated solvent with PCEs over 10 %.



Fig. S1 Variation of solubility of PM6:L14 blend in different green solvents.

Solvents	Boiling Point (°C)	V _{oc} (V)	J _{sc} (mA cm ⁻²)	FF (%)	PCE (%)
Anisole	154	0.018	4.06	24.90	0.02
MA	172	0.108	9.08	26.94	0.26
CPME	106	0.203	5.17	29.84	0.31
2-MeTHF	80	0.931	11.04	49.28	5.06
THF	66	0.910	13.94	47.81	6.06
Tul	111	0.952	19.65	64.17	12.00
<i>p</i> -XY	138	0.958	19.32	67.34	12.45
o-XY	144	0.953	22.10	74.10	15.62
1,2,4-TMB	168	0.949	20.97	69.53	13.84
CS_2	46	0.948	21.09	65.17	13.03

Table S2. Photovoltaic performance parameters of OSCs based on PM6:L14 active layers in various green solvents.



Fig. S2 The cyclic voltammetry of polymers (a) PM6 and (b) L14. (c) Energy level diagram of the materials used in all-PSCs.

Solvents	Concentration (mg mL ⁻¹)	Solubility ^a (80 °C)	Solubility ^a (60 °C)	Solubility ^a (35 °C)	Solubility ^{<i>a</i>} (R.T. ≈ 25 °C)
	16	-	-	++++	+
CS ₂	18	-	-	++++	+
	20	-	-	++++	+
	14	+++++	++++	+++ b	++
1,2,4-TMB	16	+++++	++++	+++ b	++
	18	+++++	++++	+++ b	+
	12	+++++	++++	+++ b	++
o-XY	14	+++++	++++	+++ b	++
	16	+++++	++++	+++ b	++

Table S3. The solubility test of PM6:L14 polymer blend at different temperatures with certain concentrations in various non-halogenated solvents.

^a +++++ represents good solubility, ++++ represents soluble with low viscosity, +++ represents soluble but forming nearly gel state, ++ represents poor solubility, + represents bad solubility; *^b* stirred at 80 °C for 2 h and then cooled down to 35 °C.



Fig. S3 The chemical structures of different non-halogenated solvent additives used in this work.



Fig. S4 (a) J–V characteristics and (b) EQE curves for devices based on CF solvent.

Calman ta d	A J J 44	V _{oc}	$J_{\rm sc}$	FF	PCE _{ave} ^b	PCE _{max}
Solvents "	Additives	(V)	(mA cm ⁻²)	(%)	(%)	(%)
	w/o	0.949	20.42	60.74	10.93	11.77
	ODT	0.949	19.61	58.29	10.12	10.85
CS	DPE	0.947	20.17	62.58	11.01	11.95
	NMP	0.945	20.93	61.17	11.18	12.10
	1-PN	0.948	21.09	65.17	12.06	13.03
	1-MN	0.948	21.25	63.48	11.63	12.57
	w/o	0.945	20.84	65.73	12.59	12.90
	ODT	0.949	18.63	66.32	11.47	11.72
1,2,4-TMB	DPE	0.946	20.32	68.42	12.85	13.15
	NMP	0.948	21.16	63.54	12.36	12.75
	1-PN	0.949	20.76	62.59	12.01	12.33

Table S4. Photovoltaic performance parameters of all-PSCs consisting of PM6:L14

 active layers processed with various non-halogenated solvent additives.

	1-MN	0.949	20.97	69.53	13.43	13.84
	w/o	0.955	20.61	68.16	13.12	13.41
	ODT	0.953	20.73	72.19	13.93	14.26
- VV	DPE	0.952	21.46	70.26	14.09	14.35
<i>0-X</i> Y	NMP	0.954	20.13	67.23	12.46	12.86
	1-PN	0.953	21.58	72.49	14.67	14.91
	1-MN	0.953	22.10	74.10	15.31	15.62

^{*a*} The device area is 4.5 mm²; ^{*b*} The average PCEs are obtained from 10 devices.

Table S5. Photovoltaic performance parameters of all-PSCs based on PM6:L14 active layer with different thicknesses.

Solvente (Thickness	Voc	$J_{\rm sc}$	FF	PCE _{ave} ^b	PCE _{max}
Solvents "	(nm)	(V)	(mA cm ⁻²)	(%)	(%)	(%)
	91	0.949	20.85	64.75	11.92	12.81
CS ₂	110	0.948	21.09	65.17	12.06	13.03
	127	0.946	21.46	60.58	11.49	12.30
	93	0.95	20.69	68.72	13.18	13.51
1,2,4-TMB	104	0.949	20.97	69.53	13.43	13.84
	121	0.947	21.13	66.41	12.97	13.29
	87	0.954	21.72	73.02	14.83	15.13
<i>o</i> -XY	101	0.953	22.10	74.10	15.31	15.62
	116	0.951	22.08	70.39	14.46	14.78

^{*a*} The device area is 4.5 mm²; ^{*b*} The average PCEs are obtained from 10 devices.



Fig. S5 Plots of the PCE *versus* FF for highly-efficient all-PSCs reported in the literature.

Donor: Acceptor	Solvents	V _{oc} (V)	J _{sc} (mA cm ⁻²)	FF (%)	PCE (%)	Ref.
PPDT2FBT-A:P(NDIDEG-T)	Water/ethanol	0.66	6.43	44	2.15	24
HFQx-T:N2200	THF	0.90	11.69	59	6.20	25
HFAQx-T:N2200	THF	0.92	12.47	65	7.45	25
PBDT-TS1:PPDIODT	Anisole	0.76	15.72	55.11	6.58	26
PiI-2T-PS10:P(TP)	Toluene	0.98	9.93	51	5.10	27
PBDT-TS1:PPDIODT	o-MA	0.74	13.77	52.46	5.6	28
PTB7-Th:PDI-V	THF	0.74	15.5	70.0	8.1	29

 Table S6. Representative examples of APSCs processed by various green solvents.

PM6:L14	<i>o</i> -XY	0.953	22.10	74.10	15.62	This Work
PBDB-T:PJ1:PJ2	o-XY	0.91	21.46	72.98	14.28	38
PBDB-T:PJ1	o-XY	0.90	21.92	72.61	14.34	38
PTzBI-oF:PS1	2-MeTHF	0.92	22.47	66.70	13.8	37
PTzBI-Si:N2200	CPME	0.87	14.6	72.7	9.3	36
PBDT5-TPD ₄ :P(NDI2HD-T)	2-MeTHF	1.02	14.42	55.75	8.20	35
PBDT-TPD:P(NDI2HD-T)	2-MeTHF	1.04	11.48	53.69	6.41	35
J51:PTB7-Th:N2200	CPME	0.82	17.27	67.8	9.60	34
J51:N2200	CPME	0.82	14.19	60.2	7.58	34
PTzBI-Si:N2200	2-MeTHF	0.88	17.62	75.78	11.76	33
PTzBI-Si:N2200	THF	0.85	13.6	63.4	7.6	32
PTzBI-Si:N2200	2-MeTHF	0.85	15.6	73.1	10.1	32
PTzBI-Si:N2200	СРМЕ	0.85	16.5	77.9	11.0	32
PTzBI-Si:N2200	2-MeTHF	0.865	15.76	73.76	10.1	31
PTzBI:N2200 _{HW}	2-MeTHF	0.849	15.17	70.36	9.16	30
PTzBI:N2200 _{MW}	2-MeTHF	0.85	11.48	57.17	5.67	30
PTzBI:N2200 _{LW}	2-MeTHF	0.85	10.39	51.98	4.86	30



Fig. S6 (a) *J-V* characteristics in dark, (b) dependence of J_{sc} and (c) V_{oc} on different light intensity for devices based on PM6:L14 in CS₂, 1,2,4-TMB and *o*-XY green solvents, respectively.

Table S7. J_{ph} at short-circuit condition, J_{sat} and P_{diss} parameters of all-PSCs based on PM6:L14 blends processed by CS₂, 1,2,4-TMB and *o*-XY green solvents, respectively.

Device	J_{ph}	J _{sat}	Thickness	G _{max}	P _{diss}
Device	(mA cm ⁻²) ^{<i>a</i>}	(mA cm ⁻²) ^a	(nm)	$(m^{-3} s^{-1})^{b}$	(%)
PM6:L14 (CS ₂)	21.09	22.51	110	1.28×10 ²⁸	93.68
PM6:L14 (1,2,4-TMB)	20.97	22.19	104	1.33×10^{28}	94.52
PM6:L14 (o-XY)	22.10	22.68	101	1.40×10 ²⁸	97.43

^{*a*}Average values are obtained from 5 devices. ^{*b*}Calculated by $G_{\text{max}} = J_{\text{sat}} / q \cdot L$.



Fig. S7 (a) Hole and (b) electron mobilities from SCLC model for devices based on PM6:L14 in CS₂, 1,2,4-TMB and *o*-XY green solvents, respectively.

Table S8. Hole and electron mobilities for all-PSCs based on PM6:L14 blends processed by CS₂, 1,2,4-TMB and *o*-XY green solvents, respectively.

Device ^a	$\mu_{\rm h} (\times 10^{-4} {\rm cm}^2 {\rm V}^{-1} {\rm s}^{-1})^b$	$\mu_{\rm e} (\times 10^{-4}{\rm cm}^2{ m V}^{-1}{ m s}^{-1})^b$	$\mu_{\rm h}/\mu_{\rm e}$
PM6:L14 (CS ₂)	6.48	3.84	1.69
PM6:L14 (1,2,4-TMB)	7.25	4.98	1.46
PM6:L14 (o-XY)	7.90	7.04	1.12

*^a*The device area is 4.5 mm². *^b*Average values with standard deviation were obtained from 20 devices.



Fig. S8. Statistical analysis of (a) hole and (b) electron mobilities for the corresponding devices.

Table S9. Summary of IP (100) and OOP (010) coherence lengths of the blend films processed by different green solvents.

Sampla	IP (10	0) peak	OOP (010) peak		
Sample	<i>q</i> (Å)	q (Å) $L_{\rm c}$ (nm)		L_{c} (nm)	
CS_2	0.30	9.63	1.65	1.60	
1,2,4-TMB	0.30	13.40	1.67	1.81	
o-XY	0.30	16.59	1.66	1.84	



Fig. S9 EQE curves for all-PSCs processed with CF and *o*-XY solvents under ambient condition.



Fig. S10 *J*–*V* characteristics of the corresponding device performance under ambient condition with CF and *o*-XY as fabricating solvent and PNDIT-F3N as ETL.

Table S10. Photovoltaic parameters of all-PSCs based on PM6:L14 processed in ambient condition with CF and *o*-XY as fabricating solvent and PNDIT-F3N as ETL under the illumination of AM 1.5G, 100 mW cm⁻².

Solvonta	V _{oc} (V) ^b (r	J _{sc}	FF	PCE _{max}	PCE _{ave}
Solvent		(mA/cm ²) ^b	(%) ^b	(%) ^b	(%) ^b
CF+Air	0.93	21.25	71.25	14.10	13.71
o-XY+Air	0.94	21.61	71.94	14.64	14.28
o-XY+Air (w/o MN)	0.93	21.03	66.32	12.96	12.64

*^a*The device area is 4.5 mm²; ^{*b*}Average values with standard deviation from 15 devices included in parenthesis.



Fig. S11 Contact angles of water on (a) CF and (b) o-XY processed blend films of

PM6:L14.

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