Supporting Information

Computer-aided Screening of Conjugated Polymers for Organic Solar Cell: Classification by Random Forest

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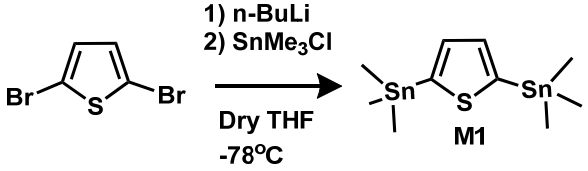
This supporting information presents the following contents.

Experimental

Machine learning. The experimental data (~1200) were manually collected from the literatures. The complied data are provided as a text file and the corresponding references are given in Supporting References (S1–S503). Modelings based on ANN and RF were performed on a notebook computer using "R studio" (a free software, https://www.rstudio.com/). The repeating unit of a polymer structure was converted to a SMILES expression using ChemDraw software and then to a MACCS, PubChem, or ECPF6 key using RDKit library of R studio.

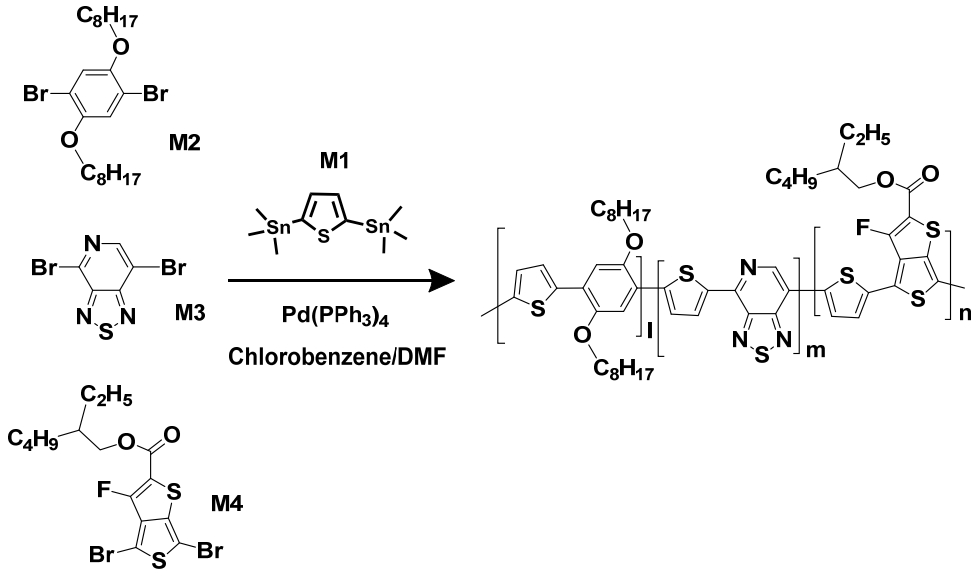
Synthesis of 2,5-bis(trimethylstannyl)thiophene (M1). 2,5-dibromothiophene (1.074 g, 4.44 mmol) and dehydrated tetrahydrofuran (30 mL) were added to a dried 100 mL flask under nitrogen. The reaction mixture was cooled to –78°C in an acetone dry ice bath, and *n*-butyllithium (6.18 mL, 1.63 M *n*-hexane solution) was added slowly via dropping funnel for 15 minutes. After stirring at –78°C for 1 h, a dry THF (18 ml) solution of trimethyltin chloride (2.215 g, 11.1 mmol) was added slowly via syringe in 15 minutes. Then the dry ice bath was removed and the reaction mixture was allowed to warm to room temperature. After stirring overnight, water was added and the mixture was extracted with *n*-hexane. The organic layer was washed with brine and dried over anhydrous $MgSO_4$. Solvent was removed under vacuum and purified by recrystallization in 20 mL isopropanol and a recycling HPLC using chloroform as the eluent. The product was obtained as a white crystal (0.795 g, 44% yield). ^{1}H NMR (400 MHz, $CDCl_3$): δ 0.34 (s, 18H), 7.37 (s, 2H).



Scheme 1. Synthesis of 2,5-bis(trimethylstannyl)thiophene.

Synthesis of P1 polymer. 1,4-Dibromo-2,5-bis(decyloxy)benzene (M2) was purchaed from Aldrich Corp. 4,7-Dibromo[1,2,5]thiadiazolo[3,4-*c*]pyridine (M3) and 2-Ethylhexyl 4,6-dibromo-3-fluorothieno[3,4-*b*]thiophene-2-carboxylate (M4) were purchased from Tokyo Chemical Industry (TCI) Co., Ltd. These monomers (M2–M4) were used without purification. Stille cross-coupling polymerization reactions were conducted in an Anton Paar Monowave 300 microwave reactor. Stannylated thiophene (M1, 75 mg, 60 μmol), M2 (29.6 mg, 60 μmol), M3 (17.7 mg, 60 μmol), M4 (28.4 mg, 60 μmol), $Pd(PPh_3)_4$ catalyst (4.8 mg, 4.2 μmol), anhydrous chlorobenzene (CB, 6 mL), and dimethylformamide (DMF, 0.3 mL) were added to

a reaction vial equipped with a stirrer bar under a flow of argon. The reaction tube was sealed and transferred to the reactor. Polymerization was performed at 180 °C with stirring at 800 rpm for 35 min. The reaction mixture was poured into methanol and the precipitate was then purified by Soxhlet extraction in methanol and hexane. The residue was reprecipitated from methanol to afford a dark blue solid (26 mg, yield: 17 wt%, 42 mol%). M_w = 1.1 kg mol^{-1}, M_n = 0.7 kg mol^{-1}, PDI = 1.6.



Scheme 2. Synthesis of P1.

General measurements. Steady-state photoabsorption spectroscopy was performed using a Jasco V-570 UV-vis spectrophotometer. Molecular weight of the polymer was determined using the gel permeation chromatography (GPC) method with polystyrene standards. GPC analyses were performed with chloroform as the eluent at a flow rate of 1 cm^3 min^{-1} at 40 °C, on a SHIMADZU LC-20AT, CBM-20A, CTO-20A chromatograph connected to a SHIMADZU SPD-M20A UV-vis detector. Photoelectron yield spectroscopy (PYS) of a polymer film on an indium-tin-oxide (ITO) glass was performed on a Bunko Keike BIP-KV2016K instrument. Atomic force microscopy (AFM) was carried out on a Bruker Innova AFM microscope. Film thicknesses were measured using a Bruker Dektak XT surface profiler. Powder X-ray diffraction (XRD) data were collected on a Rigaku MiniFlex-600 X-ray spectrometer using Cu−K$_\alpha$ radiation (λ = 1.54187 Å) at room temperature in air. Xe-flash time-resolved microwave conductivity (Xe-TRMC at ~9.1 GHz) experiments were conducted with a pseudo-sunlight white-light pulse (0.3 mJ cm^{-2} pulse^{-1}, 10 μs pulse duration) as the excitation source. The photoconductivity $\Delta\sigma$ was obtained by applying the formula $\Delta P_r/(AP_r)$,

where ΔP_r, A, and P_r are the transient power change of the reflected microwave power, the sensitivity factor, and the reflected microwave power, respectively.

OPV device. PCBM was purchased from Frontier Carbon Inc. and used as the n-type material. Chlorobenzene and DIO were purchased from Wako Chemical Inc. and TCI, respectively and used as received. A 0.8wt% chlorobenzene solution of P1:PCBM = 1: 2 (weight) with/without 3 vol% DIO was spun-coat on a ZnO layer. The device configuration was: ITO (120−160 nm)/ZnO (30 nm)/BHJ active layer (~180 nm)/MoO_3 (10 nm)/Ag (100 nm) with an active area of 7.1 mm^2. Current-voltage (*J-V*) curves were measured using a ADCMT Corp., 6241A source/monitor under AM1.5G solar illumination at 100 mW cm^{-2} (1 sun, monitored by a calibrated standard cell, Bunko Keiki SM-250KD) using a 300 W solar simulator (SAN-EI Corp., XES-301S).

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Supporting Table

Table S1. Summary of P1:PCBM = 1:2 OPV device performance.[a]

Solvent	J_{sc} /mA cm^{-2}	V_{oc} / V	FF	PCE / %	# of cells
CB with 3vol%DIO	2.28 ± 0.09 (2.40)	0.537 ± 0.028 (0.559)	0.383 ± 0.026 (0.418)	0.47 ± 0.05 (0.53)	15
CB	1.33 ± 0.02 (1.37)	0.482 ± 0.006 (0.490)	0.406 ± 0.007 (0.412)	0.26 ± 0.01 (0.27)	15

[a] Glass/ITO/ZnO/P1:PCBM/MoO_x/Ag. The value in the brackets is the maximum value. The thicknesses of the active layer were ~180 nm.

Table S2. Group ranking (A, B. …) of PCE (%) used for RF classification.

# of fraction (n)	A	B	C	D	E	F	G	H	I	J
2	>4.0	4.0–0.0	-	-	-	-	-	-	-	-
3	>5.2	5.2–2.8	2.8–0.0	-	-	-	-	-	-	-
4	>5.8	5.8–4.0	4.0–2.3	2.3–0.0	-	-	-	-	-	-
5	>6.2	6.2–4.8	4.8–3.3	3.3–1.8	1.8–0.0	-	-	-	-	-
6	>6.5	6.5–5.2	5.2–4.0	4.0–2.8	2.8–1.5	1.5–0.0	-	-	-	-
7	>6.7	6.7–5.5	5.5–4.5	4.5–3.4	3.4–2.6	2.6–1.3	1.3–0.0	-	-	-
8	>7.0	7.0–5.8	5.8–5.0	5.0–4.0	4.0–3.1	3.1–2.3	2.3–1.2	1.2–0.0	-	-
9	>7.2	7.2–6.1	6.1–5.2	5.2–4.5	4.5–3.5	3.5–2.8	2.8–2.1	2.1–1.1	1.1–0.0	-
10	>7.6	7.3–6.2	6.2–5.5	5.5–4.8	4.8–4.0	4.0–3.3	3.3–2.6	2.6–1.8	1.8–1.0	1.0–0.0

Supporting Figures.

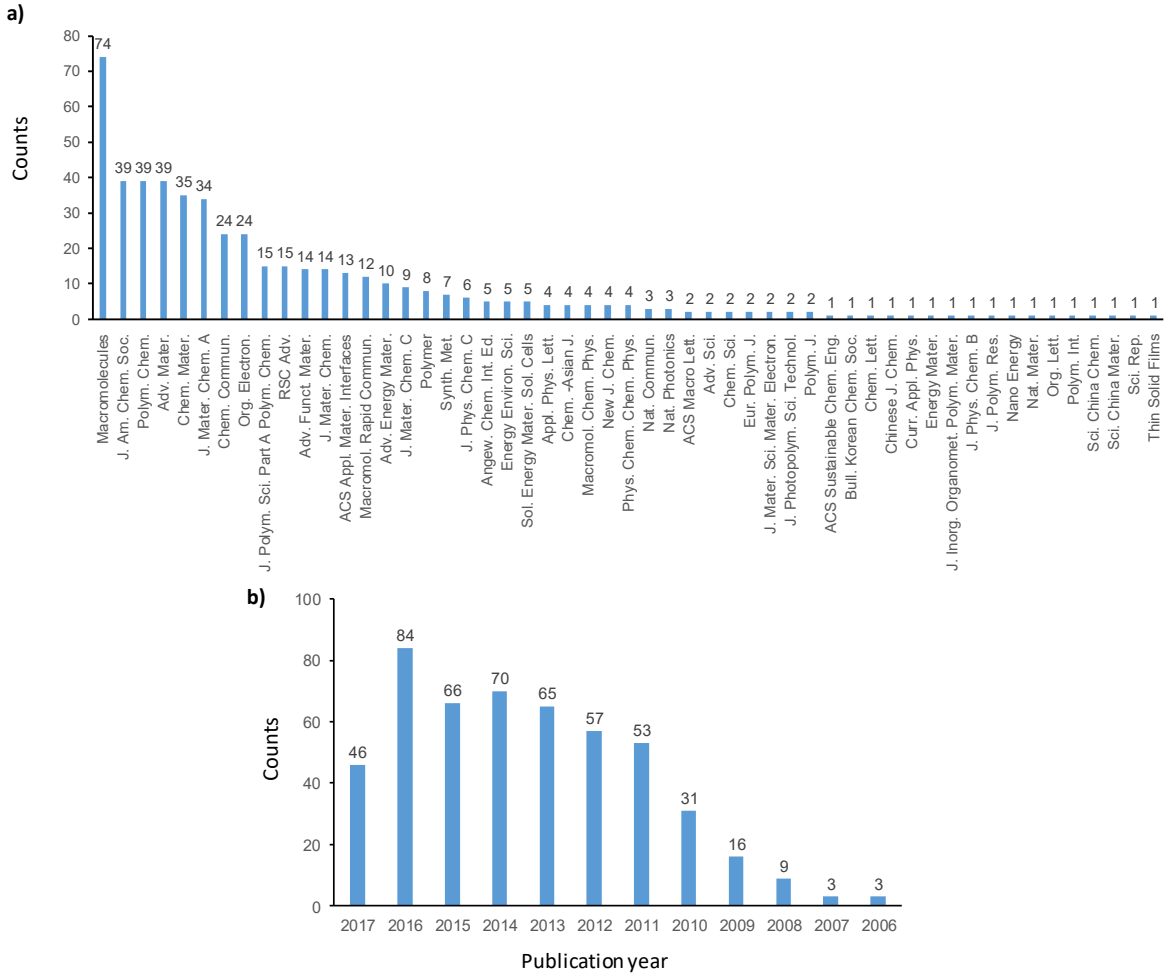

Figure S1. Statistics of collected experimental device data. (a) Sorted by journal. (b) Sorted by publication year.

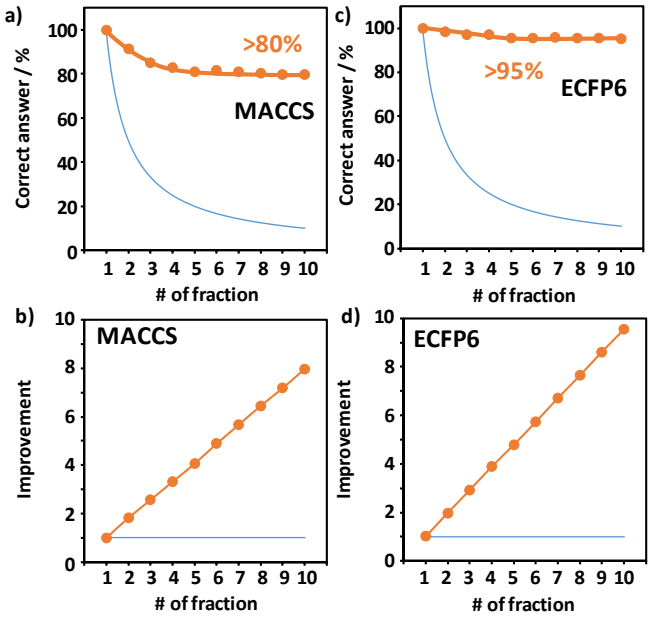

Figure S2. Correct answer obtained by the RF model when the test data and training data are identical. (a)(c) Corrected answer vs the number of fraction (*n*) obtained by the RF model with (b) MACSS and (d) ECFP6. (b)(d) Improvement of correct answer of the RF model using (b) MACCS and (d) ECFP6. The orange circles are the results of RF modeling, while the blue line indicates the random classification.

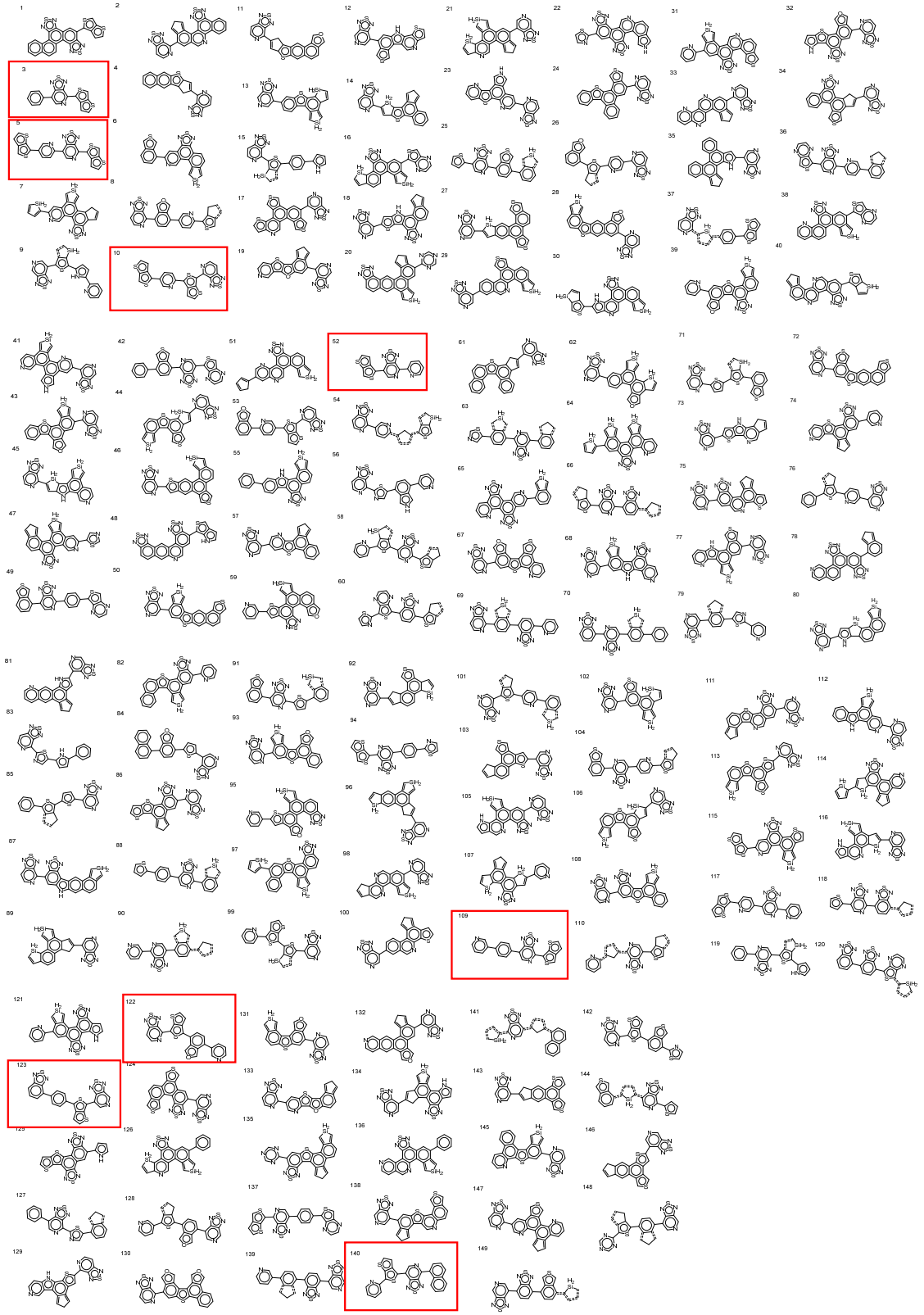

Figure S3. Chemical structures of 146 molecules extracted from 1000 molecules by using the RF screening with MACCS key (n = 4). These 146 molecules are ranked at the highest PCE group (>5.7%). The 1000 molecules were downloaded from the CEP website (see main text). The molecules enclosed by the red rectangles have a similar structure. Beside, they are relatively simple and relevent to a synthesis. The molecule of No. 3 (left top) was selected as the candidate.

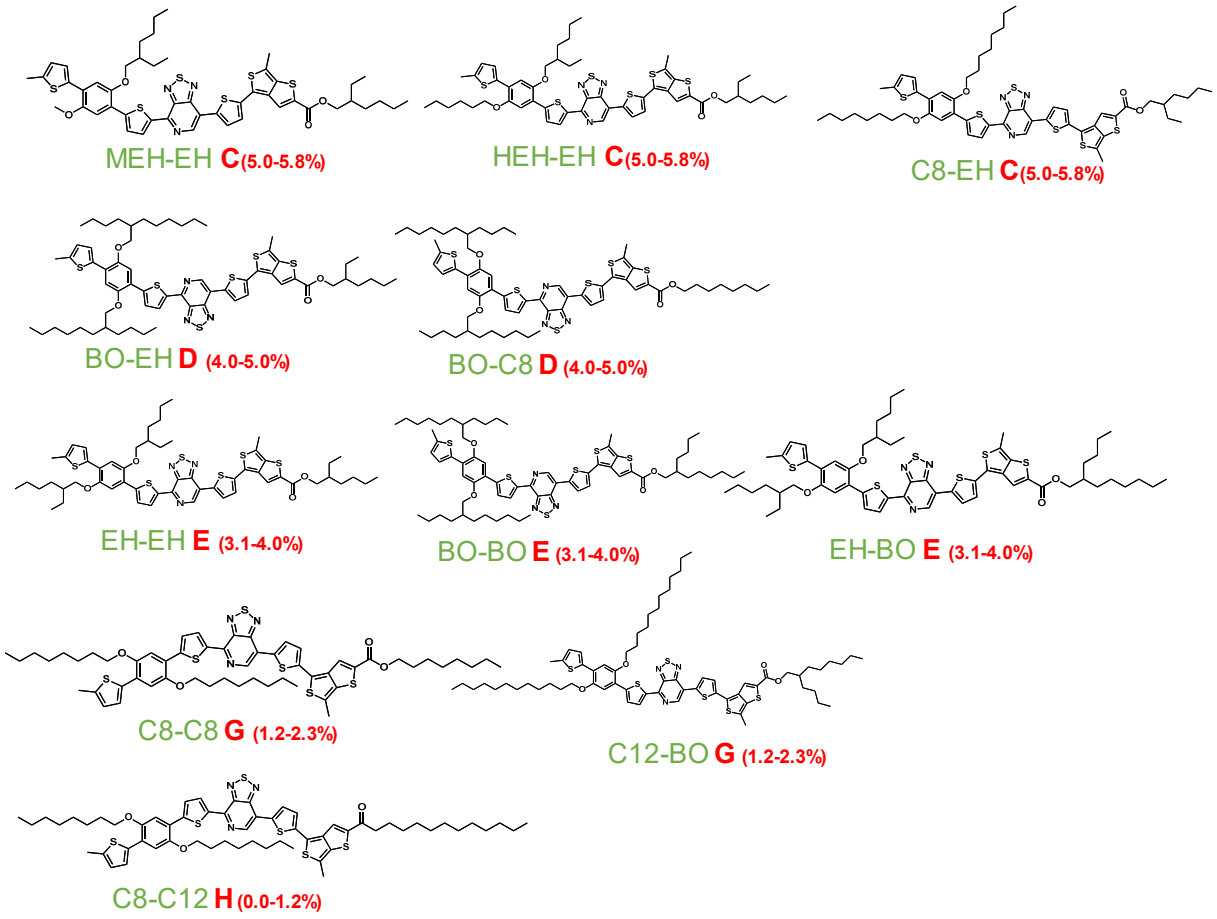

Figure S4. Screening of alkyl side chain by using RF with ECPF6 key (n = 8). The caption in green color represents the alkyl side chains. The label in red color is the predicted group along with its range of PCE.

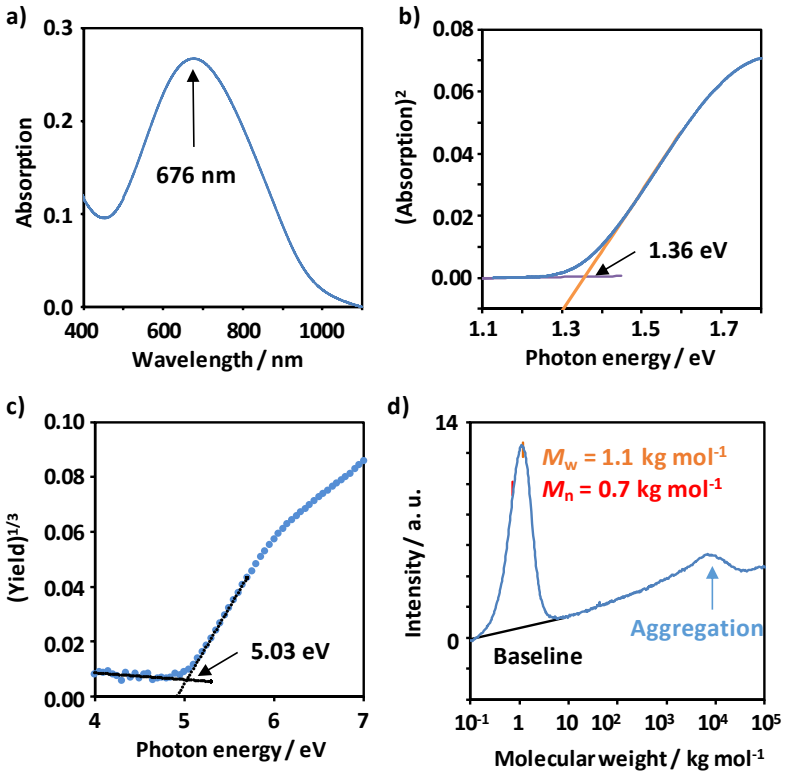

Figure S5. Optical, electronic, and polymeric data of P1. (a) UV-vis absorption spectrum in the film state. (b) Extrapolation to evaluate the optical bandgap. (c) PYS spectrum to evaluate HOMO level. (d) GPC profile.

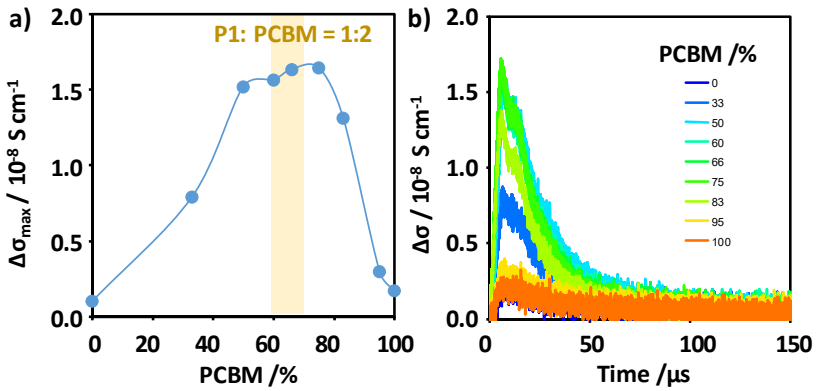

Figure S6. TRMC evaluation using a white light from a Xe lamp (Xe-TRMC). (a) Photoconductivity maxima ($\Delta\sigma_{max}$) as a function of P1: PCBM blend ratio. The films were prepared by drop-casting a chlorobenzene solution. The 0 and 100 % PCBM represent the pristine P1 film and pristine PCBM film, respectively. (b) Corresponding transients.

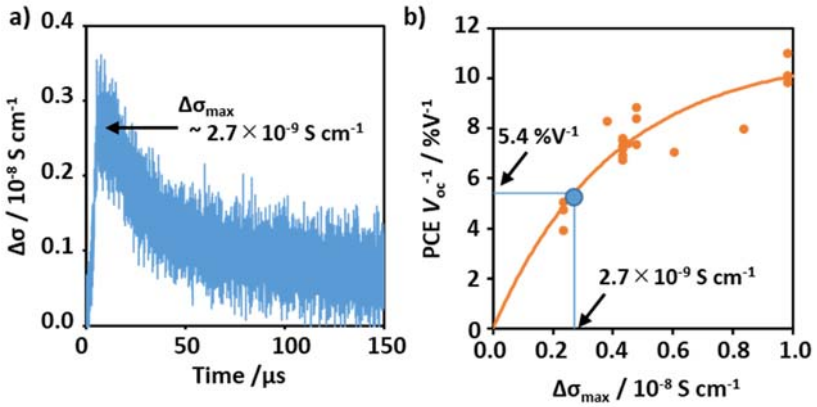

Figure S7. TRMC evaluation using a white light from a Xe lamp (Xe-TRMC) for a spin-coated film. (a) Photoconductivity transient of a P1:PCBM=1:2 film. The film was prepared by spin-coating from a chlorobenzene solution with 3 vol% DIO. (b) Correlation between PCE V_{oc}^{-1} vs $\Delta\sigma_{max}$ obtained for various polymer-fullerene film using a Xe-TRMC. The orange dot is taken from Ref. 52 in the main text. The orange solid line is the interpolated curve. The blue circle on the interpolated curve is the result of P1:PCBM film, which corresponds to $\Delta\sigma_{max} = 2.7 \times 10^{-9}$ S cm^{-1} and PCE $V_{oc}^{-1} = 5.4\%$ V^{-1}.

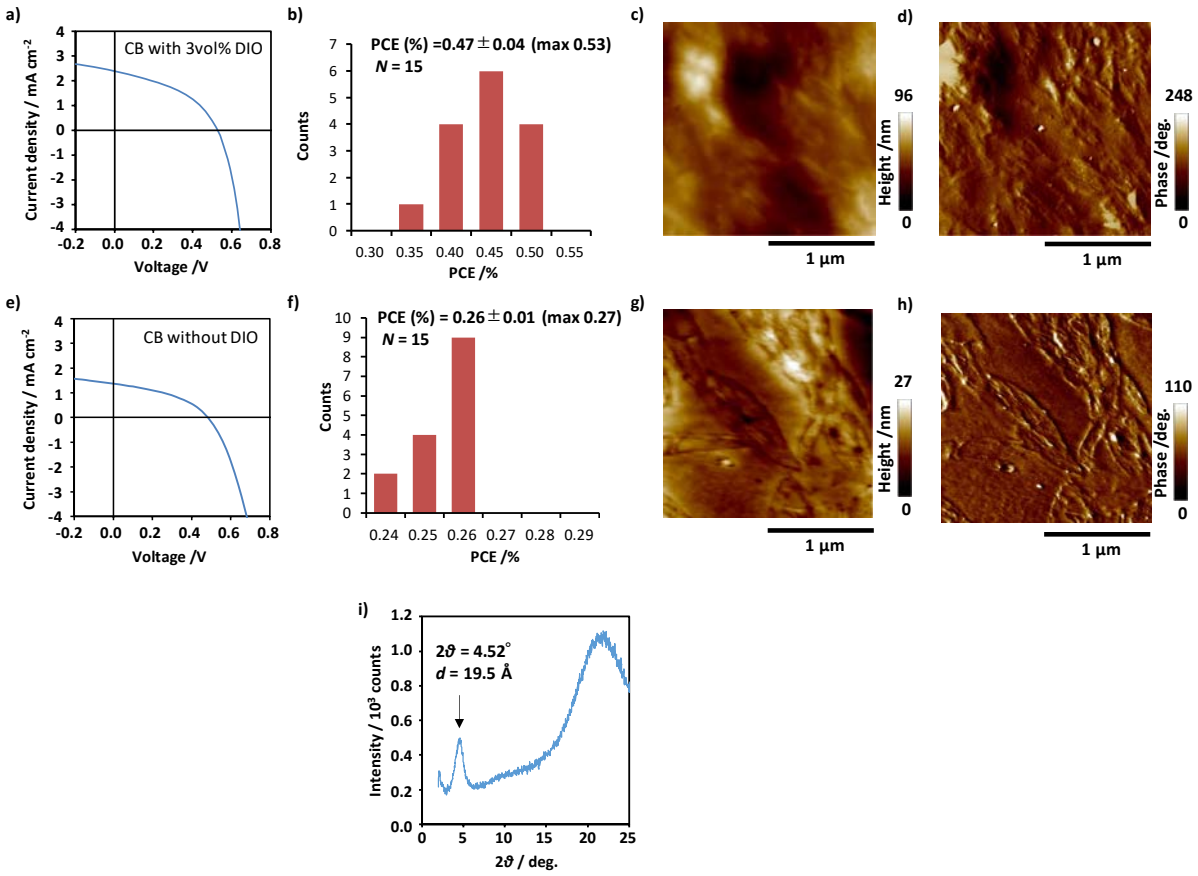

Figure S8. OPV device characterization of P1:PCBM = 1:2 prepared from (a)-(d) a chlorobenzene solution with 3vol% DIO and (e)-(h) a chlorobenzene solution. (a)(e) Current density-voltage curve of the best-performing device. (b)(f) Statistics of PCE. The number of the cells is 15. (c)(g) Topography and (d)(h) phase images of the device surface obtained by AFM. (i) XRD spectrum of the pristine P1 film.