Electronic Supplementary Information

A novel 9*H*-indeno[1,2-*b*]pyrazine-2,3-dicarbonitrile end group for efficient non-fullerene small molecule acceptor

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General Information

All solvents and reagents were used as received from commercial sources and used without further purification. 1*H*-indene-1,2(3*H*)-dione (compound **1**), 2,3-diaminomaleonitrile (compound **2**), INIC and polymer donor PBDB-T were purchased from commercial sources. ^{1}H NMR and ^{13}C NMR spectra were recorded on a Bruker Advanced II (400 MHz) spectrometer. The high resolution mass spectra (HRMS) and matrix-assisted laser desorption/ionization time of flight mass spectrometry (MALDI-TOF-MS) were performed on Thermo Scientific LTQ Orbitrap Xl using ESI and 5800 MALDI-TOF/TOF mass spectrometry (AB SCIEX, USA) in positive mode, respectively. UV-vis spectra were measured using a Shimadzu UV-2500 recording spectrophotometer. Cyclic voltammetry (CV) measurements of targeted SMA thin films were conducted on a CHI voltammetric analyzer in acetonitrile solution with 0.1 M tetrabutylammonium hexafluorophosphate (*n*-

Bu_4NPF_6) as supporting electrolyte at room temperature by using a scan rate of 100 mV s^{-1} and conventional three-electrode configuration consisting of a platinum working electrode with 2 mm diameter, a platinum wire counter electrode and a Ag/AgCl wire reference electrode. BTOIPC was calculated by using the density funcyional theory (DFT) with Gaussian 16. Ground state geometry optimization was performed using B3LYP exchange-correlation functional, the def2-SVP basis set and the density functional dispersion correction with Becke-Johnson damping.[1] Atomic force microscopy (AFM) images were obtained by using a SPM9700 microscope in the dynamic-model. Transmission electron microscopy (TEM) images of the active layers were obtained by using a JEOL JEM-1400 transmission electron microscope operated at 80 kV.

Synthesis

Synthesis of ***IPC***: To a 100 ml round-bottom flask, compound **1** (1.0 g, 6.85 mmol), compound **2** (0.74 g, 6.85 mmol) and acetic acid (30 ml) were added. The mixture were heated to reflux and stirred for 3h. After cooling to room temperature, the mixture was extracted with chloroform and washed with water for three times. The collected organic phase was evaporated and the residue was purified by column chromatography on silica gel using a mixture solvent as eluent (petroleum ether/dichloromethane, v/v = 1/1) to give a yellow solid (1.24 g, 83%). ^{1}H NMR (400 MHz, $CDCl_3$), δ (ppm): 8.21 (d, J = 8 Hz, 1H), 7.68-7.75 (m, 2H), 7.60-7.64 (m, 1H), 4.18 (s, 2H). ^{13}C NMR (100 MHz, $CDCl_3$), δ (ppm): 162.36, 157.29, 144.11, 135.34, 133.60, 129.35, 129.08, 126.04, 124.14, 113.90, 113.71, 36.33. HRMS m/z: $[M+H]^+$

calcd. for $C_{13}H_7N_4^+$, 219.06652, found 219.06627.

Synthesis of ***BTOIPC****:* To a 100 ml round bottom flask, BTO-CHO (200 mg, 0.15 mmol) and IPC (200 mg, 0.9 mmol) were added under argon protection. Then, deoxidized chloroform (40 ml) was added and stirred for a while when piperidine (0.1 ml) was added. The mixture was kept stirring at 70°C for 16 h. After removal of chloroform of reaction mixture under reduced pressure, 100 ml methanol was added and the precipitate was collected by filtration. The residue was purified by column chromatography on silica gel using a mixture solvent as eluent (petroleum ether/dichloromethane, v/v = 1/1) to give a dark solid (104 mg, 40%).^{1}H NMR (400 MHz, $CDCl_3$), δ (ppm): 8.25 (s, 2H), 8.14 (d, *J* = 7.6 Hz, 2H), 7.90 (s, 2H), 7.81 (d, *J* = 7.6 Hz, 2H), 7.60-7.65 (m, 2H), 7.49 (t, *J* = 7.6 Hz, 2H), 7.40-7.43 (m, 8H), 7.09-7.13 (m, 8H), 3.57 (s, 4H), 2.56 (t, *J* = 7.2 Hz, 8H), 1.56-1.60 (m, 10H), 1.25-1.37 (m, 40H), 0.83-0.89 (m, 32H). ^{13}C NMR (100 MHz, $CDCl_3$), δ (ppm): 155.16, 154.22, 153.44, 150.24, 145.83, 142.64, 142.09, 141.94, 138.25, 136.34, 135.23, 133.99, 132.88, 131.00, 130.04, 128.53, 128.30, 128.09, 128.00, 123.65, 121. 27, 119.39, 114.60, 114.45, 63.89, 35.57, 31.71, 31.27, 31.23, 29.23, 23.43, 22.84, 22.71, 22.59, 14.27, 14.15, 14.10, 10.98, 10.88. MALDI-TOF-MS m/z: [M] calcd. for $C_{112}H_{114}N_8O_2S_4$, 1730.79, found 1730.88.

Photovoltaic devices fabrication and characterization

The OSCs were fabricated with an conventional device configuration of

ITO/PEDOT:PSS/PBDB-T:BTOIPC/PDIN/Al. The patterned ITO-coated glass was scrubbed by detergent and then cleaned inside an ultrasonic bath by using deionized water, acetone, and isopropyl alcohol sequentially. Then, the ITO substrates was fast dried by using high pure nitrogen gas and then treated by oxygen plasma for 1 min to improve its work function and clearance. A thin PEDOT: PSS (Heraeus Clevios P VP A 4083) layer with a thickness of about 40 nm was spin-cast onto the ITO substrates at 4000 rmp for 40 s, and then dried at 150 °C for 10 min in air. The PEDOT:PSS coated ITO was transferred to a N_2 filled glove-box for further processing. The 1:1 weight ratio of donor (PBDB-T) and small molecular acceptor (BTOIPC) were dissolved in chlorobenzene (CB) to form a solution concentration of 18 mg/ml with 0.8% 1,8-diiodoctane (DIO) additive. The best-performance device was fabricated by spin-coating the mixture at 2000 rpm for 1 min and controlled the active layer thickness of ~100 nm measured by Ambios Technology XP-2 stylus Profiler. Subsequently, the active layer coated substrates were quickly transferred to a glove-box integrated thermal evaporator for PDIN and Al deposition. PDIN and Al layers were subsequently evaporated onto the active layer through a shadow mask at a vacuum pressure of ≈ 10^{-4} Pa to form top electrode. The active area of cells is about 3.8 mm^2, which is defined by the vertical overlap of ITO cathode and Al anode. The current-voltage (*J-V*) characteristic curves of all devices were measured by using a Keithley 2400 Source Meter in a high-purity nitrogen-filled glove box. AM 1.5G irradiation at 100 mW/cm^2 provided by a XES-40S2 (SAN-EI Electric Co., Ltd.) solar simulator (AAA grade, 70×70 mm^2 photobeam size), which was calibrated by

standard silicon solar cells (purchased from Zolix INSTRUMENTS CO. LTD). The external quantum efficiency (EQE) spectra of all devices were measured in air conditions by a Zolix Solar Cell Scan 100.

SCLC measurements

The electron-only SCLC devices were a stack of ITO/ZnO/PBDB-T:BTOIPC/PFN/Al, and the hole-only devices were a stack of ITO/PEDOT:PSS/PBDB-T:BTOIPC/MoO_x/Ag. The electron-only and hole-only SCLC devices fabrication processing methods are same with those for solar cell. The charge carrier mobility was determined by fitting the dark current to the model of a single carrier SCLC according to the equation:

$$J = \frac{9\varepsilon_o\varepsilon_r\mu V^2}{8L^3}$$

where J is the current density, L is the film thickness of the active layer, μ is the charge carrier mobility, ε_r is the relative dielectric constant of the transport medium, and ε_0 is the permittivity of free space. $V = V_{app} - V_{bi}$, where V_{app} is the applied voltage, V_{bi} is the offset voltage. The carrier mobility can be calculated from the slope of the $J^{1/2} \sim V$ curves.

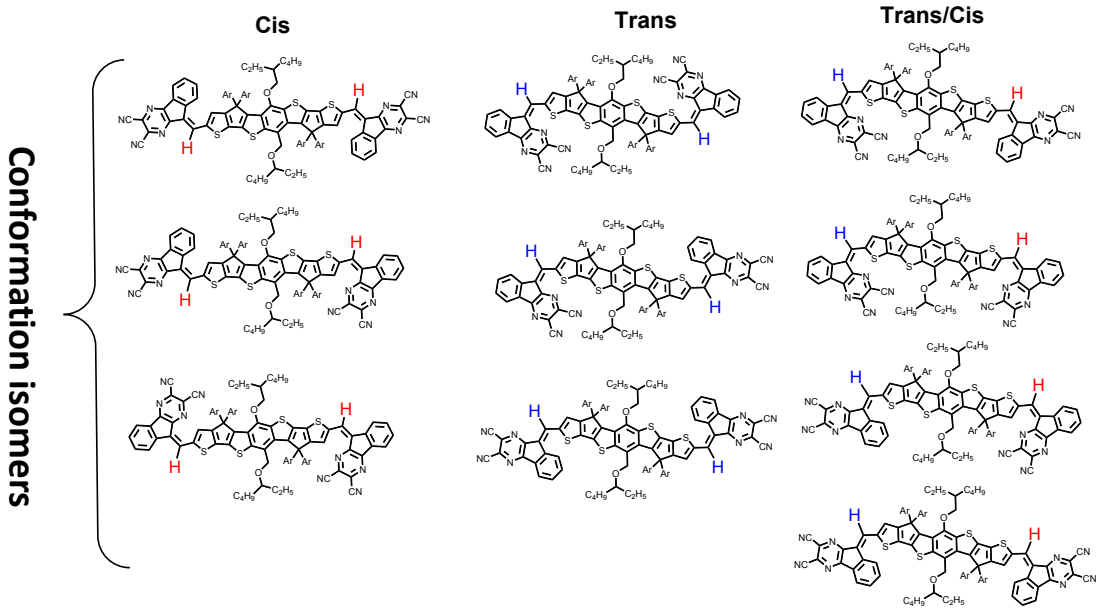

Fig. S1. The possible Cis-/Trans- isomers and conformation isomers of BTOIPC (the red is the Cis hydrogen and the blue is the Trans hydrogen).

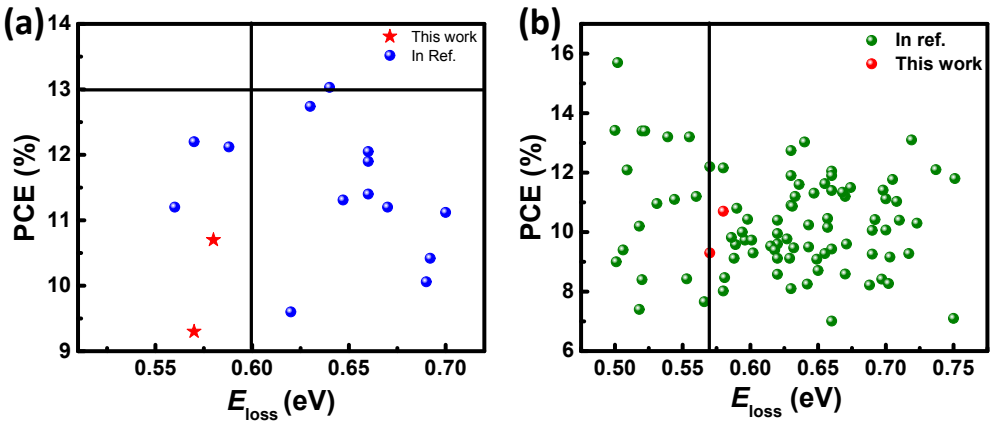

Fig. S2. The scatterplot of PCE *versus* E_{loss}: a) with PBDB-T as donor; b) with different donor.

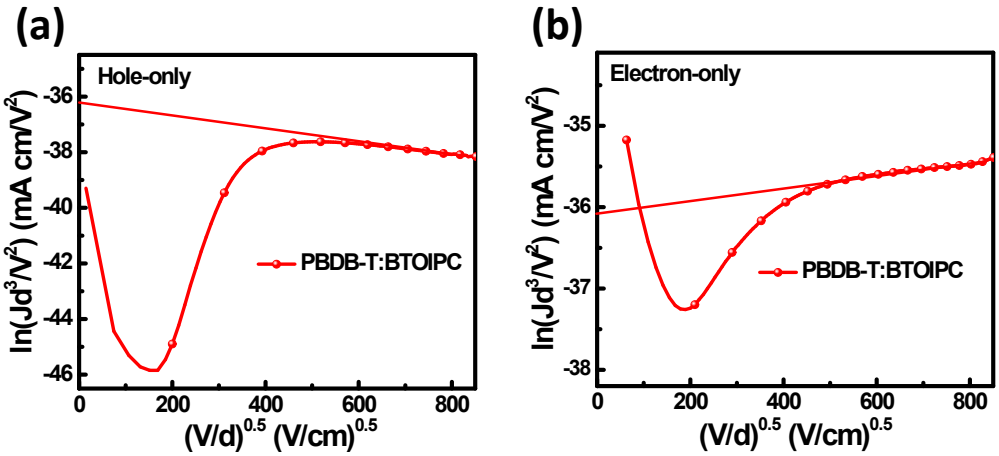

Fig. S3. a) The curve for hole-only device. b) The curve for electron-only device.

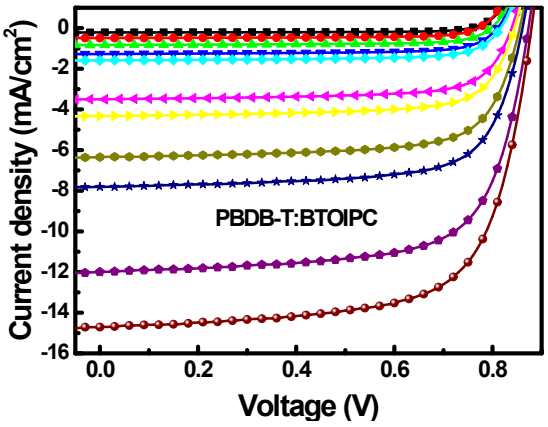

Fig. S4. The *J-V* curves of PBDB-T:BTOIPC-based OSCs under different light intensity.

Table S1 The detailed photovoltaic parameters of the 10 samples for OSCs based on PBDB-T:BTOIPC.

PBDB-T:BTOIPC	V_{oc} (V)	J_{sc} (mA cm^{-2})	FF (%)	PCE (%)
1	0.89	14.42	69.95	8.98
2	0.89	14.46	68.43	8.81
3	0.88	14.08	70.37	8.72
4	0.88	13.93	69.82	8.56
5	0.89	15.08	69.02	9.27
6	0.89	15.16	68.79	9.28
7	0.88	15.24	69.45	9.31
8	0.88	15.22	69.28	9.28
9	0.89	14.42	69.95	8.98
10	0.89	14.46	68.43	8.81

Table S2 The summarized data of E_{loss} and PCE of reported OSCs.

Acceptors	Donor	V_{OC} (V)	J_{SC} (mA cm^{-2})	E_{loss} [a)] (eV)	PCE (%)	Refs.
BTOIC	PBDB-T	0.862	18.60	0.531	10.96	1
IHIC	PTB7-Th	0.754	19.01	0.627	9.77	2
F6IC	PTB7-Th	0.611	18.07	0.750	7.1	3
F8IC	PTB7-Th	0.640	25.12	0.630	10.9	3
F10IC	PTB7-Th	0.732	20.83	0.518	10.2	3
4TIC	PTB7-Th	0.78	18.8	0.598	10.43	4
SN6IC-4F	PBDB-T	0.78	23.2	0.539	13.2	4
IEICO	PBDTTT-E-T	0.82	17.7	0.52	8.4	5
ATT-2	PTB7-Th	0.73	20.75	0.589	9.58	6
BT-CIC	PCE-10	0.70	22.5	0.633	11.2	7
DTPC-IC	PTB7-Th	0.76	21.92	0.454	10.21	8
CO*i*8DFIC	PTB7-Th	0.68	26.12	0.580	12.16	9
R12-4Cl	PBDB-T	0.75	18.5	0.602	9.3	10
Y6	PM6	0.83	25.3	0.502	15.7	11
Y1	PBDB-T	0.87	22.44	0.500	13.42	12
Y2	PBDB-T	0.82	23.56	0.520	13.40	12
ACS8	PTB7-Th	0.75	25.3	0.555	13.2	13
T1	PTB7-Th	0.72	20.95	0.586	9.82	14
T2	PTB7-Th	0.65	24.85	0.631	10.87	14
T3	PTB7-Th	0.61	22.00	0.660	9.43	14
T4	PTB7-Th	0.61	18.57	0.660	7.01	14
IEICO-4F	J52	0.734	21.9	0.506	9.4	15

IPIC-4F	PBDB-T	0.835	19.8	0.544	11.1	16
IPIC-4Cl	PBDB-T	0.813	22.2	0.522	13.4	16
CPDT-4F	PBDB-T	0.68	20.1	0.632	9.47	17
CPDT-4Cl	PBDB-T	0.65	21.3	0.655	9.28	17
SiOTIC-4F	PTB7-Th	0.65	21.6	0.501	9.0	18
COTIC-4F	PTB7-Th	0.56	20.3	0.518	7.4	18
CO5DFIC-OT	PTB7-Th	0.71	17.58	0.566	7.66	19
CO5DFIC-ST	PTB7-Th	0.74	20.71	0.596	9.73	19
CO*i*6IC	FTAZ	0.82	17.45	0.553	8.43	20
CO*i*6FIC	FTAZ	0.75	19.38	0.588	9.12	20
CO*i*6DFIC	FTAZ	0.67	20.98	0.642	8.25	20
T6Me	PM6	0.87	21.33	0.509	12.09	21
BDTIT-M	PBDB-T	1.53	0.942	0.588	12.12.	22
BDTThIT-M	PBDB-T	1.55	0.903	0.647	11.31	22
NITI	PBDB-T	1.49	0.86	0.63	12.74	23
IT-M	PBDBT	1.60	0.94	0.66	12.05	24
IT-OM-2	PBDB-T	1.59	0.93	0.66	11.9	25
ITCC	PBDB-T	1.67	1.01	0.66	11.4	26
ITIC	PBDB-T	1.57	0.90	0.67	11.2	27
FTIC-C6C8	PBDB-T	1.63	0.93	0.70	11.12	28
NFBDT	PBDB-T	1.56	0.868	0.692	10.42	29
FDICTF	PBDB-T	1.63	0.94	0.69	10.06	30
IDT-BOC6	PBDB-T	1.63	1.01	0.62	9.6	31
ZITI	PBDB-T	1.53	0.89	0.64	13.03	32
IXIC-2Cl	PBDB-T	1.30	0.73	0.57	12.2	33
IXIC-4Cl	PBDB-T	1.25	0.69	0.56	11.2	33
IT-4F	PBDB-T-SF	0.88	0.64	0.719	13.10	34
ITIC-Th1	FTAZ	0.849	0.701	0.737	12.10	35
IT-M	PB3T	1.00	0.61	0.630	11.9	36
ITCPTC	PBT1-EH	0.95	0.63	0.751	11.8	37
m-ITIC	J61	0.912	0.668	0.705	11.77	38
m-ITIC	J91	0.984	0.61	0.655	11.63	39
O-IDTBR	PvBDTTAZ	1.08	0.55	0.636	11.6	40
INIC3	FTAZ	0.852	0.628	0.674	11.5	41
ITIC	J71	0.94	0.65	0.698	11.41	42
ITIC	PBQ-4F:	0.95	0.62	0.668	11.34	43
IDIC	PTFBDT-BZS:	0.905	0.715	0.708	11.03	44
ITIC	PBPD-Th	1.01	0.54	0.590	10.8	45
BT-IC	J71	0.90	0.53	0.657	10.46	46
ITTC	HFQx	0.88	0.73	0.710	10.4	47
ITIC	PFBZ	0.89	0.68	0.620	10.4	48
ITIC	PTZ6	1.01	0.58	0.723	10.3	49
ITIC	PTzBI	0.87	0.70	0.643	10.24	50
ITIC	PDCBT	0.94	0.63	0.657	10.16	51

ATT-1	PTB7-Th	0.87	0.67	0.700	10.07	52
IEICO-4F	PBDTTT-EFT	0.739	0.501	0.594	10.0	53
IDTBR	PffBT4T-2DT	1.07	0.56	0.620	9.95	54
ITIC	3MT-Th	0.95	0.62	0.601	9.73	55
ITIC-Th	PDBT-T1	0.88	0.72	0.671	9.6	56
ITIC	J61	0.89	0.68	0.615	9.53	57
SF-PDI2	P3TEA	1.11	0.61	0.643	9.5	58
ITIC-Th	PffBT4T-B	0.972	0.628	0.618	9.4	59
TPH-Se	PDBT-T1	1.0	0.78	0.717	9.28	60
ITIC	J51	0.82	0.75	0.690	9.26	61
N2200	PTzBI	0.844	0.596	0.703	9.16	62
ITTIC	PBDB-T1	0.92	0.54	0.620	9.12	63
ITIC	PffQx-PS	0.97	0.60	0.629	9.12	64
ITIC	PBDTS-DTBTO	0.843	0.727	0.649	9.09	65
IDIC	PDBT-T1	0.89	0.73	0.650	8.71	66
NDP-V	PTB7-Th	0.74	0.87	0.670	8.59	67
IDSe-T-IC	J51	0.91	0.61	0.620	8.58	68
TPB	PTB7-Th	0.79	0.82	0.581	8.47	69
SdiPBI-Se	PDBT-T1	0.96	0.81	0.697	8.42	70
N2200	J51	0.83	0.65	0.702	8.27	71
SdiPBI-S	PBDTS-Se	0.91	0.86	0.688	8.22	72
DBFI-EDOT	PSEHTT	0.93	0.84	0.630	8.10	73
IDTIDSe-T-IC	J51	0.91	0.61	0.580	8.02	74
BTOIPC	**PBDB-T**	**1.45**	**0.88**	**0.57**	**9.3**	**This work**
BT-IC	**PBDB-T**	**1.43**	**0.85**	**0.58**	**10.7**	**This work**

[a)]Estimated from absorption edge according to $E_g^{opt} = 1240/\lambda_{onset}$.

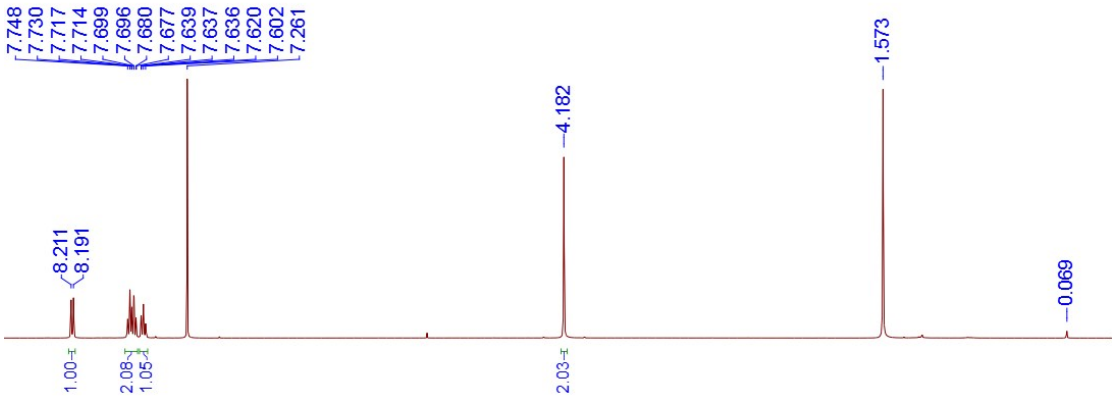

Fig. S5. The ^{1}H NMR spectrum of IPC.

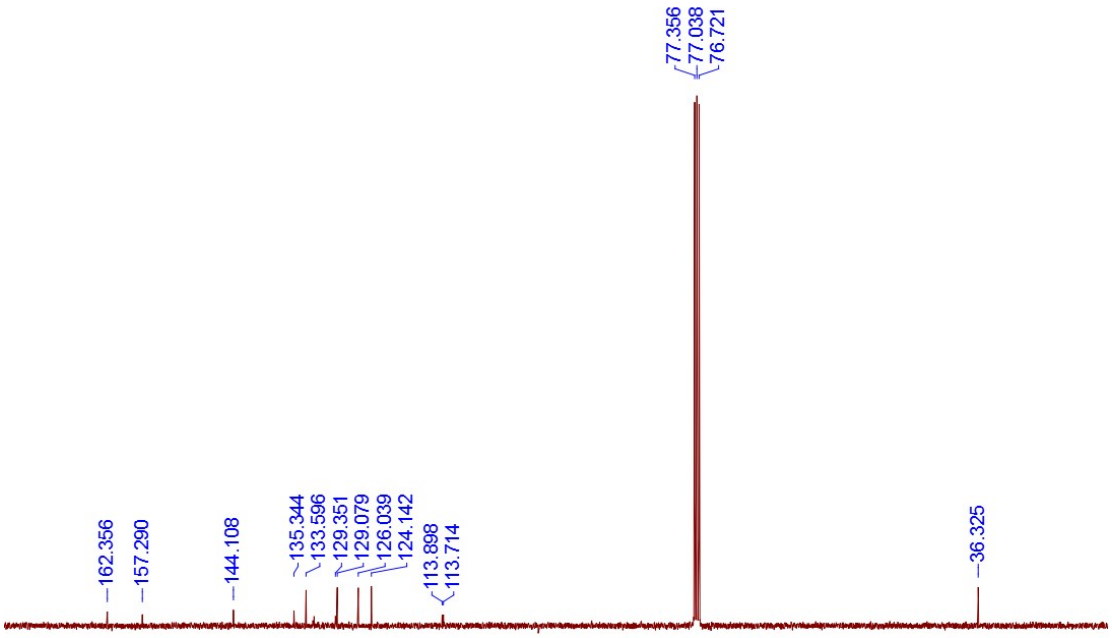

Fig. S6. The ^{13}C NMR spectrum of IPC.

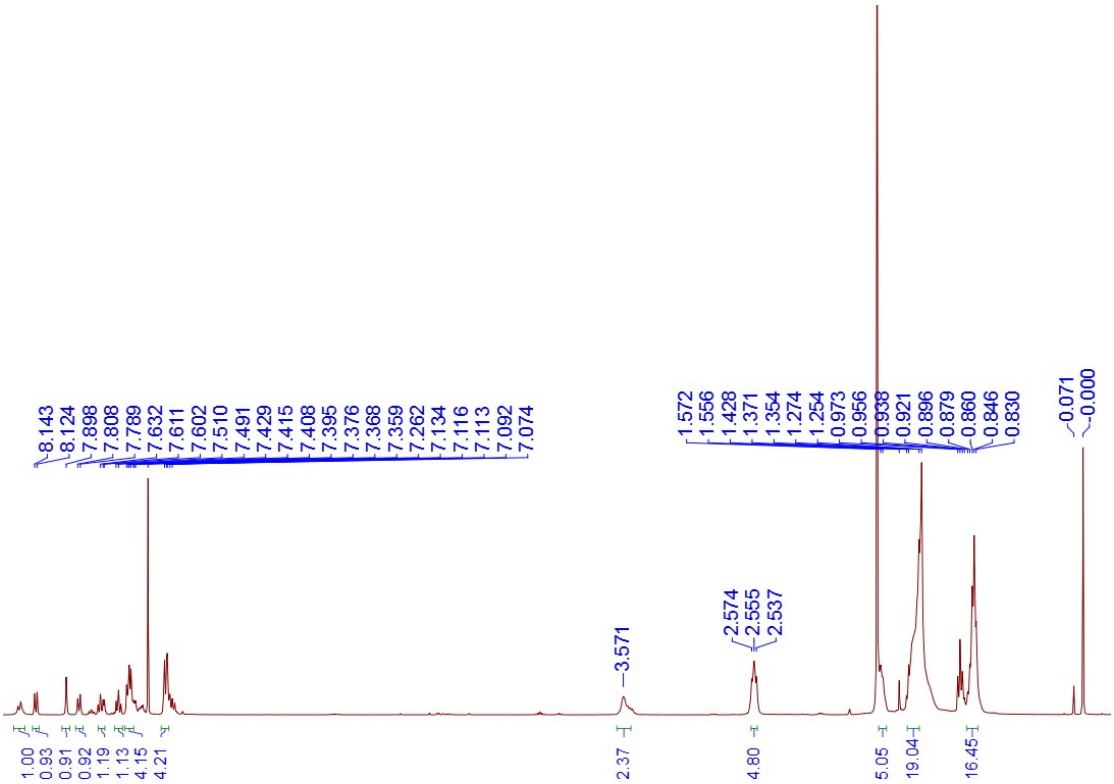

Fig. S7. The ^{1}H NMR spectrum of BTOIPC.

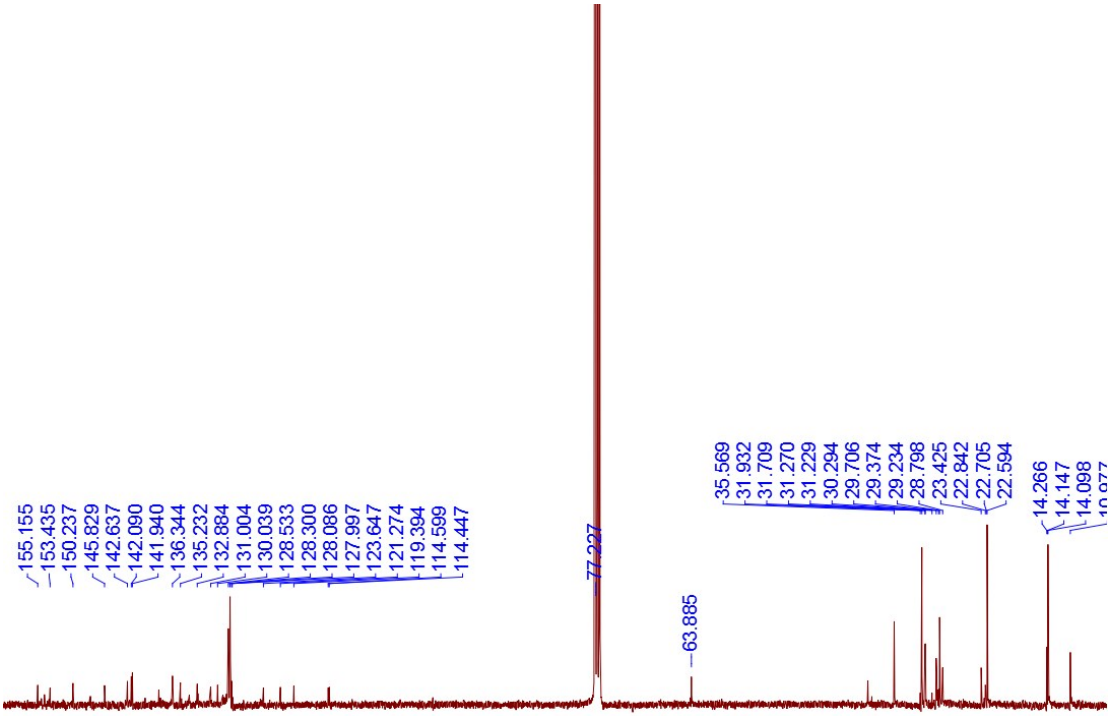

Fig. S8. The ^{13}C NMR spectrum of BTOIPC.

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