

## Supporting Information

### **A novel empirical method for quickly estimating the charge-transfer state of fullerene-donor derivatives**

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**Table S1.** The DFT calculated HOMO energy level. HOMO energy level of 152 molecules ( $\epsilon_{\text{HOMO, DFT}}$ ) which contain different functional groups. <sup>a</sup>

	$N_{\text{ECE}}$	$\epsilon_{\text{HOMO, ECEN}}$	$\epsilon_{\text{HOMO, DFT}}$	$\Delta\epsilon$
<i>Ethylene</i>	3.5480	-7.4751	-7.2526	-0.2225
<i>Propene</i>	4.8460	-6.8182	-6.7943	-0.0239
<i>Benzene</i>	6.0000	-6.4432	-6.6988	0.2556
<i>2-butylene</i>	6.1440	-6.4048	-6.3915	-0.0134
<i>2-methyl-propene</i>	6.1440	-6.4048	-6.5155	0.1106
<i>Toluene</i>	6.8146	-6.2445	-6.4018	0.1573
<i>Butadiene</i>	7.0960	-6.1849	-6.2291	0.0442
<i>Paraxylene</i>	7.6292	-6.0822	-6.1390	0.0568
<i>m-xylene</i>	7.6292	-6.0822	-6.2405	0.1583
<i>Phenol</i>	7.8580	-6.0418	-5.9546	-0.0871
<i>1,3-pentadiene</i>	8.3940	-5.9543	-5.9206	-0.0337
<i>2-methyl-butadiene</i>	8.3940	-5.9543	-6.1505	0.1961
<i>Mesitylene</i>	8.4438	-5.9467	-6.1817	0.2351
<i>Anisole</i>	8.8249	-5.8906	-5.8513	-0.0393
<i>Styrene</i>	9.5480	-5.7946	-6.0292	0.2345
<i>2-Phenylthiophene</i>	9.5800	-5.7906	-5.7384	-0.0523
<i>2,4-hexadiene</i>	9.6920	-5.7769	-5.6454	-0.1316
<i>1,1-dimethyl-butadiene</i>	9.6920	-5.7769	-5.6853	-0.0916
<i>Resorcinol</i>	9.7160	-5.7740	-5.7740	0.0000

<i>Napthalene</i>	10.0000	-5.7406	-5.7852	0.0446
<i>1,3,5-hexatriene</i>	10.6440	-5.7046	-5.6911	-0.0135
<i>1-methylnapthalene</i>	10.8146	-5.6900	-5.6712	-0.0188
<i>3-methyl-2,4-hexadiene</i>	10.9900	-5.6752	-5.5842	-0.0910
<i>Phloroglucinol</i>	11.5740	-5.6280	-5.8398	0.2118
<i>1,4-dimethylnapthalene</i>	11.6292	-5.6237	-5.5591	-0.0646
<i>2-methylnapthalene</i>	11.6292	-5.6237	-5.6957	0.0720
<i>Dimethylresorcinol</i>	11.6498	-5.6221	-5.6198	-0.0023
<i>1-Naphthol</i>	11.8580	-5.6060	-5.4204	-0.1856
<i>2-Naphthol</i>	11.8580	-5.6060	-5.5143	-0.0918
<i>1,4,5-trimethylnapthalene</i>	12.4438	-5.5626	-5.4114	-0.1512
<i>1-Methoxynapthalene</i>	12.8249	-5.5356	-5.3761	-0.1595
<i>2-Methoxynapthalene</i>	12.8249	-5.5356	-5.4133	-0.1223
<i>1-phenyl-butadiene</i>	13.0960	-5.5170	-5.5882	0.0713
<i>1,4-divinyl-benzene</i>	13.0960	-5.5170	-5.7049	0.1880
<i>2-(2-Thienyl)-5-phenyl-thiophen</i>	13.1600	-5.5126	-5.2877	-0.2249
<i>1,4-di(2-thienyl)-benzene</i>	13.1600	-5.5126	-5.4272	-0.0854
<i>1,3-di(2-thienyl)-benzene</i>	13.1600	-5.5126	-5.6957	0.1831
<i>Anilin</i>	13.1900	-5.5106	-5.2183	-0.2923
<i>1,4,5,8-tetramethylnapthalene</i>	13.2584	-5.5060	-5.2771	-0.2289
<i>Hydroquinone</i>	13.2920	-5.5038	-5.4101	-0.0937
<i>1-vinylnapthalene</i>	13.5480	-5.4868	-5.6802	0.1933

<i>2-(2-thienyl)-naphthalene</i>	13.5800	-5.4848	-5.5393	0.0545
<i>1-(2-thienyl)-naphthalene</i>	13.5800	-5.4848	-5.6095	0.1247
<i>1,5-naphthalenediol</i>	13.7160	-5.4760	-5.1626	-0.3134
<i>2,6-naphthalenediol</i>	13.7160	-5.4760	-5.2006	-0.2753
<i>Anthracene</i>	14.0000	-5.4579	-5.2235	-0.2344
<i>Trimethylphloroglucinol</i>	14.4747	-5.4286	-5.6144	0.1857
<i>9-methylanthracene</i>	14.8146	-5.4083	-5.1280	-0.2803
<i>2-methylanthracene</i>	14.8146	-5.4083	-5.1568	-0.2515
<i>1-methylanthracene</i>	14.8146	-5.4083	-5.1732	-0.2351
<i>1,4-dimethoxybenzene</i>	15.2258	-5.3845	-5.2746	-0.1099
<i>1,4-dimethylanthracene</i>	15.6292	-5.3618	-5.1234	-0.2384
<i>2,6-dimethoxynaphthalene</i>	15.6498	-5.3607	-5.0467	-0.3140
<i>1,5-dimethoxynaphthalene</i>	15.6498	-5.3607	-5.0867	-0.2740
<i>9-anthranol</i>	15.8580	-5.3493	-4.9485	-0.4008
<i>1-anthranol</i>	15.8580	-5.3493	-5.0285	-0.3208
<i>2-anthranol</i>	15.8580	-5.3493	-5.0576	-0.2917
<i>Pyrene</i>	16.0000	-5.3416	-5.3239	-0.0177
<b><i>CMPD.1</i></b>	16.7400	-5.3028	-5.1724	-0.1304
<i>1,3,5-tri(2-thienyl)benzene</i>	16.7400	-5.3028	-5.7365	0.4337
<i>2-methoxylantracene</i>	16.8249	-5.2984	-4.9798	-0.3187
<i>1-methoxylantracene</i>	16.8249	-5.2984	-4.9996	-0.2988
<i>9-methoxylantracene</i>	16.8249	-5.2984	-5.1223	-0.1761

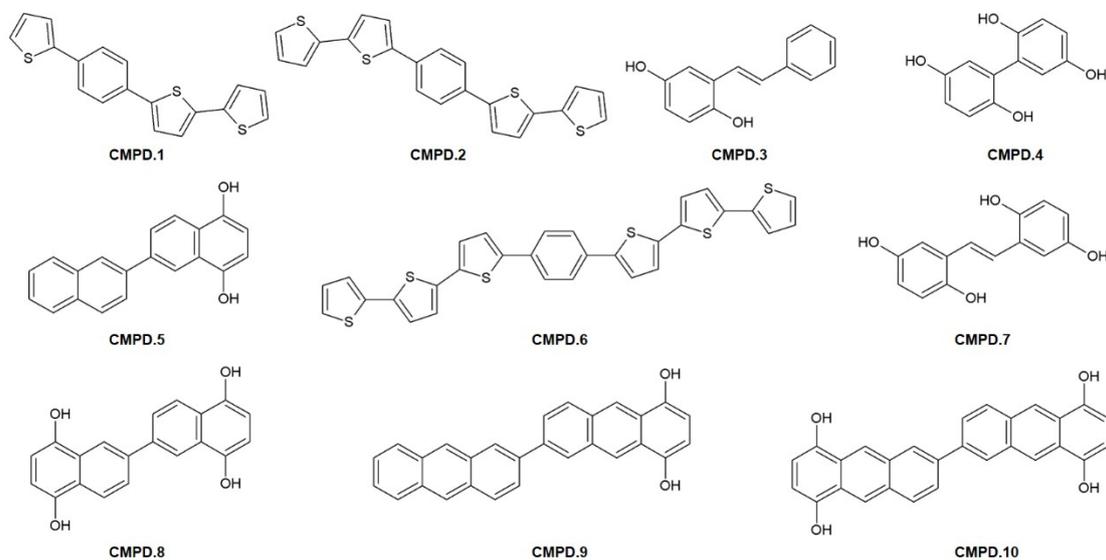
<i>1,4-divinyl-napthalene</i>	17.0960	-5.2848	-5.5455	0.2607
<i>2,6-di(2-thienyl)napthalene</i>	17.1600	-5.2816	-5.3603	0.0787
<i>1,4-di(3-thienyl)napthalene</i>	17.1600	-5.2816	-5.4506	0.1690
<i>1,5-di(4-thienyl)napthalene</i>	17.1600	-5.2816	-5.5635	0.2819
<i>1-naphthylamine</i>	17.1900	-5.2801	-5.1310	-0.1491
<i>2-naphthylamine</i>	17.1900	-5.2801	-5.1770	-0.1032
<i>1,4,5,8-tetramethylantracene</i>	17.2584	-5.2767	-5.0418	-0.2350
<i>1,4-Naphthalenediol</i>	17.2920	-5.2751	-5.0483	-0.2268
<i>1-vinylanthracene</i>	17.5480	-5.2626	-5.2112	-0.0514
<i>2-(2-thienyl)anthracene</i>	17.5800	-5.2611	-5.1707	-0.0903
<i>1-(2-thienyl)anthracene</i>	17.5800	-5.2611	-5.1876	-0.0735
<i>1,5-anthradiol</i>	17.7160	-5.2545	-4.8647	-0.3898
<i>1,8-anthradiol</i>	17.7160	-5.2545	-4.8742	-0.3803
<i>1,3,5,7-decatetraene</i>	17.7400	-5.2534	-5.1329	-0.1205
<i>Pyrenol</i>	17.8580	-5.2478	-5.3576	0.1098
<i>2-methoxypyrene</i>	18.8249	-5.2033	-5.2977	0.0944
<i>1,4,5,8,9,10-hexamethylantracene</i>	18.8876	-5.2005	-4.7527	-0.4479
<i>1,4-dimethoxynapthalene</i>	19.2258	-5.1857	-4.9926	-0.1931
<i>2-vinylpyrene</i>	19.5480	-5.1718	-5.3674	0.1956
<i>2-(2-thienyl)pyrene</i>	19.5800	-5.1704	-5.4014	0.2310
<i>1,5-dimethoxylantracene</i>	19.6498	-5.1674	-4.8109	-0.3566
<i>1,8-dimethoxylantracene</i>	19.6498	-5.1674	-4.8264	-0.3411

<i>Pyrendiol</i>	19.7160	-5.1646	-5.3785	0.2139
<i>Perylene</i>	20.0000	-5.1527	-4.9501	-0.2026
<b><i>CPD.2</i></b>	20.3200	-5.1396	-5.0565	-0.0831
<i>m-Phenylenediamine</i>	20.3800	-5.1371	-5.0309	-0.1062
<i>1,4-divinylanthracene</i>	21.0960	-5.1086	-5.1881	0.0795
<i>2,6-di(2-thienyl)anthracene</i>	21.1600	-5.1061	-5.1095	0.0034
<i>1,8-di(2-thienyl)anthracene</i>	21.1600	-5.1061	-5.1432	0.0372
<i>1,5-di(2-thienyl)anthracene</i>	21.1600	-5.1061	-5.1452	0.0391
<i>1,4-di(2-thienyl)anthracene</i>	21.1600	-5.1061	-5.1473	0.0412
<i>9-Anthracenamine</i>	21.1900	-5.1049	-4.7086	-0.3963
<i>2-Anthracenamine</i>	21.1900	-5.1049	-4.8440	-0.2609
<i>1-Anthracenamine</i>	21.1900	-5.1049	-4.8685	-0.2364
<i>1,4-Anthradiol</i>	21.2920	-5.1010	-4.7951	-0.3059
<i>Dimethoxypyrene</i>	21.6498	-5.0873	-5.2346	0.1474
<i>hydroxylperylene</i>	21.8580	-5.0794	-4.9382	-0.1413
<i>1,12-benzoperylene</i>	22.0000	-5.0741	-5.2009	0.1268
<i>Dimethoxylperylene</i>	22.8249	-5.0441	-4.9088	-0.1353
<b><i>CPD.3</i></b>	22.8400	-5.0436	-5.1922	0.1487
<i>Divinylpyrene</i>	23.0960	-5.0345	-5.4057	0.3712
<i>2,7-di(2-thienyl)pyrene</i>	23.1600	-5.0322	-5.4702	0.4379
<i>Aminopyrene</i>	23.1900	-5.0312	-5.2219	0.1907
<i>1,4-dimethoxylanthracene</i>	23.2258	-5.0299	-4.7614	-0.2686

<i>Vinylperylene</i>	23.5480	-5.0188	-4.9069	-0.1119
<i>1-N, N-dimethyl-naphthylamine</i>	23.6750	-5.0144	-5.2534	0.2390
<i>2,11-dihydroxylperylene</i>	23.7160	-5.0130	-4.9240	-0.0890
<i>2,8-dihydroxylperylene</i>	23.7160	-5.0130	-4.9292	-0.0838
<i>2,5-dihydroxylperylene</i>	23.7160	-5.0130	-4.9395	-0.0735
<i>Coronene</i>	24.0000	-5.0034	-5.4479	0.4445
<i>1,3,5,7-tetra(2-thienyl)naphthalene</i>	24.3200	-4.9927	-5.2667	0.2740
<i>1,3,5,7,9,11,13-tetradecheptene</i>	24.8360	-4.9759	-4.8440	-0.1318
<i>2,11-methoxylperylene</i>	25.6498	-4.9500	-4.8664	-0.0837
<i>2,8-methoxylperylene</i>	25.6498	-4.9500	-4.8710	-0.0791
<i>2,5-methoxylperylene</i>	25.6498	-4.9500	-4.8800	-0.0701
<b><i>CMPD.4</i></b>	26.5840	-4.9216	-5.1568	0.2353
<i>Divinylperylene</i>	27.0960	-4.9064	-4.8669	-0.0395
<i>Aminoperylene</i>	27.1900	-4.9037	-4.7867	-0.1170
<b><i>CMPD.5</i></b>	27.2920	-4.9007	-5.0483	0.1476
<i>Tetrahydroxylperylene</i>	27.4320	-4.8967	-4.9284	0.0317
<b><i>CMPD.6</i></b>	27.4800	-4.8953	-4.9069	0.0116
<i>1,3,5-trianimobenzene</i>	27.5700	-4.8927	-4.9292	0.0364
<i>1-(N, N-dimethyl)-aminoanthracene</i>	27.6750	-4.8897	-4.9678	0.0781
<i>9-(N, N-dimethyl)-aminoanthracene</i>	27.6750	-4.8897	-5.1364	0.2467
<i>1,4,5,8-tetra(2-thienyl)anthracene</i>	28.3200	-4.8716	-5.0369	0.1653
<i>1,8-diaminoanthracene</i>	28.3800	-4.8699	-4.6808	-0.1891

<i>1,3,5,7,9,11,13,15-hexadecoctene</i>	28.3840	-4.8698	-4.7461	-0.1237
<i>1,4,5,8-anthratetraol</i>	28.5840	-4.8643	-4.5253	-0.3391
<i>N-phenyl-N-methyl-aminobenzene</i>	29.2700	-4.8457	-5.0347	0.1890
<b>CMPD.7</b>	30.1320	-4.8231	-4.9681	0.1450
<i>Diaminopyrene</i>	30.3800	-4.8167	-4.9080	0.0912
<i>Tetramethoxylperylene</i>	31.2996	-4.7936	-4.8141	0.0205
<i>1,4,5,8-tetramethoxylantracene</i>	32.4516	-4.7658	-4.4736	-0.2922
<i>N, N-dimethyl-aminobenzene</i>	33.3500	-4.7448	-5.0138	0.2690
<i>Diaminoperylene</i>	34.3800	-4.7216	-4.6610	-0.0606
<b>CMPD.8</b>	34.5840	-4.7171	-4.8865	0.1694
<b>CMPD.9</b>	35.2920	-4.7017	-4.7951	0.0934
<i>1,3,5,7,9,11,13,15,17,19-eicosdecene</i>	35.4800	-4.6977	-4.6678	-0.0299
<i>2-(N, N-dimethyl)-aminonaphthalene</i>	37.3500	-4.6589	-5.0937	0.4348
<i>1,4-diaminobenzene</i>	39.2000	-4.6227	-4.6441	0.0214
<i>2-(N, N-dimethyl)-aminoanthracene</i>	41.3500	-4.5831	-4.8416	0.2585
<b>CMPD.10</b>	42.5840	-4.5614	-4.6860	0.1246
<i>1,4-diaminonaphthalene</i>	43.2000	-4.5508	-4.6232	0.0724
<i>(N, N-dimethyl)-aminopyrene</i>	43.3500	-4.5483	-5.0429	0.4946
<i>1,4-diaminoanthracene</i>	47.2000	-4.4863	-4.4997	0.0134
<i>(N, N-dimethyl)-aminoperylene</i>	47.3500	-4.4840	-4.7978	0.3138
<i>Tetraminoperylene</i>	48.7600	-4.4628	-4.4428	-0.0200
<i>1,4-di(N, N-dimethylamino)-benzene</i>	79.5200	-4.1243	-4.2696	0.1453

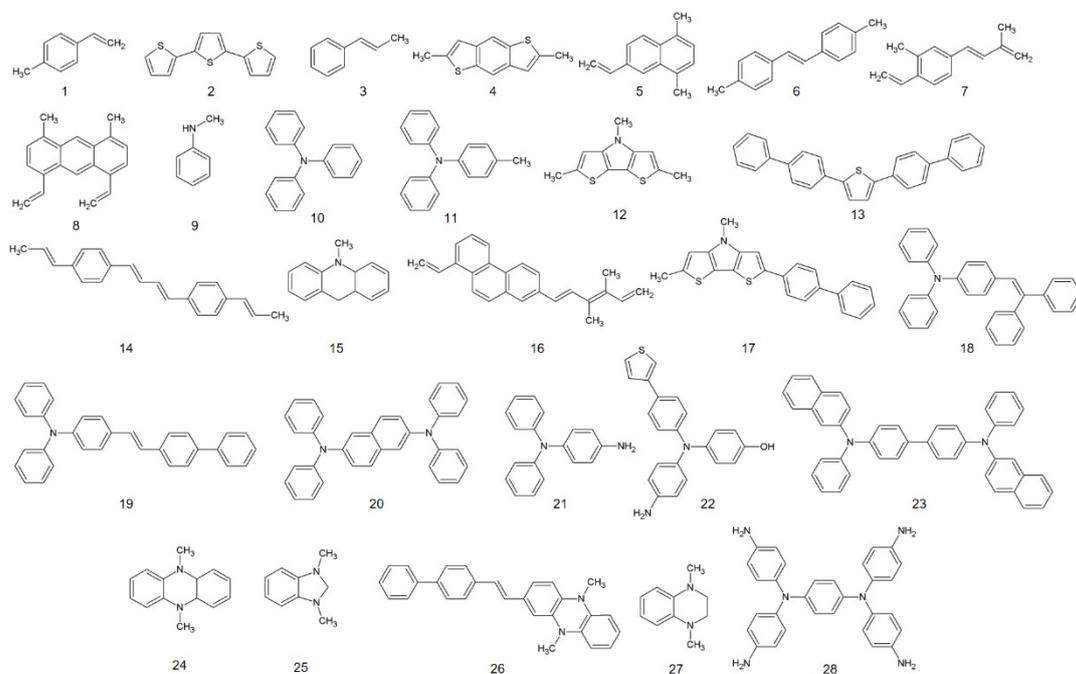
<sup>a</sup> All the values are used for the mathematic model building. Compounds are arranged by Equivalent Conjugated Electron Number ( $N_{ECE}$ ). The model predicted value ( $\epsilon_{HOMO, ECEN}$ ) and difference to the DFT calculated values ( $\Delta\epsilon$ ) are also listed in the table. Some of the compounds are with complicated structures and denoted as CMPD.X (X = 1-10) in the table, of which the structures are listed below the table.

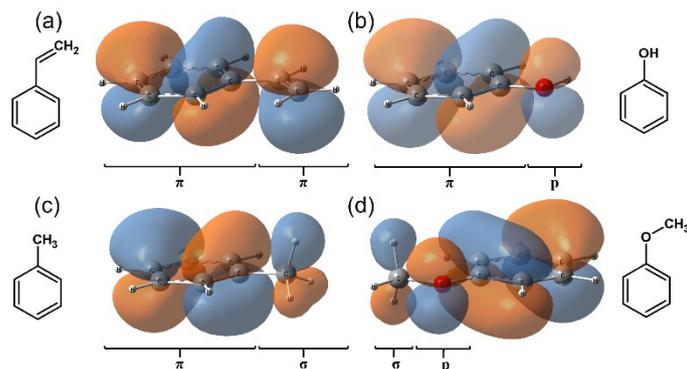


**Table S2.** Predicted HOMO of 28 molecules in the evaluation set and comparison with DFT calculated values. All energy items are in eV. The structures of the compounds are listed after the table.

<b>Compound</b>	<b>N<sub>ECE</sub></b>	<b><math>\epsilon_{\text{HOMO, ECEN}}</math></b>	<b><math>\epsilon_{\text{HOMO, DFT}}</math></b>	<b><math>\Delta\epsilon</math></b>
1	10.3626	-5.72297	-5.84256	0.119587
2	10.74	-5.69082	-5.60266	-0.08816
3	10.846	-5.68202	-5.78517	0.103144
4	13.304	-5.50213	-5.29883	-0.2033
5	13.4032	-5.4957	-5.508	0.012304
6	17.1772	-5.2851	-5.28142	-0.00367
7	20.0546	-5.15774	-5.35459	0.196854
8	22.7252	-5.05718	-5.06083	0.003656
9	23.27	-5.03834	-5.17126	0.132923
10	25.19	-4.97582	-4.94442	-0.0314
11	26.0046	-4.95094	-4.86962	-0.08132
12	27.026	-4.92099	-4.78448	-0.13651
13	27.58	-4.90529	-5.24987	0.344585
14	28.788	-4.87228	-4.90797	0.035688
15	30.0846	-4.83859	-5.01704	0.17845
16	32.086	-4.78976	-5.14461	0.354853
17	37.728	-4.6691	-4.77469	0.105585
18	37.738	-4.66891	-4.79373	0.12482
19	40.738	-4.613	-4.80325	0.190252
20	48.38	-4.48976	-4.64413	0.154368
21	51.2	-4.44988	-4.61938	0.169499
22	56.638	-4.37969	-4.49752	0.117828
23	58.38	-4.35885	-4.72518	0.366339
24	65.36	-4.282	-4.35064	0.068643
25	69.44	-4.24135	-4.12651	-0.11484

26	70.828	-4.22815	-4.34574	0.117591
27	79.52	-4.15177	-4.19533	0.043561
28	167.24	-3.69302	-3.87246	0.179442



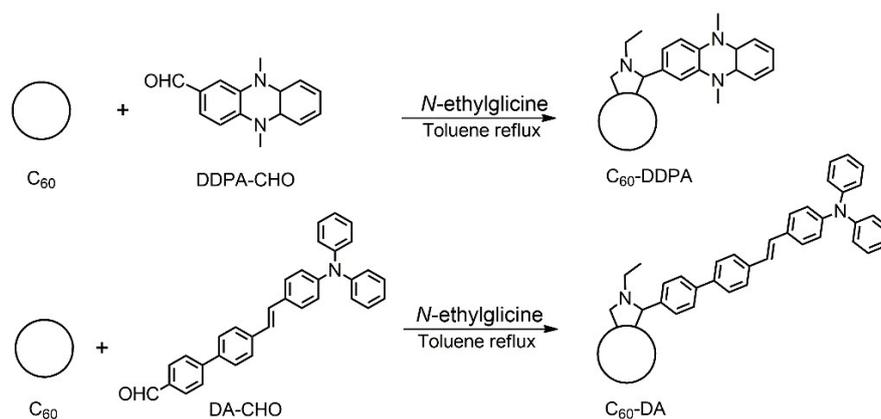


**Fig. S1** The HOMO of different conjugation types. (a)  $\pi$ - $\pi$  conjugation; (b)  $p$ - $\pi$  conjugation; (c)  $\sigma$ - $\pi$  conjugation and (d)  $\sigma$ - $p$  conjugation. The blue and orange colors indicate the positive and negative phase of the wavefunctions, respectively.

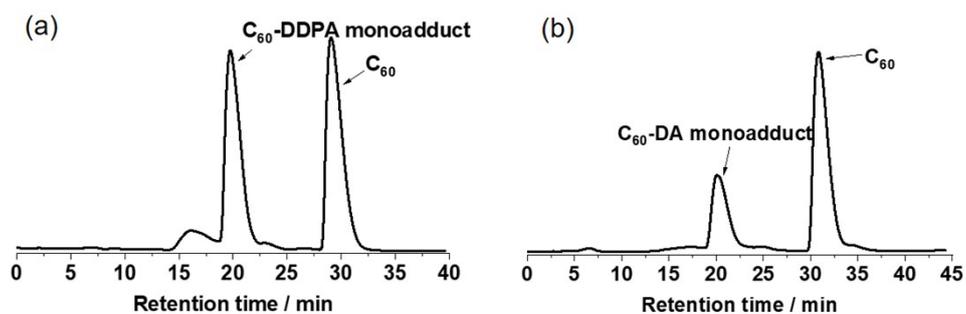
For fullerene modified with electron donating functional groups, the donors would always contribute a certain number of electrons through conjugation effect. As shown in Fig. S1, the four kinds of conjugation effects are considered: First, the  $\pi$ - $\pi$  conjugation, in which the interactions among the olefin chain, polyaromatic hydrocarbon ring and heterocyclic structures such as thiophenes are considered, e.g., styrene in Fig. S1a; Second, the  $p$ - $\pi$  conjugation, in which the atoms of O, N with lone pair electrons are considered, e.g. phenol in Fig. S1b; Third, the  $\sigma$ - $\pi$  conjugation, in which the methyl groups connected with the aromatic cores and the olefin chains are considered, e.g. toluene in Fig. S1c; Forth, the  $\sigma$ - $p$  conjugation, in which the methyl groups on the strong electron donor of N and O atoms are considered, e.g., anisole in Fig. S1d.

## 1. Synthesis, Purification and Characterization of C<sub>60</sub>-DDPA and C<sub>60</sub>-DA dyads

C<sub>60</sub> was heated with *N*-ethylglycine and aldehyde precursors which were synthesized as literature methods<sup>1,2</sup> at 120 °C to give corresponding fullerene derivatives with a yield of nearly 50% and 30% in toluene solution for 120 min. Pure C<sub>60</sub>-DDPA and C<sub>60</sub>-DA were isolated by HPLC using Buckyprep column (Fig. S2 and S3).

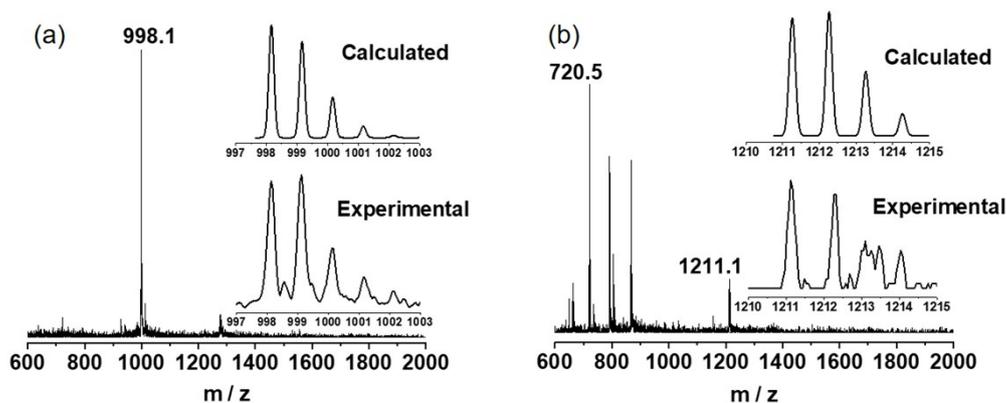


**Fig. S2** The synthesis of C<sub>60</sub>-DDPA and C<sub>60</sub>-DA derivatives.



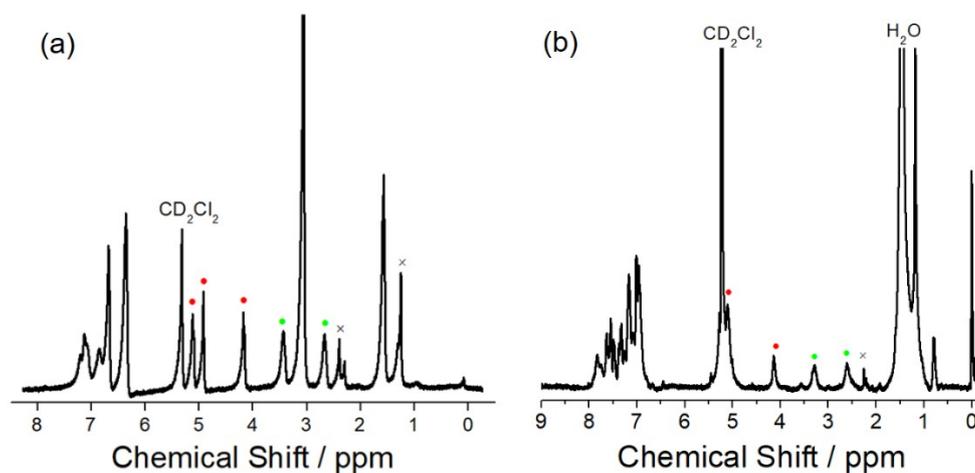
**Fig. S3** HPLC profile of the cycloaddition of (a) C<sub>60</sub>-DDPA and (b) C<sub>60</sub>-DA. HPLC conditions: 10×250 mm Buckyprep column, 6 mL/min flow rate with toluene, 330 nm detection.

The mass spectrometric analysis was performed on Shimadzu MALDI-TOF-MS Spectrometer using 1,9,10-trihydronaphthalene as the matrix (Fig. S4).



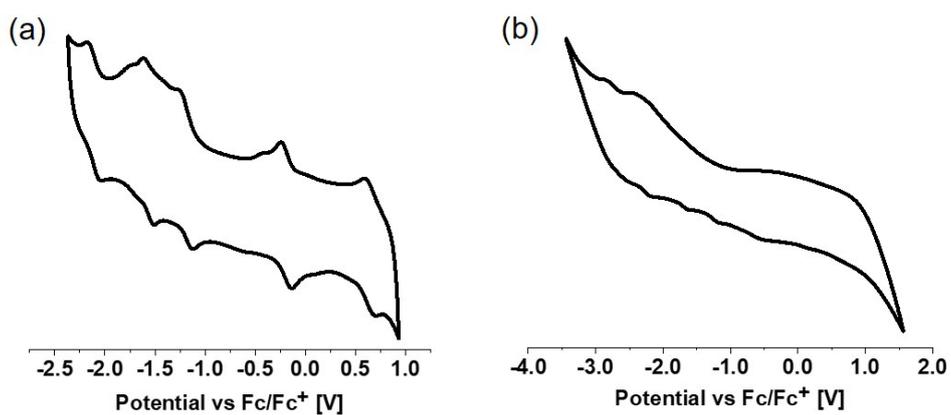
**Fig. S4** MALDI-TOF mass spectra of the pure (a) C<sub>60</sub>-DDPA (m/z = 998.1) and (b) C<sub>60</sub>-DA (m/z = 1211.1). The insets show the experimental and calculated isotope distributions.

<sup>1</sup>H NMR spectra of C<sub>60</sub>-DDPA and C<sub>60</sub>-DA were measured in dichloromethane-*d*<sub>2</sub> on a Bruker 400 MHz spectrometer (Fig. S5).



**Fig. S5** The <sup>1</sup>H NMR spectra of (a) C<sub>60</sub>-DDPA and (b) C<sub>60</sub>-DA between 0 and 9 ppm at 400 MHz in dichloromethane-*d*<sub>2</sub> (CD<sub>2</sub>Cl<sub>2</sub>) at 293 K. Signals marked by red and green circles are attributed respectively to the pyrrolidine geminal and methyne protons.

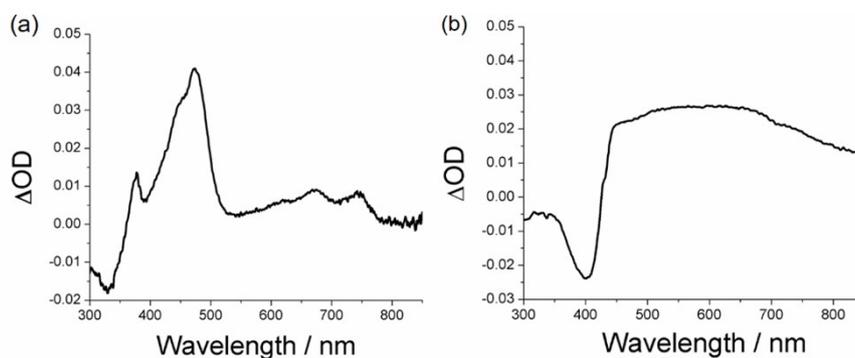
Electrochemical measurements were performed on a Princeton electrochemical workstation. The experiment was carried out in *o*-dichlorobenzene (*o*-DCB) in room temperature, using 0.05 M tetrabutylammonium hexafluorophosphate (TBAPF<sub>6</sub>) as supporting electrolyte and a scan rate of 50 mV/s, silver wire as the reference electrode, platinum wires as the counter electrode, glassy carbon electrode as the working electrode. The ferrocene/ferrocenium couple was used as the internal standard (Fig. S6).



**Fig. S6** Cyclic voltammogram of (a) C<sub>60</sub>-DDPA and (b) C<sub>60</sub>-DA measured in room temperature. Using 0.05 M TBAPF<sub>6</sub> in *o*-dichlorobenzene (*o*-DCB), silver wire as the reference electrode, platinum wires as the counter electrode, glassy carbon electrode as working electrode, ferrocene as internal standard, scan rate: 50 mV/s.

## 2. Spectroelectrochemistry Measurements.

Spectroelectrochemical measurement was performed in *o*-DCB solution with 0.05 M TBAPF<sub>6</sub> as supporting electrolyte employing a three-electrode setup. The working electrode was a platinum gauze with the counter electrode being a platinum wire and a silver wire as a reference electrode. The sample in a home-built quartz cuvette was put into the UV/Vis spectrometer in conjunction with the CHI 660D potentiostat to monitor absorption spectrum in situ (Fig. S7).



**Fig. S7** Differential absorption changes ( $\Delta OD$ ) in the UV-vis region obtained upon electrochemical oxidation of (a) DDPA at an applied bias of +0.7 V and (b) DA at an applied bias of +0.9 V in argon-saturated *o*-DCB with 0.05 M TBAPF<sub>6</sub> as supporting electrolyte at room temperature.

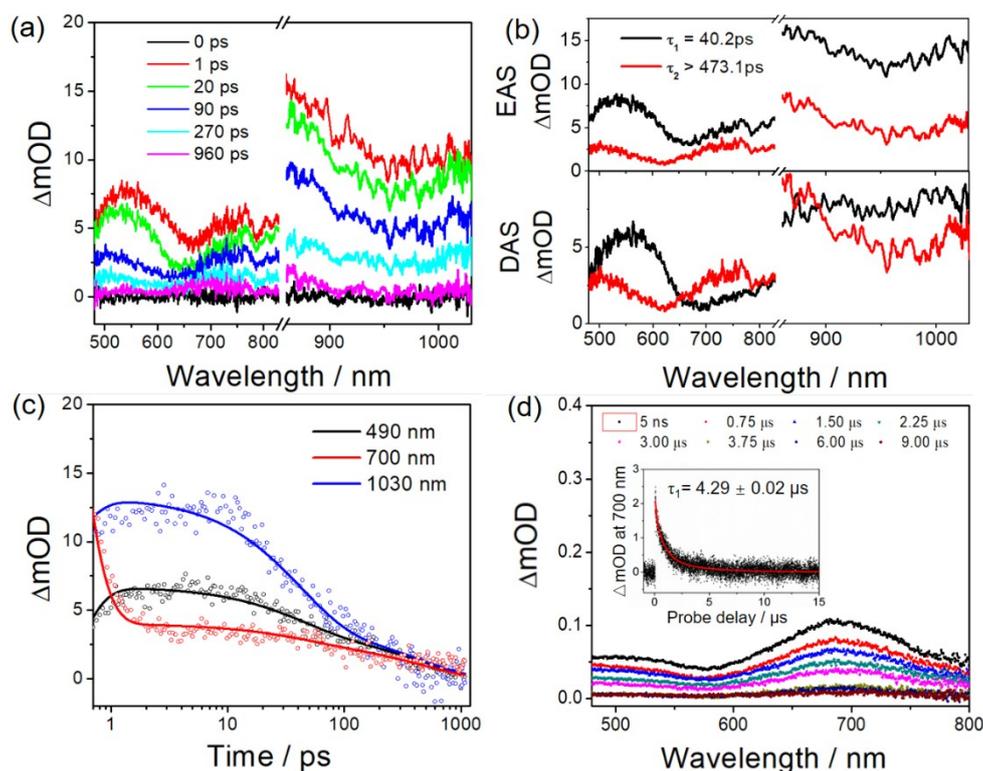
## 3. Transient Absorption Measurements of fullerene derivatives.

Femtosecond TA measurements were carried out on a homemade spectroscopy setup. The laser source is a commercial femtosecond amplifier laser system (35 fs, 1 kHz, 800 nm, Spitfire Ace, Spectra Physics). The output pulse was split into two beams. The first beam was used to pump a commercial wavelength conversion system (TOPAS prime and wavelength mixing unit from Spectra Physics) which will output tunable femtosecond laser pulses from 350 nm to 2600 nm. The pulses centered at 400 nm were selected as the excitation pulse. The second beam with weaker energy was focused into a CaF<sub>2</sub> plate (4 mm thickness) or an yttrium aluminum garnet plate to generate a white light continuum for the visible or near-infrared probe, respectively. The time delay between the pump beam and the probe pulses was controlled by a motorized

delay stage. In the transient absorption experiment, a laser frequency synchronized fiber optical spectrometer (AvaSpec-ULS2048CL-EVO, Avantes) was used to collect the probe light. During the experiment, the pump intensity was set to be 1 mJ/ cm<sup>2</sup> for the 400 nm laser pulses.

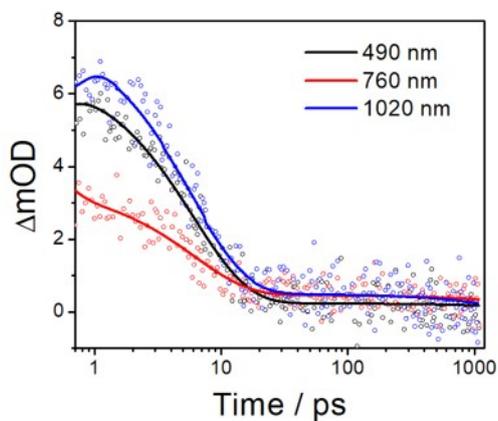
Transient absorption measurements on the nanosecond time scale were performed with by Continuum Surelite Laser pumping 400 nm pump light of ~5 ns pulse duration at 10 Hz. The pulse laser was coupled to a transient absorption detection system (Edinburgh Instruments), equipped with a monochromator and a pulsed Xenon lamp. The sample was contained in a 1x1cm optical glass sample cuvette. Transient absorption was collected at a right angle to the incoming laser beam by to a CCD camera to record transient absorption spectra. The output was processed with Edinburgh Instruments L900 software.

All samples were freshly prepared and deaerated for 20 min with argon prior to transient absorption investigation.



**Fig. S8** (a) Femtosecond transient absorption spectra of C<sub>60</sub>-DA following the excitation at 400 nm in toluene. (b) EAS (top) and DAS (bottom) derived from the global analysis of C<sub>60</sub>-DA; (c) Time-absorption profile at 490 nm (black), 700 nm (red) and 1030 nm (blue). (d) Differential absorption spectra obtained upon nanosecond flash photolysis of C<sub>60</sub>-DA (10<sup>-4</sup> M) in toluene

with several time delays between 5 and 9000 ns at room temperature, with time-decay profile of optical density at 700 nm (inset).



**Fig. S9** Time decay profiles of optical density at 490 nm (black), 760 nm (red), 1020 nm (blue) of C<sub>60</sub>-DDPA.

#### REFERENCE

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2. M. Klikar, P. le Poul, A. Ruzicka, O. Pytela, A. Barsella, K. D. Dorkenoo, F. R.-I. Guen, F. Bures and S. Achelle, *J. Org. Chem.*, 2017, **82**, 9435-9451.