SUPPORTING INFORMATION

Ruthenium (II) Complexes incorporating Carbazole-Diazafluorene based bipolar ligands for Dye sensitized solar cell applications

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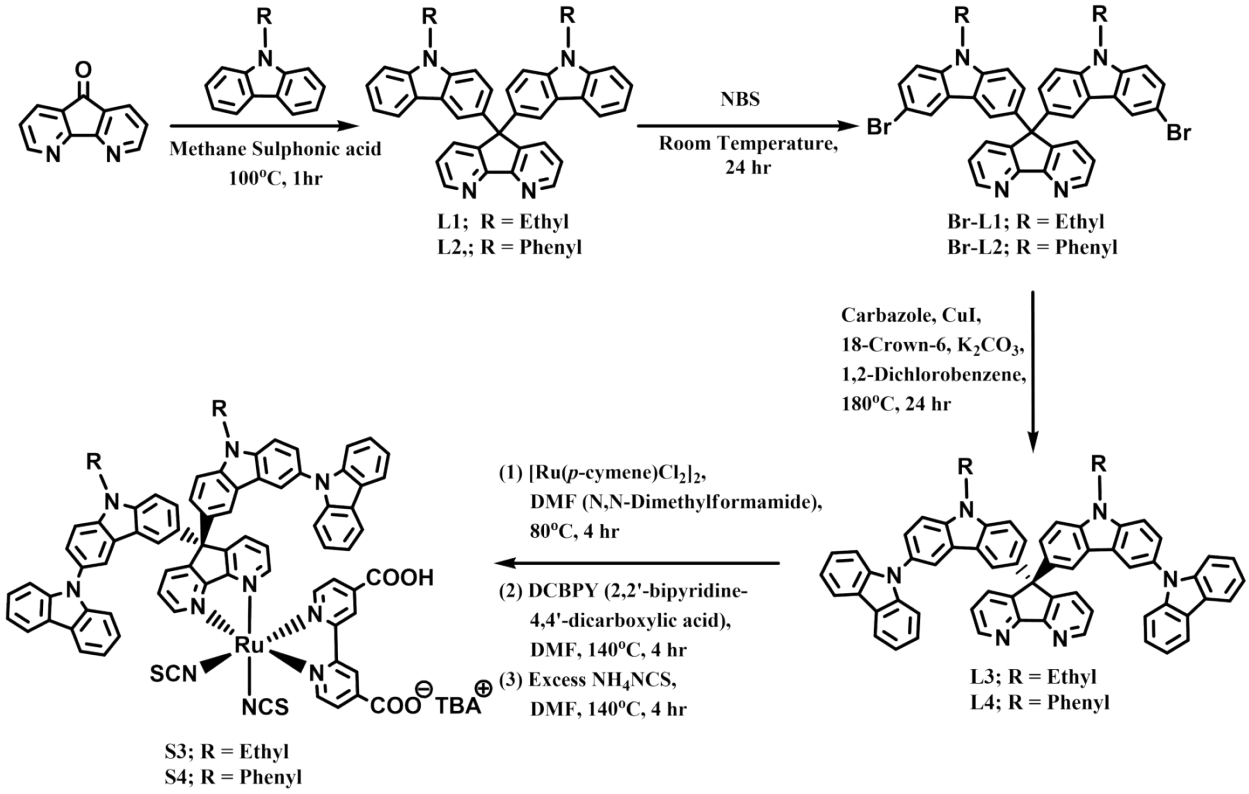
Captions	Table of Contents	Page
1	Synthesis of intermediates compounds	**2**
Scheme S1.	Synthetic routes for the preparation of ligands **L1-L4** and Ruthenium sensitizers **S3** and **S4**.	**2**
Fig. S1.	Frontier molecular orbitals **HOMO-3** and **LUMO +3** of sensitizers of **S3** and **S4** as determined from DFT calculations in DMF as solvent.	**6**
DFT (DATA)	XYZ coordinates of sensitizer **S3** optimized at B3LYP/6-31G(d) level using G09 package.	**7**
DFT (DATA)	XYZ coordinates of sensitizer **S4** optimized at B3LYP/6-31G(d) level using G09 package.	**11**

Scheme S1. Synthetic routes for the preparation of ligands **L1-L4** and Ruthenium sensitizers **S3** and **S4**.



1. Synthesis of intermediates compounds:

1.1. Synthesis of 9,9-bis(6-bromo-9-ethylcarbazol-3-yl)-4,5-diazafluorene (Br-L1) and 9,9-bis(6-bromo-9-phenylcarbazol-3-yl)-4,5-diazafluorene (Br-L2)

Br-L1: N-bromo-succinimide (NBS) (0.37 g, 2.1mmol) was added slowly in a solution of 9,9-bis(9-ethylcarbazol-3-yl)-4,5-diazafluorene (0.56 g, 1mmol) in DCM (20 mL). The mixture was stirred for 24 hours at room temperature. After pouring into brine solution and washing, it was extracted with DCM. The organic extracts were dried with $MgSO_4$ and concentrated by rotary evaporation. The pure product was obtained through column chromatography on silica gel (dichloromethane/acetone; 9/1) as a white solid, yield 0.70 g, 97%.

^{1}H NMR ($CDCl_3$): δ = 8.80 (d, 2H), 8.03 (d, 2H), 7.94-7.92 (d, 2H), 7.90 (d, 2H), 7.51-7.53 (d, 2H), 7.4-7.38 (d, 2H), 7.36-7.30 (m, 4H), 7.28-7.25 (d, 2H), 4.29-4.34 (q, 4H), 1.45-1.41 (t, 6H). ^{13}C NMR ($CDCl_3$): δ = 157.63, 150.75, 146.54, 141.95, 139.84, 139.50, 128.96, 126.73, 125.83, 125.60, 123.03, 122.45, 120.24, 119.53, 109.78, 109.57, 109.12, 60.61, 37.95, 14.04. Elemental analysis: Anal. Calcd (%) for $C_{39}H_{28}Br_2N_4$: C, 65.75; H, 3.96; N, 7.86; Found C, 65.25; H, 4.22; N, 8.25.

Br-L2: This ligand was prepared by following the same procedure used for **Br-L1** except that compound **L2** (0.65 g, 1 mmol) was used as starting material. The pure product was obtained through column chromatography on silica gel (dichloromethane/acetone; 9/1) as a white solid, yield 0.77 g, 95%. ^{1}H NMR ($CDCl_3$): δ = 8.81 (d, 2H), 8.1 (d, 2H), 7.96 (d, 2H), 7.94 (d, 2H), 7.62-7.58 (m, 4H), 7.52-7.44 (m, 8H), 7.38-7.31 (m, 6H), 7.29-7.24 (t, 2H). ^{13}C NMR ($CDCl_3$): δ =157.53, 150.25, 146.76, 140.42, 139.94, 136.99, 136.17, 133.92, 130.07, 128.94, 127.94, 126.88, 124.74, 123.68, 123.14, 122.33, 119.46, 112.86, 111.41, 110.36, 61.71. Elemental analysis: Anal. Calcd (%) for $C_{47}H_{28}Br_2N_4$: C, 69.82; H, 3.49; N, 6.93 Found C, 69.40; H, 3.65; N, 6.76.

1.2. Synthesis of 9,9-bis(6-carbazol-9-yl-9-ethylcarbazol-3-yl)-4,5-diazafluorene (L3) and 9,9-bis(6-carbazol-9-yl-9-phenylcarbazol-3-yl)-4,5-diazafluorene (L4)

L3: A mixture of **Br-L1** (0.43 g, 0.6mmol), carbazole (0.40 g, 2.4 mmol), copper (I) iodide (0.11 g, 0.6 mmol), potassium carbonate (0.40 g, 2.88 mmol) and 18-crown-6 (0.016 g, 0.06 mmol) were dissolved in 1,2-dichlorobenzene (5 mL) under nitrogen atmosphere. The mixture was then stirred at 180 ºC for 24 h. After completion of the reaction, the solvent was removed by reduced pressure distillation and the remains were extracted with DCM. The combined extracts were dried over anhydrous $MgSO_4$, the solvent was removed by rotary

evaporation and the pure product was obtained through column chromatography on silica gel (dichloromethane/acetone; 9/1) as a white solid. Yield 0.44 g, 82%. ^{1}H NMR ($CDCl_3$): δ = 8.81 (d, 2H), 8.19-8.17 (d, 4H), 8.05 (d, 2H), 7.90 (d, 2H), 7.58 (d, 4H), 7.51-7.36 (m, 8H), 7.34-7.26 (m, 10H), 7.15 (t, 2H), 4.46-4.40 (q, 4H), 1.54-1.50 (t, 6H). ^{13}C NMR ($CDCl_3$): δ = 157.76, 150.78, 147.56, 141.95, 139.84, 139.50, 128.96, 126.73, 125.83, 125.60, 123.64, 123.03, 122.45, 120.24, 119.84, 119.53, 109.78, 109.57, 109.12, 61.54, 37.95, 14.04. Elemental analysis: Anal. Calcd (%) for $C_{63}H_{44}N_6$: C, 85.49; H, 5.01; N, 9.50; Found C, 84.34; H, 5.51; N, 9.25.

L4: This ligand was prepared by following the same procedure used for **L3** except that compound **Br-L2** (0.49 g, 0.6mmol) was used as starting material. The pure product was obtained through column chromatography on silica gel (dichloromethane/acetone; 9/1) as a white solid, yield 0.50 g, 85%. ^{1}H NMR ($CDCl_3$): δ = 8.80 (d, 2H),8.17-8.15 (d, 4H), 8.07 (d, 2H), 7.91 (d, 2H), 7.65-7.59 (m, 8H), 7.55-7.52 (m, 4H), 7.4-7.37 (m, 8H), 7.31-7.26 (m, 12H), 7.10 (t, 2H). ^{13}C NMR ($CDCl_3$): δ = 157.78, 150.45, 147.07, 141.8, 140.83, 140.44, 137.12, 130.07, 129.97, 127.98, 127.04, 126.93, 125.90, 125.85, 124.05, 123.04, 122.86, 120.26, 119.66, 119.60, 119.46, 111.01, 110.53, 109.72, 61.95. Elemental analysis: Anal. Calcd (%) for $C_{71}H_{44}N_6$: C, 86.91; H, 4.52; N, 8.57; Found C, 85.19; H, 4.61; N, 9.43.

1.3. Synthesis of Complexes

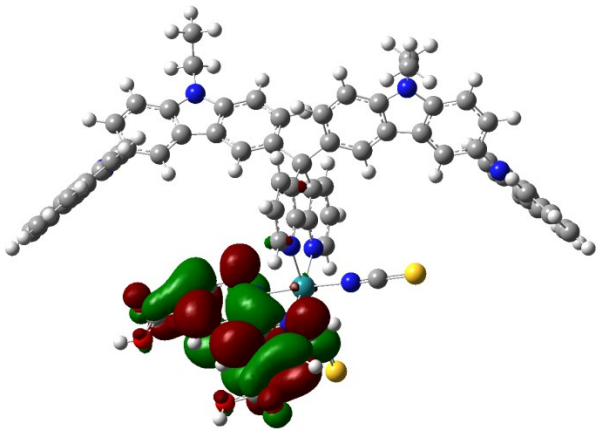
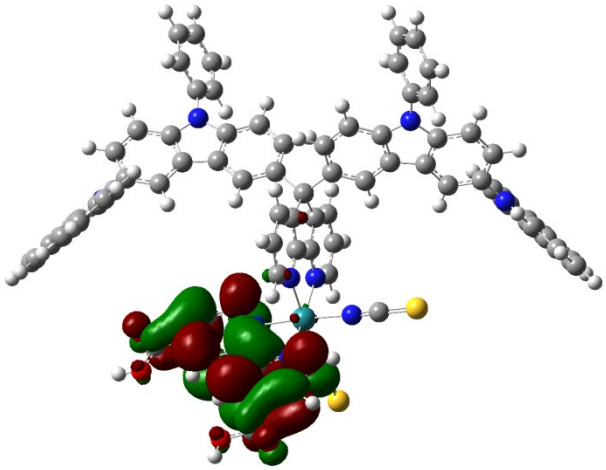
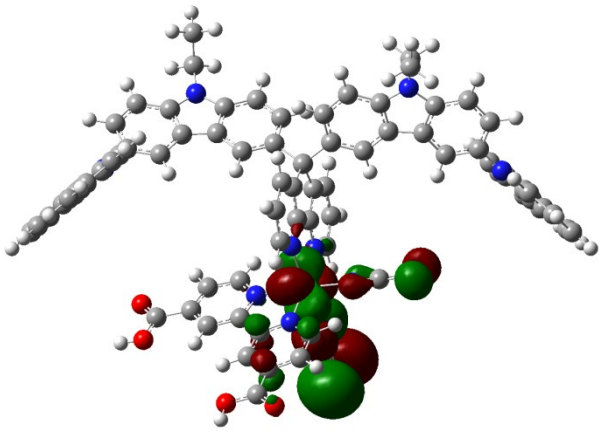
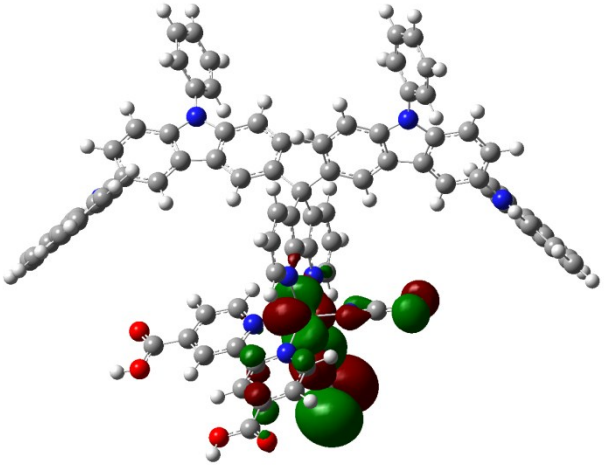
Synthesis of Tetrabutylammonium [ruthenium (4-carboxylic acid-4'-carboxylate-2,2'-bipyridine)(9,9-bis(6-carbazol-9-yl-9-ethylcarbazol-3-yl)-4,5-diazafluorene)(NCS)$_2$]

(S3): L3 (0.35 g, 0.4mmol) and [Ru(p-cymene) Cl_2]$_2$ (0.12 g, 0.2mmol) were dissolved in dry DMF (30 mL). The solution was heated to 80 ^{o}C under N_2 atmosphere for 4 h in the dark. Then it was added to 4,4'-dicarboxylic acid-2,2'-bipyridine (0.10 g, 0.4mmol) and the reaction mixture

was refluxed at 160 °C for 4 h. To the resulting solution, ammonium thiocyanate (0.30 g, 4.0mmol) was added and further heated at 130 °C for 4 h. The volume of the solvent was reduced on a rotary evaporator to about ~5 mL, and the purple residue was suspended in water to induce the precipitation. The resulting solid was filtered off, washed with water and diethyl ether and then dried under vacuum. The crude product was dissolved in basic methanol (with TBAOH) and further purified on the Sephadex LH-20 with methanol as an eluent. The main band was collected, concentrated and precipitated with 0.02 N HNO_3 to obtain the title complex **S1,** yield 0.17 g, 26 %. ^{1}H NMR (DMSO-d_6): δ = 8.86 (d, 2H), 8.64 (d, 2H), 8.52-8.47 (d, 2H),8.31 (d, 2H), 8.22-8.18 (d, 4H), 8.09 (d, 2H), 7.83 (s, 2H), 7.60 - 7.54 (m, 4H), 7.42-7.33 (m, 12H), 7.25-7.18 (m, 8H), 4.49 (m, 4H), 3.16 (t, 8H), 1.56 (m, 8H), 1.39-1.28 (m, 14H), 0.94-0.91 (t, 12H).Elemental analysis: Anal. Calcd(%) for $C_{97}H_{99}N_{11}O_4RuS_2$: C, 70.69; H, 6.05; N, 9.35; S; 3.89; Found C, 70.87; H, 6.08; N, 9.37; S, 3.80.

Synthesis of Tetrabutylammonium [ruthenium (4-carboxylic acid-4'-carboxylate-2,2'-bipyridine)(9,9-bis(6-carbazol-9-yl-9-phenylcarbazol-3-yl)-4,5-diazafluorene)(NCS)$_2$] (S4): Using the same conditions as for synthesizing complex **S3** and, starting from the ligand **L4** (0.39 g, 0.4 mmol), compound **S4** was obtained. Yield 0.16 g, 23 %. 1H NMR (DMSO-d6): δ = 8.85 (d, 2H), 8.65 (d, 2H), 8.52-8.45 (d, 2H), 8.32-8.28 (d, 2H), 8.24 (d, 4H), 8.13 (d, 2H), 7.90 (s, 2H), 7.70-7.67 (m, 4H), 7.54 (m, 8H), 7.49-7.35 (m, 20H), 7.26-7.21 (t, 2H), 3.17 (t, 8H), 1.57 (m, 8H), 1.30 (m, 8H), 0.93 (t, 12H). Elemental analysis: Anal. Calcd (%) for C105H99N11O4RuS2: C, 72.30; H, 5.72; N, 8.83; S, 3.68; Found C, 72.05; H, 5.99; N, 8.22; S, 3.82.

Fig. S1. Frontier molecular orbitals **HOMO-3** and **LUMO +3** of sensitizers of **S3** and **S4** as determined from DFT calculations in DMF as solvent.

 **LUMO +3 (1.86 eV)**	 **LUMO +3 (1.86 eV)**
 **HOMO -3 (5.61 eV)**	 **HOMO -3 (5.61 eV)**
S3	**S4**

DATA: XYZ coordinates of sensitizer **S3** optimized at B3LYP/6-31G(d) level using G09 package.

Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms) X	Y	Z
1	44	0	-0.117400	2.996655	-0.423256
2	7	0	1.877713	3.079047	-0.997255
3	6	0	2.628239	4.024479	-0.362678
4	6	0	2.457544	2.304109	-1.933865
5	6	0	3.981809	4.191426	-0.660865
6	6	0	3.796886	2.421231	-2.276141
7	1	0	1.815666	1.577746	-2.415222
8	6	0	4.577982	3.381350	-1.627435
9	1	0	4.571939	4.941606	-0.152208
10	1	0	4.229839	1.780541	-3.034744
11	7	0	0.589557	4.514358	0.792082
12	6	0	1.905675	4.831485	0.636150
13	6	0	-0.155061	5.201449	1.679872
14	6	0	2.493276	5.852375	1.385695
15	6	0	0.370968	6.226404	2.453039
16	1	0	-1.193204	4.903164	1.751402
17	6	0	1.720216	6.560299	2.306259
18	1	0	3.538325	6.100788	1.259411
19	1	0	-0.253752	6.760884	3.158372
20	7	0	-0.707827	1.278703	-1.641820
21	6	0	-1.205251	1.071930	-2.876271
22	6	0	-0.569070	0.180600	-0.888836
23	6	0	-1.538637	-0.211853	-3.326786
24	1	0	-1.332844	1.947365	-3.502774
25	6	0	-0.870315	-1.134670	-1.224911
26	6	0	-1.373434	-1.341224	-2.507704
27	1	0	-1.932680	-0.323313	-4.331045
28	1	0	-1.634566	-2.327166	-2.878379
29	7	0	0.286548	1.415305	1.006841
30	6	0	-0.050891	0.248097	0.441911
31	6	0	0.745772	1.328781	2.270692
32	6	0	0.017699	-1.025120	0.997030
33	6	0	0.849947	0.099700	2.933429
34	1	0	1.028319	2.255377	2.757052
35	6	0	0.482369	-1.102859	2.307856
36	1	0	1.222281	0.090389	3.951969
37	1	0	0.563869	-2.044508	2.841092
38	6	0	-0.507291	-2.064017	-0.032888
39	7	0	-0.589652	4.342043	-1.915736
40	7	0	-2.083384	3.054749	0.280756

41	6	0	-3.194780	3.046442	0.672139
42	6	0	-0.856852	5.103589	-2.773715
43	16	0	-4.748784	3.042703	1.225795
44	16	0	-1.234196	6.165732	-3.978324
45	6	0	2.282119	7.665459	3.140769
46	6	0	6.020564	3.504821	-1.996824
47	8	0	1.631161	8.297959	3.949154
48	8	0	6.563709	2.811916	-2.834838
49	8	0	3.586924	7.883726	2.899799
50	1	0	3.876228	8.614878	3.479486
51	8	0	6.652327	4.466591	-1.300927
52	1	0	7.582459	4.484755	-1.599072
53	6	0	-1.728838	-2.781405	0.580623
54	6	0	-3.034282	-2.343334	0.349468
55	6	0	-1.511700	-3.855274	1.475851
56	6	0	-4.103796	-2.975095	0.995924
57	1	0	-3.230347	-1.517586	-0.326895
58	6	0	-2.555245	-4.497677	2.133575
59	1	0	-0.497948	-4.201308	1.651278
60	6	0	-3.859442	-4.054312	1.885423
61	1	0	-2.349422	-5.317277	2.814399
62	6	0	0.579127	-3.054466	-0.505747
63	6	0	0.187849	-4.184435	-1.262783
64	6	0	1.937463	-2.829823	-0.275674
65	6	0	1.109018	-5.085959	-1.784765
66	1	0	-0.868604	-4.367901	-1.432164
67	6	0	2.885997	-3.725375	-0.785199
68	1	0	2.268670	-1.970746	0.298473
69	6	0	2.467704	-4.850119	-1.543209
70	1	0	0.769800	-5.945774	-2.353419
71	6	0	4.329925	-3.790770	-0.718300
72	6	0	4.714786	-4.953587	-1.442403
73	6	0	5.307040	-2.976416	-0.136754
74	6	0	6.061221	-5.315539	-1.571656
75	6	0	6.648822	-3.340229	-0.259839
76	1	0	5.036787	-2.072933	0.401531
77	6	0	7.016562	-4.504085	-0.967631
78	1	0	6.364070	-6.206697	-2.111525
79	1	0	8.067104	-4.769208	-1.032483
80	6	0	-5.537734	-2.780280	0.973485
81	6	0	-6.090559	-3.752536	1.853275
82	6	0	-6.379989	-1.880869	0.311742
83	6	0	-7.471346	-3.833867	2.070550
84	6	0	-7.757601	-1.971574	0.516206
85	1	0	-5.979170	-1.117226	-0.347943
86	6	0	-8.293684	-2.944611	1.385740

87	1	0	-7.901587	-4.568269	2.743479
88	1	0	-9.369946	-2.996187	1.516725
89	7	0	-5.066242	-4.523517	2.385227
90	7	0	3.580462	-5.576384	-1.944455
91	7	0	-8.638179	-1.076292	-0.162875
92	7	0	7.663051	-2.534154	0.339377
93	6	0	-8.821085	-1.002657	-1.545540
94	6	0	-8.222073	-1.757539	-2.559626
95	6	0	-9.785167	0.000117	-1.833914
96	6	0	-8.595415	-1.487455	-3.875365
97	1	0	-7.491663	-2.526657	-2.330592
98	6	0	-10.141767	0.252015	-3.165796
99	6	0	-9.543528	-0.492436	-4.180106
100	1	0	-8.143572	-2.059691	-4.681104
101	1	0	-10.877338	1.016236	-3.403378
102	1	0	-9.810727	-0.305930	-5.216392
103	6	0	-9.471907	-0.134053	0.442812
104	6	0	-9.622512	0.172597	1.799453
105	6	0	-10.200926	0.555960	-0.562395
106	6	0	-10.526020	1.178177	2.140178
107	1	0	-9.055783	-0.353543	2.560760
108	6	0	-11.103238	1.562485	-0.192186
109	6	0	-11.262439	1.867144	1.157920
110	1	0	-10.661467	1.434546	3.187420
111	1	0	-11.668858	2.099877	-0.948994
112	1	0	-11.959245	2.644794	1.457516
113	6	0	8.685255	-1.869108	-0.341338
114	6	0	9.499114	-1.181708	0.598365
115	6	0	8.939579	-1.807040	-1.715685
116	6	0	10.592225	-0.430444	0.145513
117	6	0	10.032642	-1.052519	-2.139273
118	1	0	8.308313	-2.327776	-2.428295
119	6	0	10.854506	-0.371410	-1.221473
120	1	0	11.224855	0.101205	0.851650
121	1	0	10.251633	-0.989662	-3.201751
122	1	0	11.699474	0.206584	-1.584765
123	6	0	7.808325	-2.283956	1.705996
124	6	0	7.039580	-2.747903	2.778839
125	6	0	8.938704	-1.447460	1.907481
126	6	0	7.412743	-2.352951	4.062735
127	1	0	6.181867	-3.392893	2.618243
128	6	0	9.291484	-1.064613	3.208779
129	6	0	8.525105	-1.518024	4.280157
130	1	0	6.830691	-2.699718	4.912322
131	1	0	10.153416	-0.424697	3.379282
132	1	0	8.787476	-1.227312	5.293408

133	6	0	3.556294	-6.835121	-2.684727
134	1	0	2.701108	-6.802850	-3.366112
135	1	0	4.452855	-6.870232	-3.310989
136	6	0	3.479669	-8.069278	-1.780503
137	1	0	2.570882	-8.052185	-1.169986
138	1	0	3.465994	-8.978142	-2.391662
139	1	0	4.343849	-8.119321	-1.110097
140	6	0	-5.222650	-5.587847	3.372929
141	1	0	-4.437906	-6.328149	3.190275
142	1	0	-6.175146	-6.088268	3.174893
143	6	0	-5.165677	-5.086917	4.819380
144	1	0	-5.286177	-5.928922	5.509491
145	1	0	-4.205264	-4.605077	5.029963
146	1	0	-5.964142	-4.364165	5.016696

DATA: XYZ coordinates of sensitizer **S4** optimized at B3LYP/6-31G(d) level using G09 package.

Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms) X	Y	Z
1	44	0	-0.107741	3.487727	0.421163
2	7	0	-2.132787	3.490210	0.887062
3	6	0	-2.878899	4.428788	0.237278
4	6	0	-2.734797	2.677372	1.776358
5	6	0	-4.248761	4.554097	0.475701
6	6	0	-4.091460	2.751059	2.057113
7	1	0	-2.097040	1.956488	2.271178
8	6	0	-4.866945	3.706614	1.395197
9	1	0	-4.834340	5.301592	-0.042057
10	1	0	-4.541874	2.081252	2.779548
11	7	0	-0.802188	5.001600	-0.806016
12	6	0	-2.133844	5.274989	-0.711396
13	6	0	-0.037048	5.724470	-1.646660
14	6	0	-2.716127	6.287638	-1.476152
15	6	0	-0.556895	6.743313	-2.431998
16	1	0	1.012482	5.460417	-1.669635
17	6	0	-1.921733	7.032580	-2.348035
18	1	0	-3.773867	6.500236	-1.400360
19	1	0	0.084327	7.307074	-3.098694
20	7	0	0.479724	1.767439	1.638592
21	6	0	0.913453	1.552399	2.895503
22	6	0	0.426144	0.681385	0.857654
23	6	0	1.268053	0.272200	3.340163
24	1	0	0.972013	2.418250	3.545166
25	6	0	0.757199	-0.628613	1.185638
26	6	0	1.191702	-0.844861	2.491616
27	1	0	1.607582	0.153737	4.363324
28	1	0	1.466269	-1.828921	2.857834
29	7	0	-0.375577	1.921998	-1.057479
30	6	0	-0.022304	0.757005	-0.497825
31	6	0	-0.769004	1.844146	-2.343850
32	6	0	-0.016016	-0.506599	-1.078562
33	6	0	-0.797267	0.625166	-3.032179
34	1	0	-1.061008	2.769678	-2.826620
35	6	0	-0.416632	-0.575585	-2.410904
36	1	0	-1.121042	0.622032	-4.067204
37	1	0	-0.440728	-1.508619	-2.964446
38	6	0	0.500477	-1.544870	-0.043050
39	7	0	0.227047	4.819493	1.961711
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41	6	0	3.022299	3.685781	-0.496444
42	6	0	0.415579	5.572145	2.847950
43	16	0	4.602994	3.766512	-0.961758
44	16	0	0.681339	6.621531	4.092874
45	6	0	-2.477153	8.131229	-3.195407
46	6	0	-6.327407	3.785638	1.701088
47	8	0	-1.808364	8.793589	-3.964348
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51	8	0	-6.952014	4.746866	0.998073
52	1	0	-7.894765	4.734950	1.253829
53	6	0	1.784754	-2.189831	-0.606463
54	6	0	3.056371	-1.729045	-0.262615
55	6	0	1.663410	-3.221300	-1.567634
56	6	0	4.187078	-2.299149	-0.861523
57	1	0	3.180361	-0.932001	0.462912
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59	1	0	0.675830	-3.585478	-1.832961
60	6	0	4.037046	-3.337976	-1.813468
61	1	0	2.643670	-4.590064	-2.913056
62	6	0	-0.560274	-2.595187	0.351214
63	6	0	-0.150732	-3.746454	1.064285
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65	6	0	-1.056115	-4.694756	1.527822
66	1	0	0.906875	-3.907766	1.247510
67	6	0	-2.851373	-3.342389	0.552849
68	1	0	-2.263311	-1.528060	-0.446288
69	6	0	-2.414583	-4.482124	1.272683
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71	6	0	-4.290851	-3.447850	0.441510
72	6	0	-4.664142	-4.650545	1.098125
73	6	0	-5.274321	-2.628372	-0.122478
74	6	0	-6.003817	-5.039910	1.198607
75	6	0	-6.609937	-3.024126	-0.040538
76	1	0	-5.013447	-1.694199	-0.610417
77	6	0	-6.965710	-4.222550	0.613554
78	1	0	-6.291016	-5.954163	1.706219
79	1	0	-8.011909	-4.508420	0.654969
80	6	0	5.608074	-2.053133	-0.735797
81	6	0	6.258042	-2.952018	-1.622556
82	6	0	6.366491	-1.162409	0.031608
83	6	0	7.652176	-2.989958	-1.729615
84	6	0	7.757443	-1.198583	-0.072872
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87	1	0	8.149917	-3.687404	-2.394326
88	1	0	9.474359	-2.129501	-0.987664
89	7	0	5.296429	-3.729009	-2.270635
90	7	0	-3.518499	-5.271965	1.598168
91	7	0	8.553663	-0.310155	0.710032
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93	6	0	8.618332	-0.283643	2.105298
94	6	0	7.961141	-1.092172	3.038744
95	6	0	9.525808	0.732080	2.508513
96	6	0	8.216734	-0.861941	4.389797
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105	6	0	10.030324	1.345379	1.296932
106	6	0	10.561295	2.078005	-1.344211
107	1	0	9.175729	0.526879	-1.942247
108	6	0	10.930963	2.388140	1.040581
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115	6	0	-8.966531	-1.613036	1.435277
116	6	0	-10.610211	-0.186762	-0.396155
117	6	0	-10.085555	-0.903602	1.868996
118	1	0	-8.338593	-2.152091	2.137089
119	6	0	-10.903103	-0.198317	0.965818
120	1	0	-11.239557	0.363625	-1.090759
121	1	0	-10.328431	-0.895594	2.928092
122	1	0	-11.768751	0.343115	1.336660
123	6	0	-7.750876	-1.901035	-1.979217
124	6	0	-6.949120	-2.296420	-3.055272
125	6	0	-8.895888	-1.081493	-2.166099
126	6	0	-7.304292	-1.849442	-4.327166
127	1	0	-6.080317	-2.929242	-2.906458
128	6	0	-9.229802	-0.644885	-3.455364
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130	1	0	-6.696605	-2.142583	-5.178977
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133	6	0	5.555860	-4.743636	-3.239341
134	6	0	6.254557	-4.427652	-4.411161
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149	6	0	-3.966337	-8.866866	2.456509
150	1	0	-4.413044	-7.633651	0.739686
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