Electronic Supplementary Material (ESI) for New Journal of Chemistry.

Synthesis, characterization and electroluminescence studies of cyanopyridine-based π-conjugative polymers carrying benzo[c][1,2,5]thiadiazole and naphtho[1,2-c:5,6-c']bis([1,2,5]thiadiazole) units

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Contents

1. Synthesis

*Synthesis of 4,6-bis-(3'-formyl-biphenyl-4-yl)-2-heptyloxy-nicotinonitrile (**Cy_1**)*

Compound **3** (0.33 g, 0.767 mmol) was dissolved in 8 mL of DMF. 1-Bromoheptane (0.15 mL, 0.777 mmol) was then added, followed by 148 mg of potassium carbonate (1.072 mmol) and the reaction mixture was stirred at 90 ^{o}C for overnight. The reaction mixture was cooled and poured into ice cold water with constant stirring. The obtained precipitate was filtered, washed with water and dried to obtain creamy yellow dibromo derivative of O-alkylated cyanopyridine (monomer) **Cy_1**. The obtained crude product was dissolved in a minimum amount of $CHCl_3$, followed by re-precipitation using methanol to get the pure monomer. Finally, it was recrystallized from $CHCl_3$ and methanol mixture. Yield: 280 mg, 75%. FTIR (cm^{-1}): 1540 (C=O), 2219 (C≡N), 2918 (Aromatic CH stretching). ^{1}H NMR (300 MHz, $CDCl_3$, δ ppm) : 0.88 (t, J = 4.8 Hz, 3H), 1.33 (t, J = 6.4 Hz, 3H), 1.39 (t, J = 4Hz, 3H), 1.51 (d, J = 7.2 Hz, 2H), 1.89 (m, J = 6.8 Hz, 2H), 4.576 (t, J = 6.4 Hz, 2H), 7.26 (s, 1H) 7.52 (d, J = 6.8 Hz, 2H), 7.61 (d, J = 2 Hz, 1H), 7.63 (d, J = 2 Hz, 1H), 7.66 (d, J = 2 Hz, 1H), 7.68 (d, J = 2 Hz, 1H), 7.92 (d, J = 2 Hz, 1H), 7.94 (d, J = 2 Hz, 1H). ESI-MS, exact mass calculated for ($C_{25}H_{24}Br_2N_2O$), $[M+H]^+$: 528.28; Found : 529.4. Elemental Anal. Calcd (%) for $C_{25}H_{24}Br_2N_2O$: C, 56.84; H, 4.58; N, 5.30; Found: C, 56.82; H, 5.54; N, 5.33.

*Synthesis of 1,4-bis-(2'-formyl-phenylene-thiophene-5-yl)-2-hexadecoxyl-nicotinonitrile (**Cy_2**)*

Compound, 2,5-*bis*(4-bromothiophene)-2-hydroxypyridine-3-carbonitrile (**6**, 0.5 g, 1.131 mmol) was dissolved in 8 mL of DMF. To this 1-bromohexadecane (0.35 mL, 1.146 mmol) was then added, followed by 148 mg of potassium carbonate (1.072 mmol) and the reaction mixture was stirred at 80 ^{0}C for overnight. Progress of the reaction was monitored using TLC. It was then cooled and poured onto 150 mL of ice cold water with constant stirring. The obtained precipitate was filtered, washed with water and dried to obtain creamy yellow product. It was recrystallized using chloroform-methanol to give product, **Cy_2**. Yield 75%; mp 71.1 ^{0}C; ATR-IR (cm^{-1}): 2220 (C≡N), 2919 (C-H). ^{1}H NMR (300 MHz, $CDCl_3$, δ ppm): 0.81-1.86 (m, 31H), 4.56-4.58 (t, 2H), 7.73-7.86 (m, 5H). Elemental Anal. Calcd (%) for $C_{30}H_{38}Br_2N_2OS_2$: C, 54.06; H, 5.75; N, 4.20; Found: C, 54.05; H, 5.76; N, 4.21.

2. Characterization (FTIR, NMR and GPC)

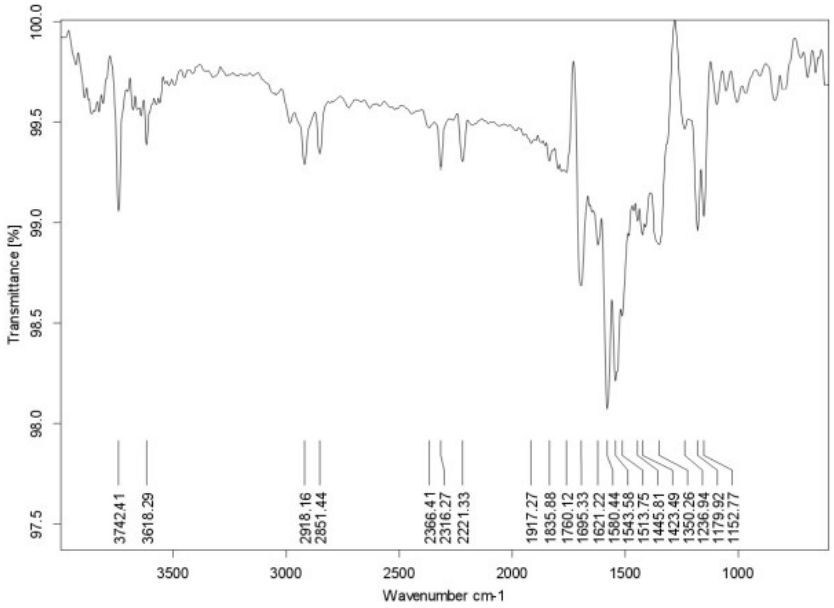

Fig S1: FTIR spectrum of monomer ***Cy$_1$***

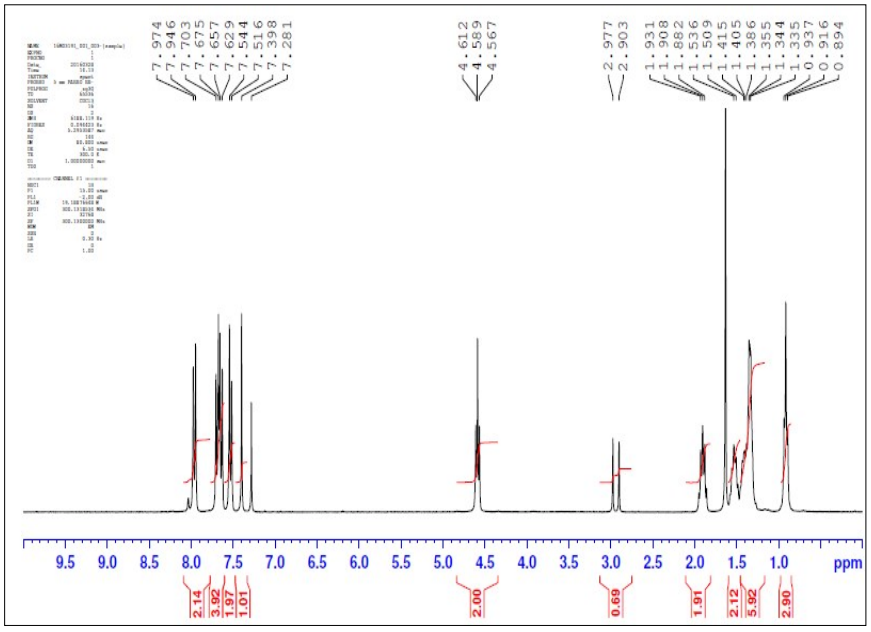

Fig S2: ^{1}H NMR spectrum of monomer ***Cy$_1$***

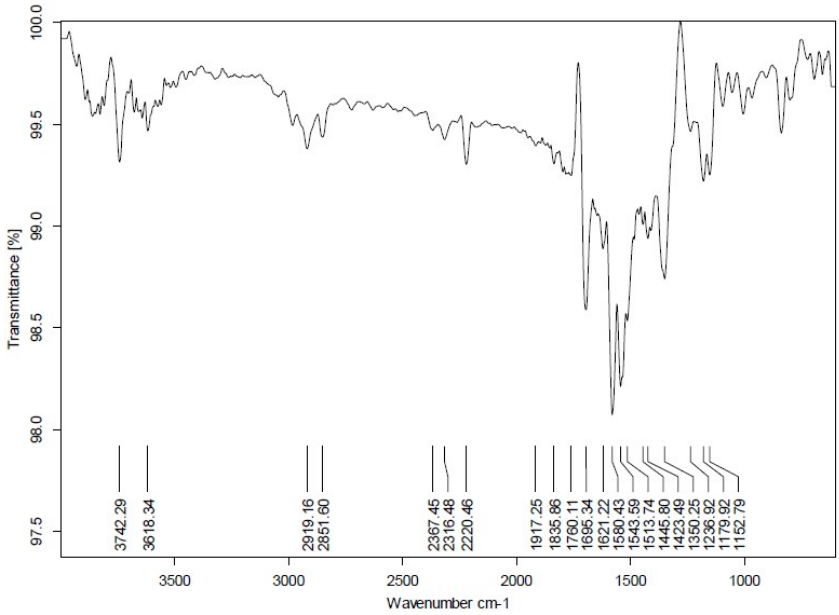

Fig S3: FTIR spectrum of intermediate **Cy_2**

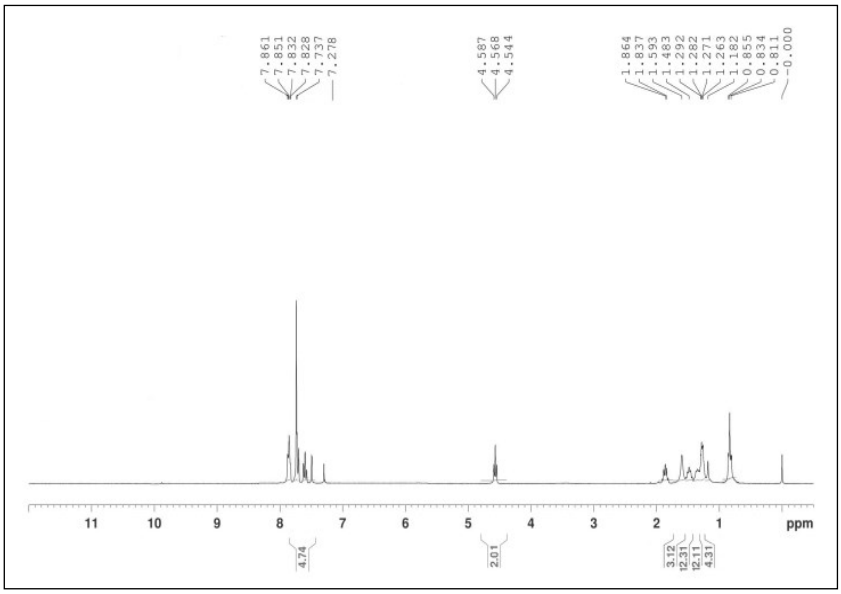

Fig S4: ^{1}H NMR spectrum of intermediate **Cy_2**

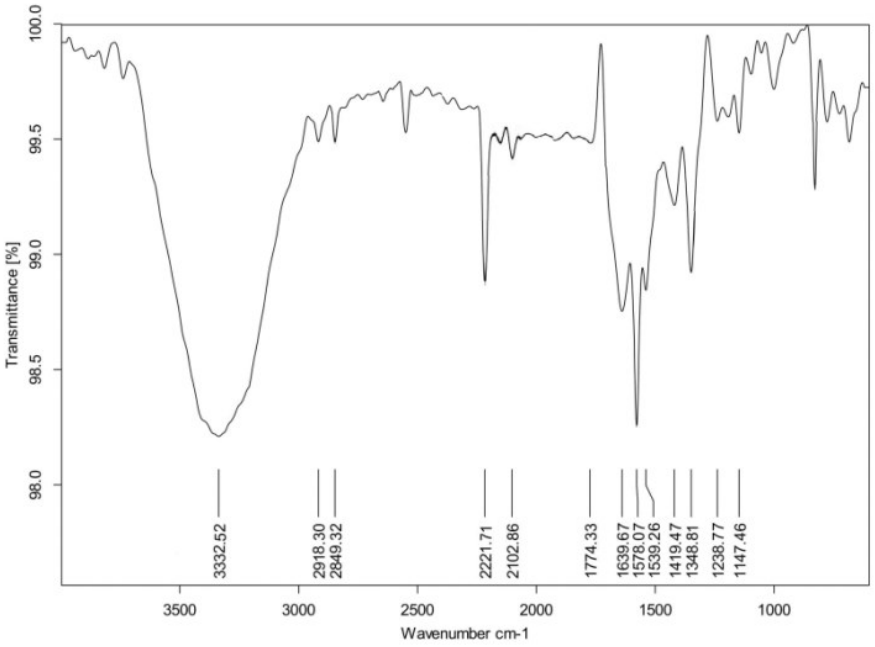

Fig S5: FTIR spectrum of polymer **$TDPy_1$**

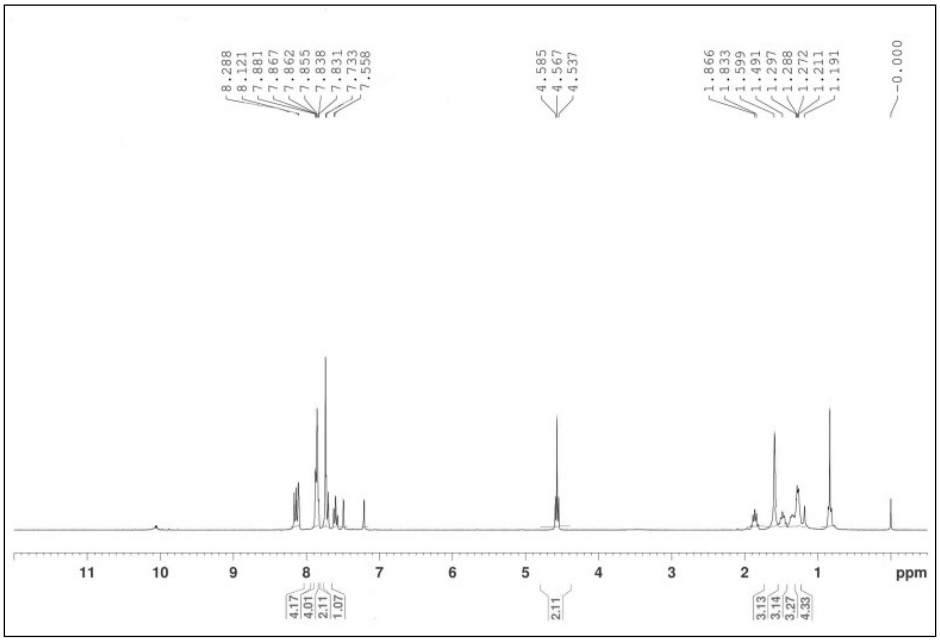

Fig S6: ^{1}H NMR spectrum of polymer **$TDPy_1$**

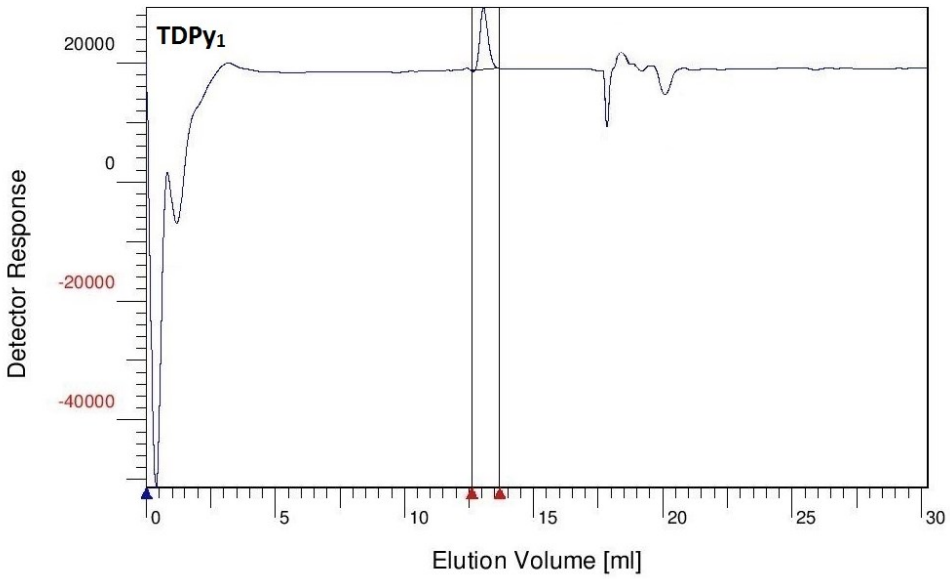

Fig S7: GPC plot of polymer **$TDPy_1$**

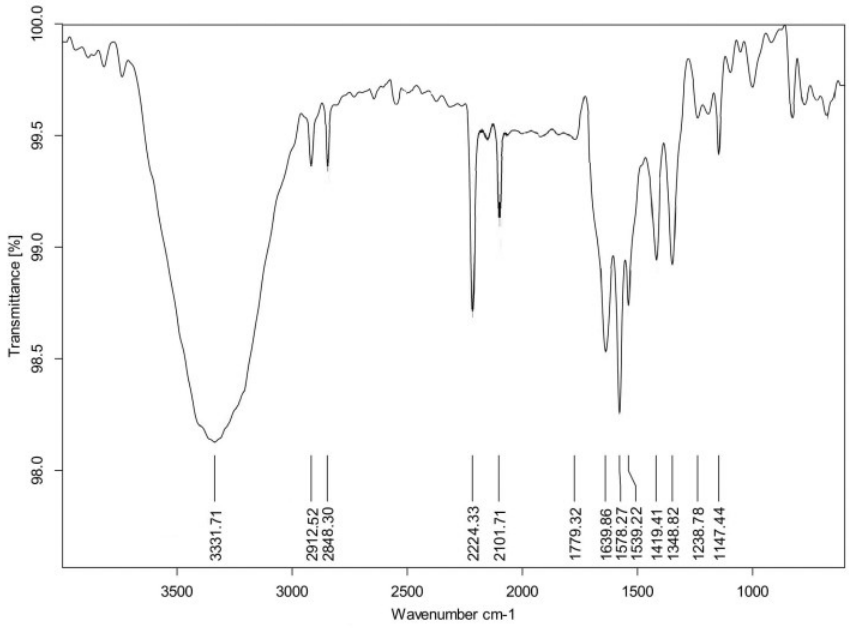

Fig S8: FTIR spectrum of polymer **$TDPy_2$**

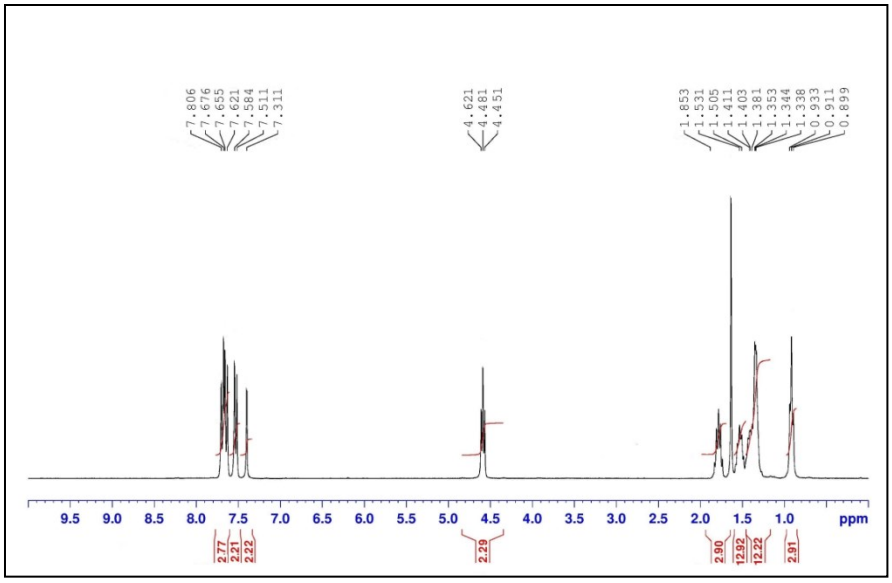

Fig S9: ^{1}H NMR spectrum of polymer **TDPy$_2$**

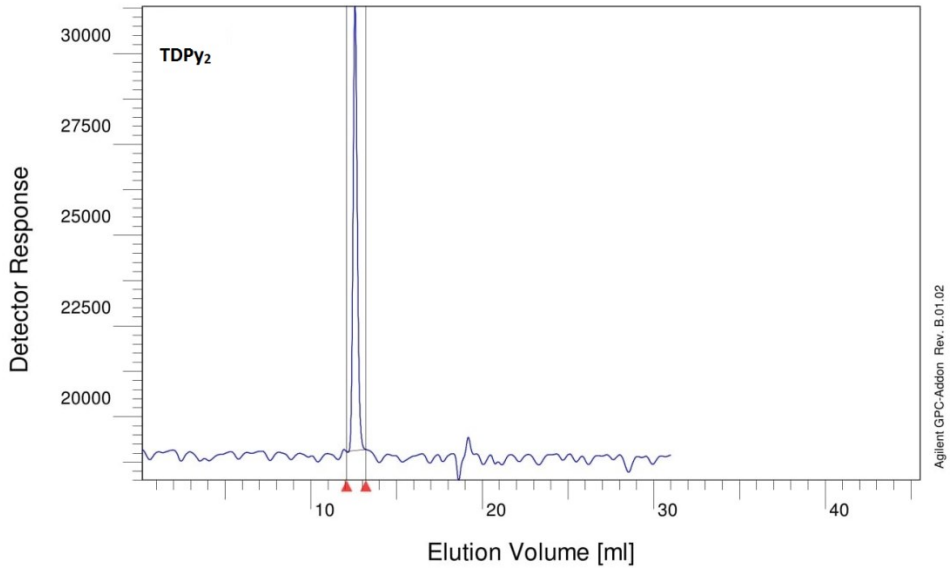

Fig S10: GPC plot of polymer **TDPy$_2$**

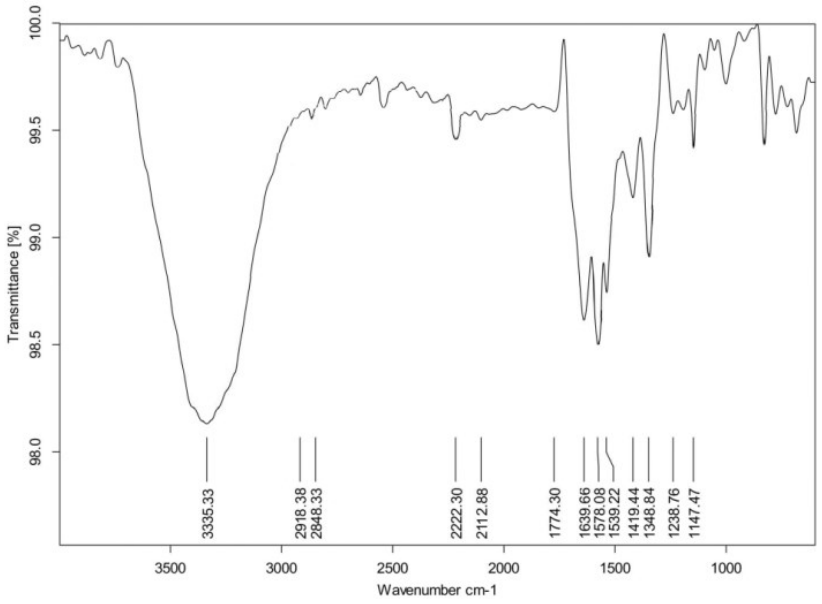

Fig S11: FTIR spectrum of polymer **TDPy$_3$**

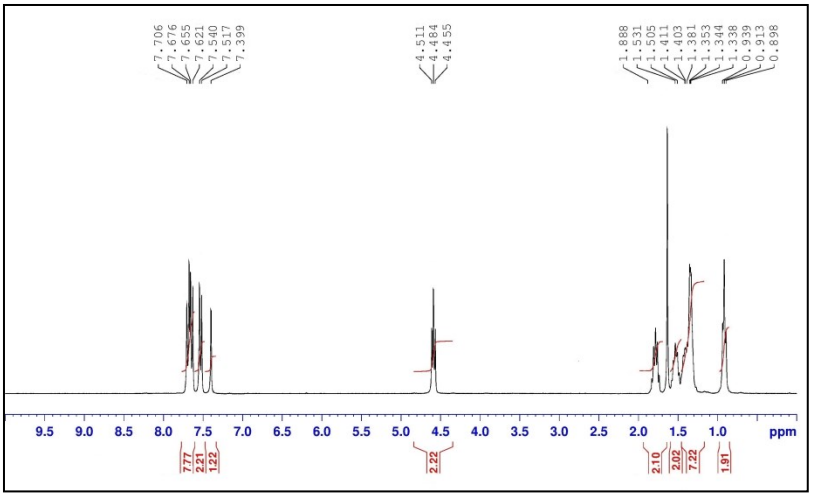

Fig S12: ^{1}H NMR spectrum of polymer **TDPy$_3$**

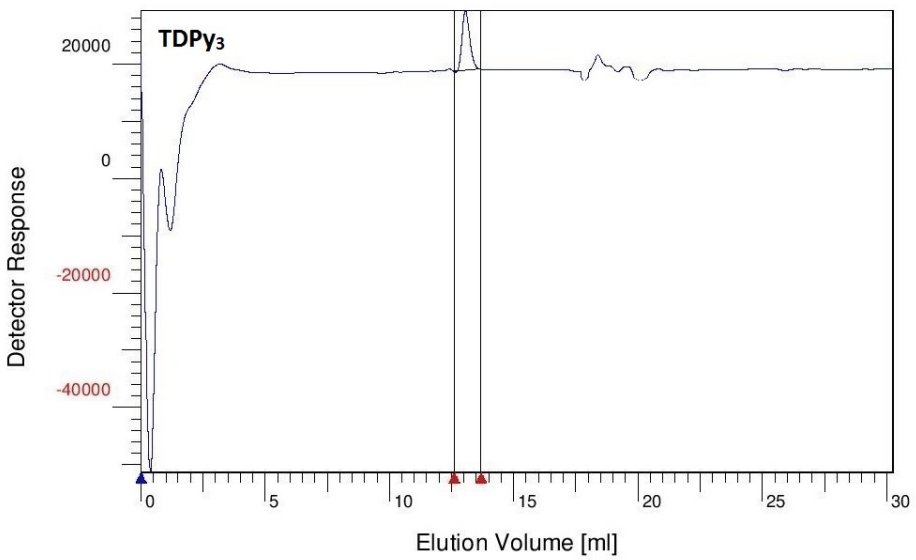

Fig S13: GPC plot of polymer **$TDPy_3$**

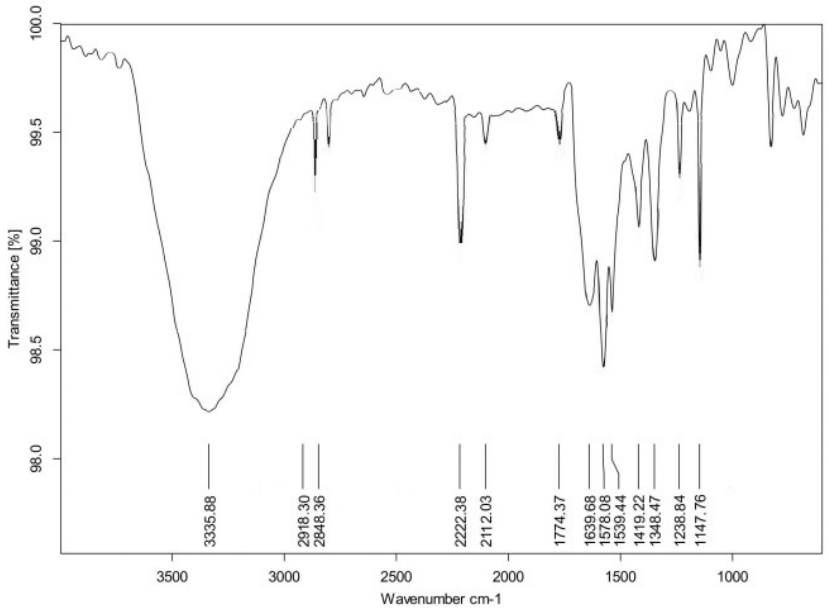

Fig S14: FTIR spectrum of polymer **$TDPy_4$**

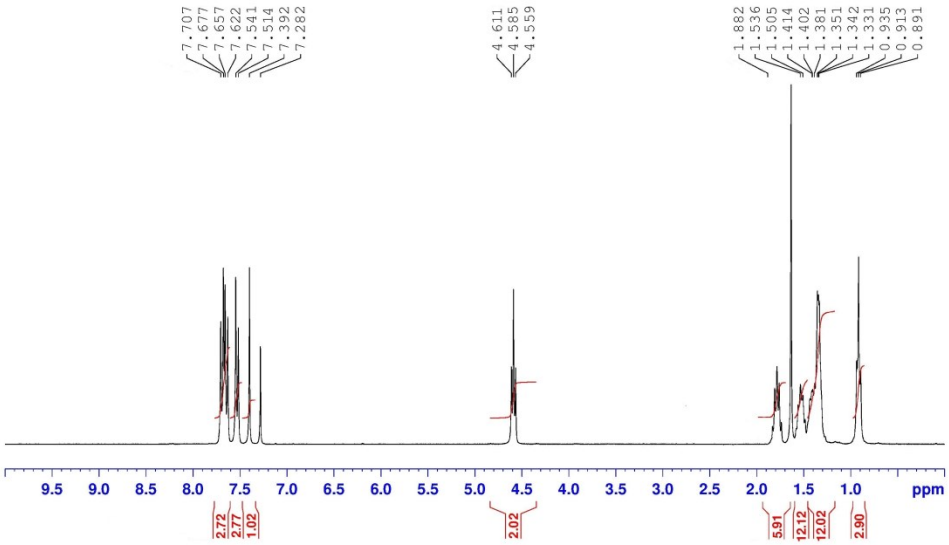

Fig S15: ^{1}H NMR spectrum of polymer **TDPy$_4$**

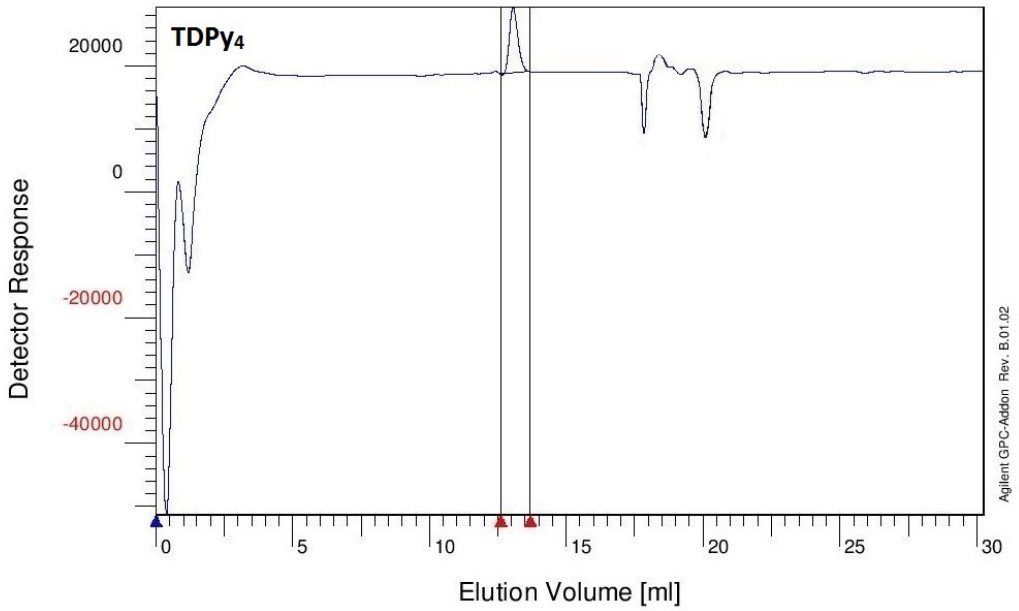

Fig S16: GPC plot of polymer **TDPy$_4$**

3. CV traces of polymers $TDPy_{1-4}$

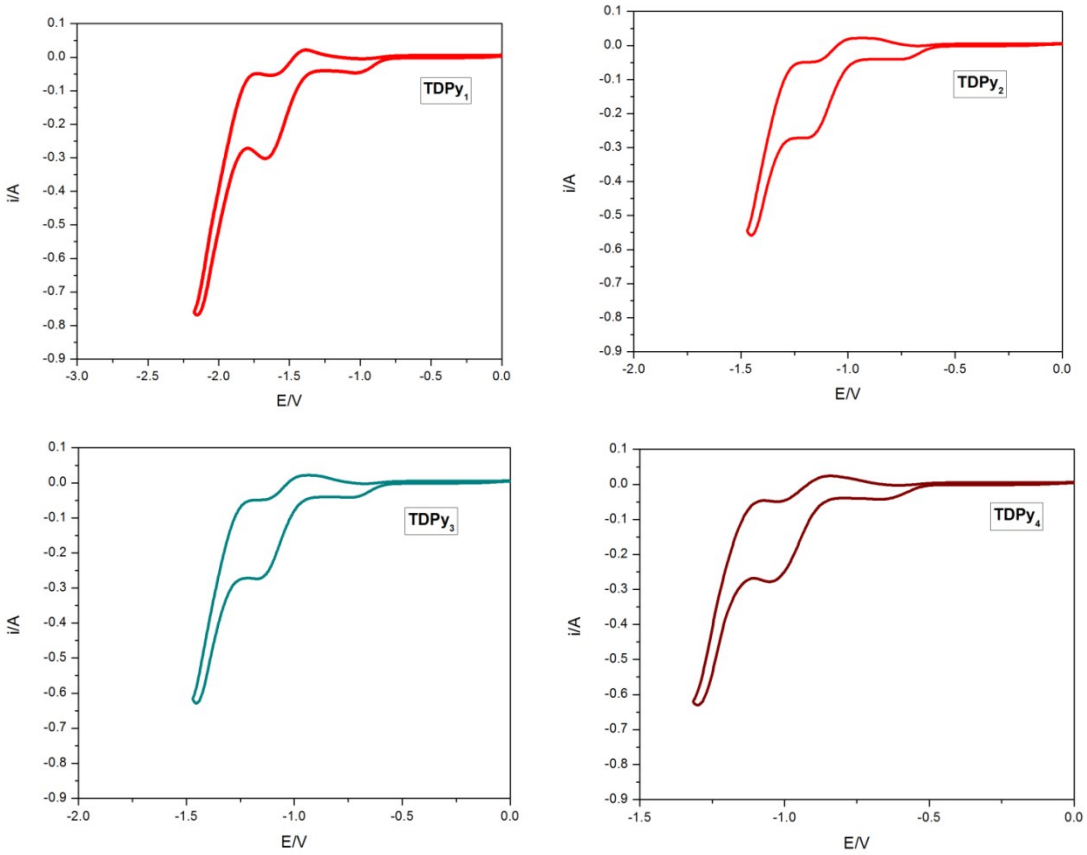

Fig S17: Cyclic voltamograms of polymers **$TDPy_{1-4}$**

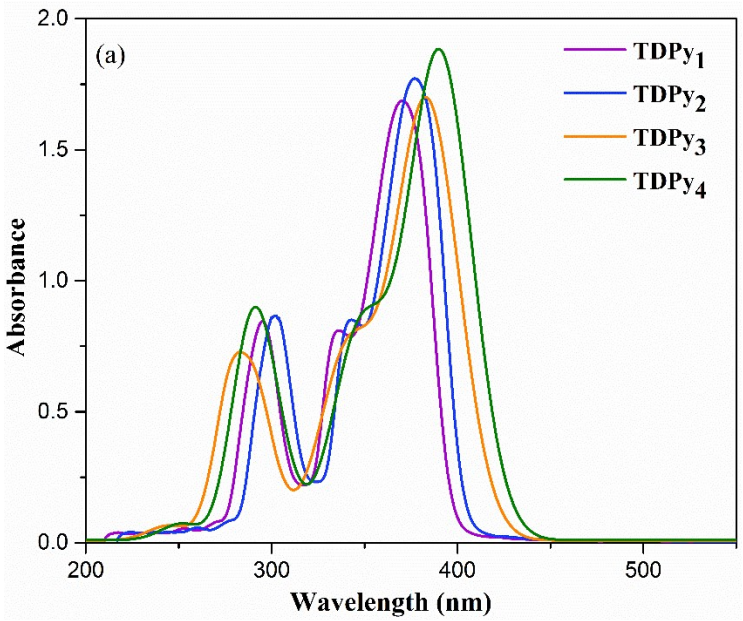

Fig S18: (a) UV-vis absorption spectra of polymers **$TDPy_{1-4}$** in solution state

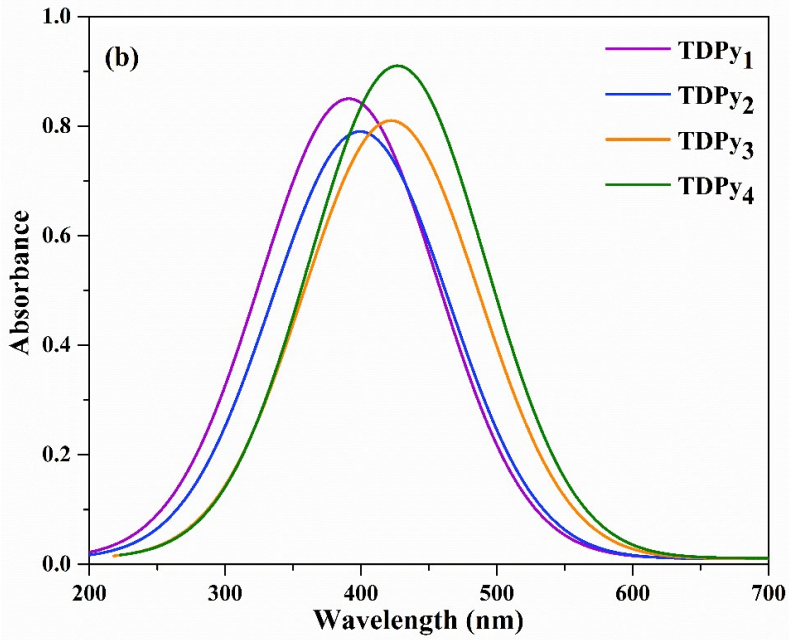

Fig S18: (b) UV-vis absorption spectra of polymers **TDPy$_{1-4}$** in film state

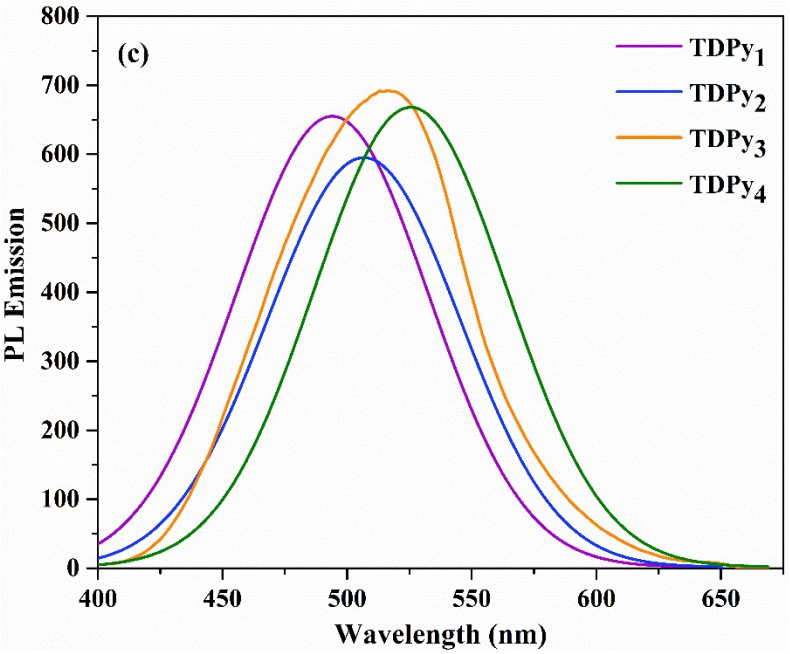

Fig S18: (c) PL emission spectra of polymers **TDPy$_{1-4}$** in solution (THF)

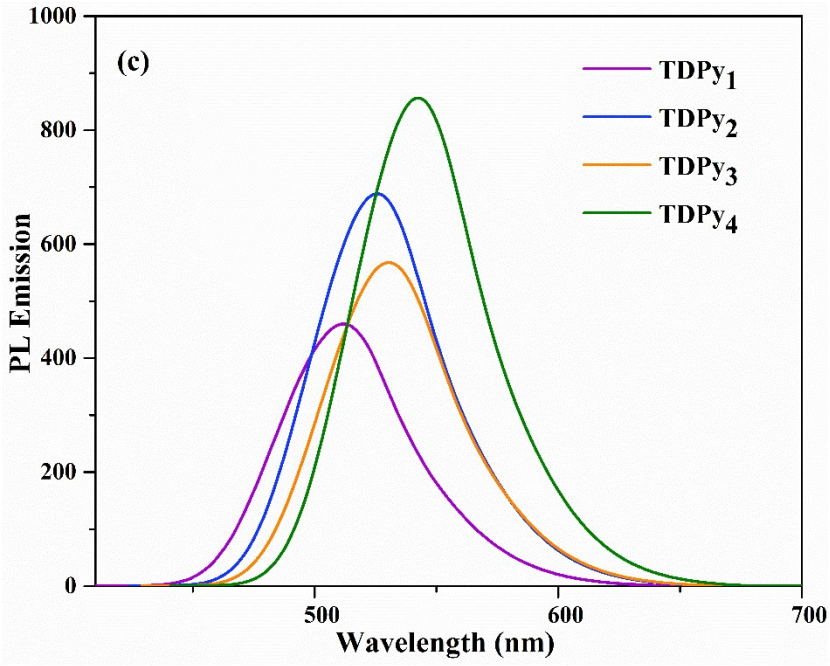

Fig S18: (d) PL emission spectra of polymers **$TDPy_{1-4}$** in film state

4. DFT Calculations

4.1 Cartesian coordinates of the polymer **$TDPy_1$**

ATOM	CARTESIAN COORDINATES		
1 n	7.51197802867245	4.26841610178193	0.05248880909229
2 c	6.68149552311365	1.88270139382775	0.16792303776909
3 c	8.34396612577010	-0.12009936965931	0.63310107219312
4 c	10.86874544673859	0.35801121689119	1.16752320573641
5 c	11.69376238696886	2.90274789364439	1.16683995553343
6 c	9.91367449557519	4.78802309261410	0.47974658812195
7 c	3.94943177473175	1.48184775301882	-0.29611662875276
8 c	12.59794422273307	-1.77087483247333	1.74154249709907
9 c	2.48435217002516	3.42695354910371	-1.33354562136104
10 c	-0.07381392539708	3.10397242651231	-1.78319701830522
11 c	-1.28029904370389	0.81271593747886	-1.22440304959483
12 c	0.18205067621647	-1.12047912776230	-0.15498822378265
13 c	2.74024837209059	-0.79573315713032	0.29912459679479
14 c	11.88004618116071	-3.64346133293719	3.46484079463190
15 c	13.46374889499448	-5.67035771140810	3.98502017258513

16	c	15.80602680450772	-5.91383911998506	2.79403308716085
17	c	16.50506413132597	-4.05370264150802	1.05588436995767
18	c	14.94194767900715	-2.01049421548972	0.54460621938994
19	c	-3.98798804642411	0.39075710421467	-1.76970298751448
20	c	17.55000836238587	-8.08060137494233	3.40509285048423
21	c	-4.86251419646875	-1.88948317897097	-2.65740835051687
22	c	-7.45290452148308	-2.38413868364503	-3.19062376064950
23	c	-9.32041513896059	-0.62456996011381	-2.86624697544094
24	c	-8.51947830330912	1.78273967547575	-1.93983014479658
25	c	-5.87766672988995	2.30703983856728	-1.40167835059558
26	c	-12.05467192197964	-1.11073962694871	-3.42407050496591
27	n	-10.05388967684600	3.72608836532793	-1.47849778535010
28	s	-8.26109554616916	6.05091698700695	-0.42754251937670
29	n	-5.48409302612534	4.63766546961862	-0.53301502020564
30	o	10.44145368074019	7.27206597638368	0.28058920207217
31	c	12.86019807129476	8.16905891826089	-0.56036145046179
32	c	14.13901202433838	3.53364327254690	2.09590663377272
33	n	16.09379790581108	4.06022885184195	2.91359290009979
34	h	7.70192740129171	-2.05438905600213	0.54434201410410
35	h	3.39349080938131	5.20119297619191	-1.78141283676287
36	h	-1.15146747730838	4.64225826605524	-2.58396639253155
37	h	-0.70961909145527	-2.88218297603429	0.38314460167679
38	h	3.77690315369392	-2.32225359023676	1.18137428185765
39	h	10.08649186288586	-3.48741015653788	4.43978732329516
40	h	12.87093507287375	-7.08296418875846	5.34603472440412
41	h	18.30333489096338	-4.20292794302055	0.08525229365277
42	h	15.53687838854762	-0.60992395872806	-0.82272135860538
43	h	16.49969710583967	-9.73867260193187	4.03824931584846
44	h	18.68138776751324	-8.62019690672387	1.76744016909513
45	h	18.86214920253539	-7.56322653393410	4.91755451946489
46	h	-3.51564153078867	-3.38732935783612	-3.01975040444348

47 h	-7.94791786320831	-4.24218853352273	-3.89877644436380
48 h	-12.76859090072145	0.21692353006753	-4.83604938386352
49 h	-12.33387509044878	-3.02874664865583	-4.12268480317718
50 h	-13.21135822364440	-0.84902044497542	-1.73335763394823
51 h	12.48581425177028	10.00189371544681	-1.41423195259446
52 h	14.17133135998788	8.38624068524043	1.00915731772048
53 h	13.67181790143265	6.90490254804929	-1.97475240125417

4.2 Cartesian coordinates of the polymer **$TDPy_2$**

ATOM	CARTESIAN COORDINATES		
1 n	-28.81787209557776	-0.00728935362425	-2.02821102478397
2 c	-31.07034280952243	0.32611370775006	-0.87336692758397
3 c	-31.39324023293652	2.11040583857051	1.03145137128877
4 c	-29.38427003066686	3.66594032842914	1.78244686974499
5 c	-27.05130784136233	3.32473759149024	0.54868174199052
6 c	-26.90824795375151	1.41876041759431	-1.33823179662296
7 c	-33.10604341388002	-1.32738686513775	-1.71978474245963
8 c	-29.84386351279671	5.53838589704346	3.76594724050953
9 c	-32.95329327220526	-3.22169580313348	-3.48543216013148
10 c	-35.25632418317495	-4.49543546077649	-3.91174370026151
11 c	-37.23499738891773	-3.61014776772174	-2.47168965138747
12 s	-36.20150997944442	-1.09097046257682	-0.55222189693120
13 c	-32.05649557862465	6.82981087862975	4.17754618948868
14 c	-31.98674374287851	8.44322923471777	6.31304996130150
15 c	-29.73630847799244	8.38940347619692	7.58133969521489
16 s	-27.61843866280411	6.33906008392737	6.10085969239272
17 c	-39.84670133555963	-4.47243432053811	-2.36032389383433
18 c	-28.98893697962345	9.83271572387673	9.89362848889487
19 c	-41.48126820246002	-3.74608277700096	-0.46349626550060
20 c	-44.03913156189719	-4.54178482619007	-0.31121914018875
21 c	-45.13069181537053	-6.13259099109307	-2.03586502840441
22 c	-43.52606562871757	-6.95992924529163	-4.04301240650539

23 c	-40.90625151720427	-6.15051434909372	-4.22810628079414
24 c	-47.82261269833246	-6.99906001118441	-1.89518072137210
25 n	-44.24233840013337	-8.50310934934974	-5.89928464596613
26 s	-41.76690679144808	-8.89798188000808	-7.74897748135501
27 n	-39.71548176530777	-7.10639636435615	-6.22690207512774
28 c	-24.87209768454209	4.82497667838460	1.01638608059309
29 n	-23.07700396348954	6.02364830034042	1.34086086722791
30 o	-24.65404390510381	1.13100580013054	-2.45690036220135
31 c	-24.42218173066322	-0.73306294537494	-4.41058993815383
32 h	-33.18404025115212	2.27001606119191	1.99803905750969
33 h	-31.20266929528610	-3.66524928146749	-4.43527730649884
34 h	-35.48408686161514	-6.02311923523908	-5.24036678028662
35 h	-33.67935182831343	6.66397021238619	2.94654838127831
36 h	-33.55503352405427	9.62010801656825	6.89293614958018
37 h	-30.58505738309764	10.95471203823151	10.55838378444012
38 h	-28.40930198981152	8.57757492832653	11.42793240961189
39 h	-27.41835378895881	11.11971410373946	9.51559313606180
40 h	-40.79491376487036	-2.51462886840844	1.02058122442797
41 h	-45.15987058844865	-3.85626126749728	1.26081900345539
42 h	-47.93224257395933	-9.05918632495101	-1.79157236754572
43 h	-48.76572832510342	-6.20518361608399	-0.24470410868438
44 h	-48.86890934216147	-6.43962137159676	-3.58596852101866
45 h	-22.46875117850702	-0.60554601159792	-5.03252473879803
46 h	-24.82431048088931	-2.61655192952327	-3.67812733950133
47	-25.70526666096957	-0.32364180063899	-5.96983112433364

4.3 Cartesian coordinates of the polymer **$TDPy_3$**

ATOM	CARTESIAN COORDINATES		
1 n	-20.03262293552890	-2.07339459087150	-1.41346597781957
2 c	-21.75252869453379	-0.37804592752307	-0.60236616482902
3 c	-21.06935190730324	2.10892727354505	-0.07507266280531
4 c	-18.56058122037199	2.90148839305931	-0.32650112428292

5 c	-16.77886212818609	1.10610338692262	-1.13000908795377
6 c	-17.66324567989456	-1.36752813861790	-1.66493127325582
7 c	-24.38873404747794	-1.28793246384187	-0.35865087067437
8 c	-17.88192500438947	5.55944100970487	0.24262750172744
9 c	-25.13798300355500	-3.52644750371739	-1.55458166388567
10 c	-27.59210359673407	-4.41580066858791	-1.35846549940794
11 c	-29.41314406548381	-3.09195502130733	0.03617136075348
12 c	-28.65482445050237	-0.86746170573588	1.25863730825416
13 c	-26.19759025077860	0.01541091913604	1.06801765244029
14 c	-18.76335632191797	6.71076074013875	2.45287393734796
15 c	-18.17681232266927	9.21136804537356	2.99326268963266
16 c	-16.71435706602415	10.65769827276064	1.34122266812874
17 c	-15.85949576305685	9.50841297564456	-0.87674135050339
18 c	-16.41513316105400	7.00334736425345	-1.41673117421595
19 c	-32.06841385380442	-3.94640503138469	0.19565179233197
20 c	-16.03416860862225	13.35518186771160	1.94799281188911
21 c	-34.03108750147338	-2.25704279513378	0.13417855394927
22 c	-36.62801854878424	-2.97420396630399	0.29521993246850
23 c	-37.37752647604881	-5.49614948147550	0.53057558564007
24 c	-35.37917161289365	-7.33229492394836	0.61395795174728
25 c	-32.74868193014811	-6.57593687626231	0.43671626067048
26 n	-35.70977182604745	-9.80665226719504	0.86418127471833
27 s	-32.86638504342172	-11.09905124878094	0.88225278295308
28 n	-31.14958349032060	-8.50888151456950	0.57191369623904
29 c	-38.62494738891635	-1.13077610284883	0.20032148793384
30 c	-41.24361055866941	-1.90801458094686	0.34950235091851
31 c	-41.92376222519325	-4.52495866233705	0.59879501510938
32 c	-39.98329161140572	-6.22013664689598	0.68084773052873
33 n	-38.31363524396942	1.34745637433045	-0.02249483026767
34 s	-41.17406308814449	2.61555610770956	-0.04481954153858
35 n	-42.87010877471435	0.00269699994763	0.23340889885186

36 c	-44.65225002806240	-5.26970435609683	0.75472023654824
37 o	-15.92470804737669	-3.03715019279830	-2.44593855317486
38 c	-16.72587114942633	-5.57521437758005	-2.96054163455870
39 c	-14.14933604041022	1.63215601505393	-1.34790732316802
40 n	-12.01030634483545	2.02442730287483	-1.52615712898650
41 h	-22.48206359552829	3.48074390126776	0.45702005902364
42 h	-23.75989560416205	-4.56085445084851	-2.65184548436472
43 h	-28.10821864456105	-6.14902084202863	-2.30689012594584
44 h	-29.99401184796004	0.15492554097962	2.42029567228743
45 h	-25.68373477302275	1.70813779744424	2.09469666952088
46 h	-19.87996883688999	5.62702634254383	3.78353927366536
47 h	-18.86414828318858	10.04896261560391	4.73254830608561
48 h	-14.73817488355921	10.59046544121117	-2.20740351028712
49 h	-15.73004659795056	6.17865365660912	-3.15796969318744
50 h	-17.33478720337330	14.16879514935432	3.32539703892421
51 h	-14.12935718356138	13.47212198134504	2.74465464724098
52 h	-16.06459331113765	14.53686779215225	0.25710512065441
53 h	-33.63862788191215	-0.26331350967597	-0.09445255439566
54 h	-40.38531165290795	-8.21816399778432	0.86634876636397
55 h	-45.56976043387647	-4.36856711731710	2.37077832603724
56 h	-45.68152882574720	-4.65153168607590	-0.92587005594785
57 h	-44.84925397972346	-7.31182610231167	0.93616327651216
58 h	-15.01075377654577	-6.57272262719216	-3.49192906653757
59 h	-17.57403513006124	-6.43041636535462	-1.28940782598575
60	-18.08747865897753	-5.61414804336973	-4.50692372844407

4.4 Cartesian coordinates of the polymer **$TDPy_4$**

ATOM	CARTESIAN COORDINATES		
1 n	-30.95939631181428	2.15233010872340	-0.99334470939324
2 c	-33.46972046631649	2.02373916681010	-0.66896696962161
3 c	-34.97298593689713	4.19503890390020	-0.59130539335859
4 c	-33.90147948354745	6.55371123156451	-1.04510987402810

5 c	-31.26728532386833	6.65677994324127	-1.52719198512430
6 c	-29.87496771096672	4.36536667082316	-1.35836211748434
7 c	-34.50878826092406	-0.50773150522924	-0.33342978779808
8 c	-35.54633103021289	8.77987631664373	-1.05732655153099
9 c	-33.19089683839425	-2.74281658105416	-0.29339782130289
10 c	-34.72557289153883	-4.89165690738034	0.06387336496524
11 c	-37.26303087662160	-4.35111078755865	0.29850426682536
12 s	-37.74801716352498	-1.06246448161783	0.08936181904422
13 c	-37.94640737794162	8.93146757266238	-2.02233957959965
14 c	-39.11533880340111	11.31251529524403	-1.63846462212386
15 c	-37.64342041132479	12.99908852610023	-0.34980839397221
16 s	-34.72075003082239	11.65433405286830	0.38978035564060
17 c	-39.39643278221829	-6.05240056769121	0.64941609725094
18 c	-38.25247968577978	15.65891579218203	0.38864984435533
19 c	-41.85718759541219	-5.24667348144878	0.41268921001538
20 c	-43.99995096336055	-6.83305594833793	0.76548727778811
21 c	-43.77258393455953	-9.39496392077870	1.37153930356453
22 c	-41.23814239542171	-10.33516256684886	1.61620267759481
23 c	-39.06416450231009	-8.69473606652280	1.27782588265067
24 n	-40.64171850859925	-12.70494679395084	2.18192550751716
25 s	-37.52686512438640	-12.86035196033971	2.27698836069170
26 n	-36.87599540597955	-9.88200927153804	1.59724711148400
27 c	-46.53553869409941	-5.87973687454765	0.50708263419746
28 c	-48.68474311243935	-7.53300837486119	0.86603315475818
29 c	-48.35651996155914	-10.15456408617283	1.49025525524690
30 c	-45.92823836984142	-10.99295475022432	1.71923718767328
31 n	-47.15487121281348	-3.51780505457379	-0.05892324023317
32 s	-50.28097410278708	-3.39962198938510	-0.13131947291340
33 n	-50.89898241923314	-6.38534924247057	0.56438249798365
34 c	-50.62013505314761	-11.81678424608174	1.85502339514718
35 o	-27.35098988401822	4.20548075381010	-1.64985715236389

36 c	-25.67734914396232	6.15284260471157	-0.76479961796577
37 c	-30.12373617325769	8.89973169722630	-2.46814791019956
38 n	-29.18030090359908	10.67461328836763	-3.32147067139117
39 h	-36.96521091313058	4.07128042499541	-0.15975911299719
40 h	-31.16301844733604	-2.79364782406155	-0.51928349872050
41 h	-34.01080130559976	-6.79715962495006	0.15883423251248
42 h	-38.83102632719979	7.38837263758118	-3.02907455823642
43 h	-40.99309702807862	11.77023413818103	-2.30593752673976
44 h	-40.16555787676380	16.12689123628383	-0.21881778501936
45 h	-38.14980769806674	15.93083425747246	2.43357086989107
46 h	-36.95774719535294	17.00965358328925	-0.48588015878214
47 h	-42.24547908956880	-3.30033428359559	-0.08306326745422
48 h	-45.57013071889386	-12.95278397445461	2.18769337982206
49 h	-51.82465126958836	-11.09873597677706	3.37170890674592
50 h	-51.78460799324382	-11.85907254098788	0.14972204355032
51 h	-50.05377988285143	-13.74221840627358	2.31704388995716
52 h	-23.92455922099314	5.18494885722720	-0.29576094295755
53 h	-25.33012280322350	7.56141911032447	-2.22202331308269
54 h	-26.43064232099488	7.06858430680095	0.92320108886982

5. Device fabrication

PLED devices fabricated with a configuration of ITO/PEDOT:PSS/Polymer/Al were used to investigate the electroluminescent (EL) behavior of the polymers **TDPy$_{1-4}$** as emissive materials for polymer light emitting diodes. The device fabrication follows sandwich type of structure with poly(3,4-ethylenedioxythiophene):poly(styrenesulphonate) (PEDOT: PSS) coated indium tin oxide (ITO) glass as the anode, the spin coated newly synthesized polymer as the emissive layer and aluminum as the cathode. To fabricate PLEDs of device configuration ITO/PEDOT: PSS/polymer/Al, first the indium tin oxide (ITO) coated glass substrates with a sheet resistance of 20 Ω/square and a thickness (ITO) of 120 nm were cleaned using deionized water, acetone, trichloroethylene and isopropyl alcohol sequentially for about 30 min each using an ultrasonic bath and dried in a vacuum oven. Then ITO surface was treated with oxygen plasma for about 10 min to increase its work function. Later, a hole injection layer of PEDOT:

PSS was spin coated on the cleaned and patterned ITO substrate at 4000 rpm with about 50-60 nm in thickness and was dried by baking at 120 ^{0}C in vacuum for 50 min. Then, the emitting polymer layer was spin cast onto the PEDOT: PSS layer at a speed of 2000 rpm from chlorobenzene solution (10 mg/mL) through a 0.45 μm teflon filter, followed by vacuum annealing at 150 ^{0}C for ~2 h in order to remove the organic fraction. Finally, the coated ITO was transferred to a deposition chamber, where a layer of Al electrode was vacuum deposited on the polymer layer with about 200 nm in thickness by thermal evaporation method at a pressure of 1 × 10^{-6} Torr. Four pixels, each of active area of 4×4 mm^2 were defined per substrate and used to assess the reproducibility of the device performance. The complete fabricated devices were finally annealed at 100 ^{o}C in vacuum for 5 min before being characterized. All the characterizations of the light-emitting diode devices were carried out at room temperature under ambient conditions without protective encapsulation.