Electronic Supplementary Material (ESI) for RSC Advances.

Poly(Ethylene Glycol)-Poly(Propylene Glycol)-Poly(Ethylene Glycol) and Polyvinylidene Fluoride Blend Doped with the Oxydianiline Based Thiourea Derivatives as a Novel and Modest Gel Electrolyte System for Dye-Sensitized Solar Cell Applications

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1. Synthesis of organic additives:

1.1. Synthesis of OPMT

4-Methoxyphenyl isothiocyanate (2.2 mmol) with 4,4'-oxydianiline (1 mmol) was stirred in anhydrous methanol at room temperature for 2-3 days. The product obtained was white precipitate. It was then filtered, washed with methanol and dried under vacuum to obtain **1,1'-(oxybis(4,1-phenylene))bis(3-(4-methoxyphenyl)thiourea) (OPMT)** as white solid with 78% yield. The relevant scheme is given in the Figure S1. ^{1}H NMR (400 MHz, DMSO d_6) δ 9.62 (s, 2H), 9.58 (s, 2H), 7.46 (d, *J* = 8.8 Hz, 4H), 7.33 (d, *J* = 8.8 Hz, 4H), 6.99 (d, *J* = 8.8 Hz, 4H), 6.92 (d, *J* = 8.8 Hz, 4H), 3.75 (s, 6H); ^{13}C NMR (100 MHz, DMSO d_6) δ 180.56, 157.07, 154.06, 135.37, 132.61, 126.63, 126.43, 118.90, 114.15, 55.71. HRMS (ESI): calcd. For $C_{28}H_{26}N_4O_3S_2$ $[M+H]^+$ 530.1446 found to be 531.1519.

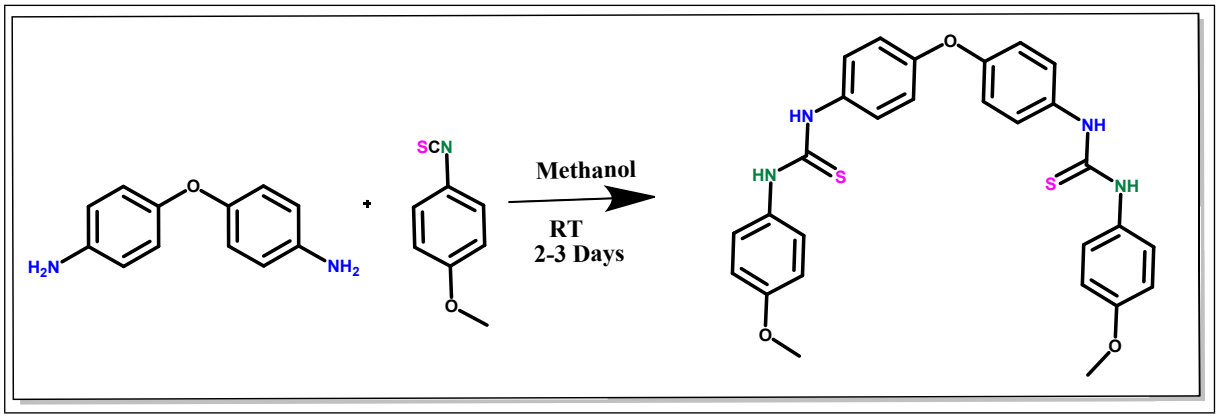

Figure S1. Scheme for the synthesis of organic additive-1 (OPMT)

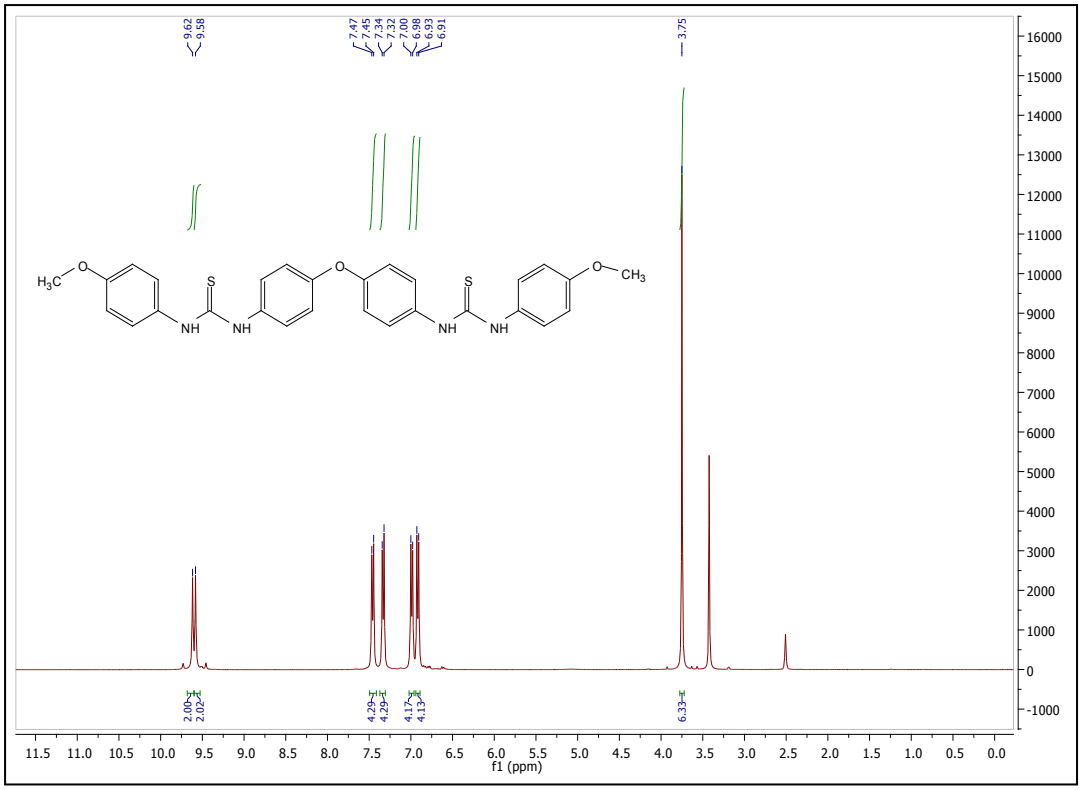

Figure S2.^{1}H NMR spectrum of OPMT

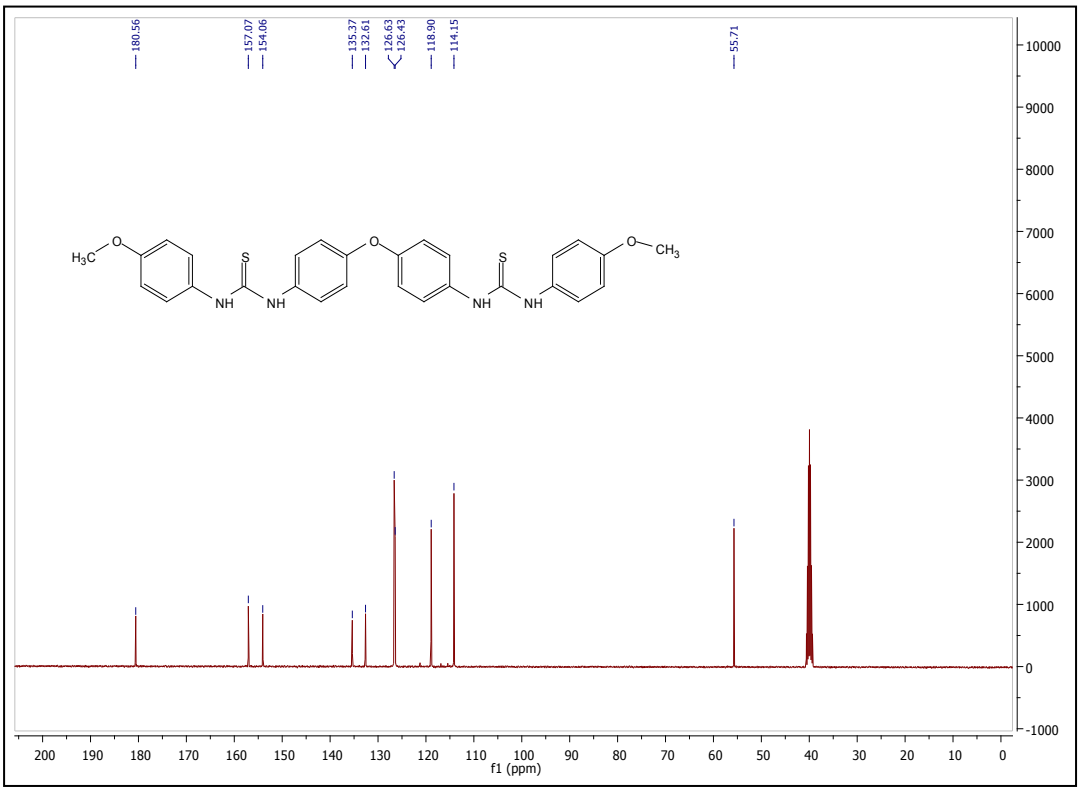

Figure S3. ^{13}C NMR spectrum of OPMT

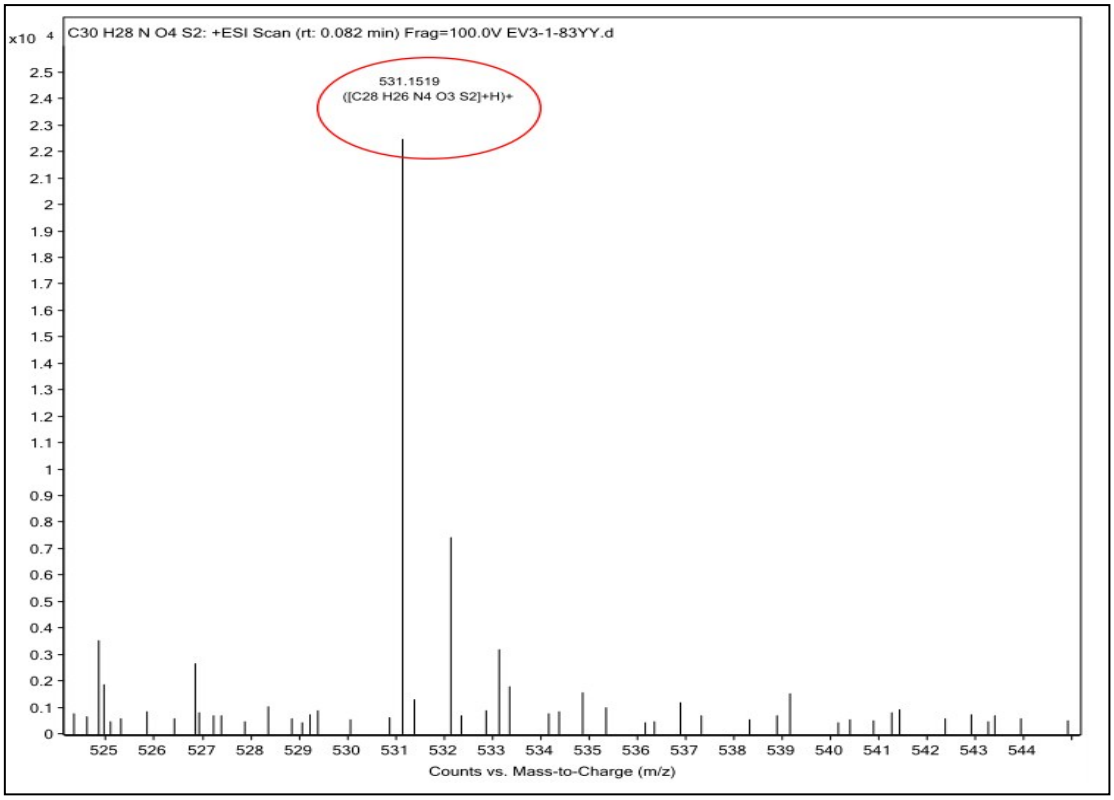

Figure S4. HRMS spectrum of OPMT

1.2. Synthesis of OPBT

4-n-Butylphenyl isothiocyanate (2.2 mmol) with 4,4'-oxydianiline (1 mmol) was stirred in anhydrous methanol at room temperature for 2-3 days. The product obtained was white precipitate which was then filtered, washed with methanol and dried under vacuum to obtain **1,1'-(oxybis(4,1-phenylene))bis(3-(4-butylphenyl)thiourea)(OPBT)** as a white solid with 75% yield. The relevant scheme is given in the Figure S5.^{1}H NMR (400 MHz, DMSO d_6) δ 9.72 (s,2H), 9.70 (s, 2H), 7.48 (d, J = 8.6 Hz, 4H), 7.38 (d, J = 8.1 Hz, 4H), 7.15 (d, J = 8.1 Hz, 4H), 6.99 (d, J = 8.6 Hz, 4H), 2.55 (m, 4H), 1.57 – 1.43 (m, 4H), 1.32 (m, 4H), 0.91 (t, J = 7.3 Hz, 6H), ^{13}CNMR (100MHz, DMSO d_6) δ 180.19, 154.02, 139.10, 137.47, 135.36, 128.71, 126.29, 124.31, 118.88, 34.81, 33.67, 22.24, 14.28; HRMS (ESI):calcd. For $C_{34}H_{38}N_4OS_2$ $[M+H]^+$ 582.2487 found to be 583.2560.

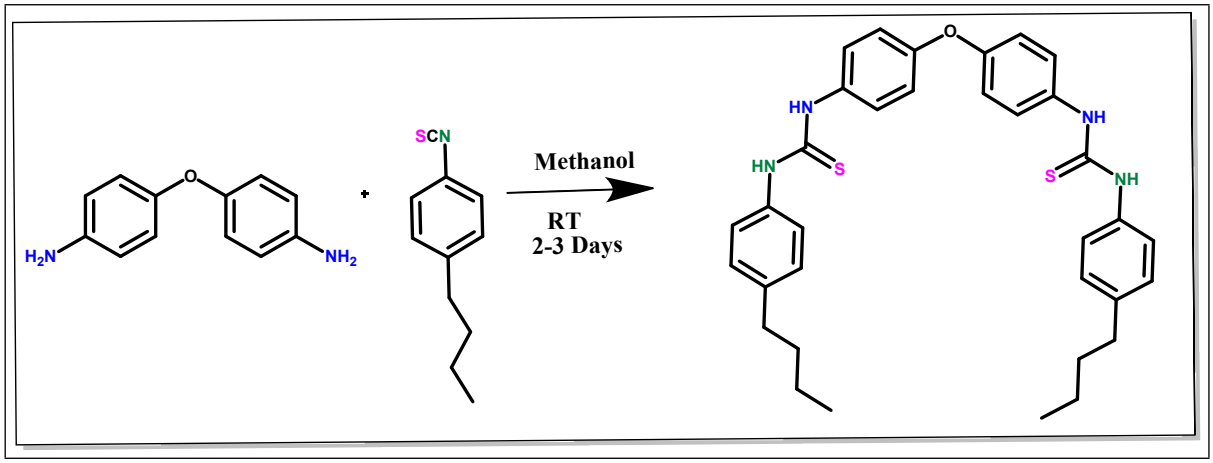

Figure S5. Scheme for the synthesis of organic additive-2 (OPBT)

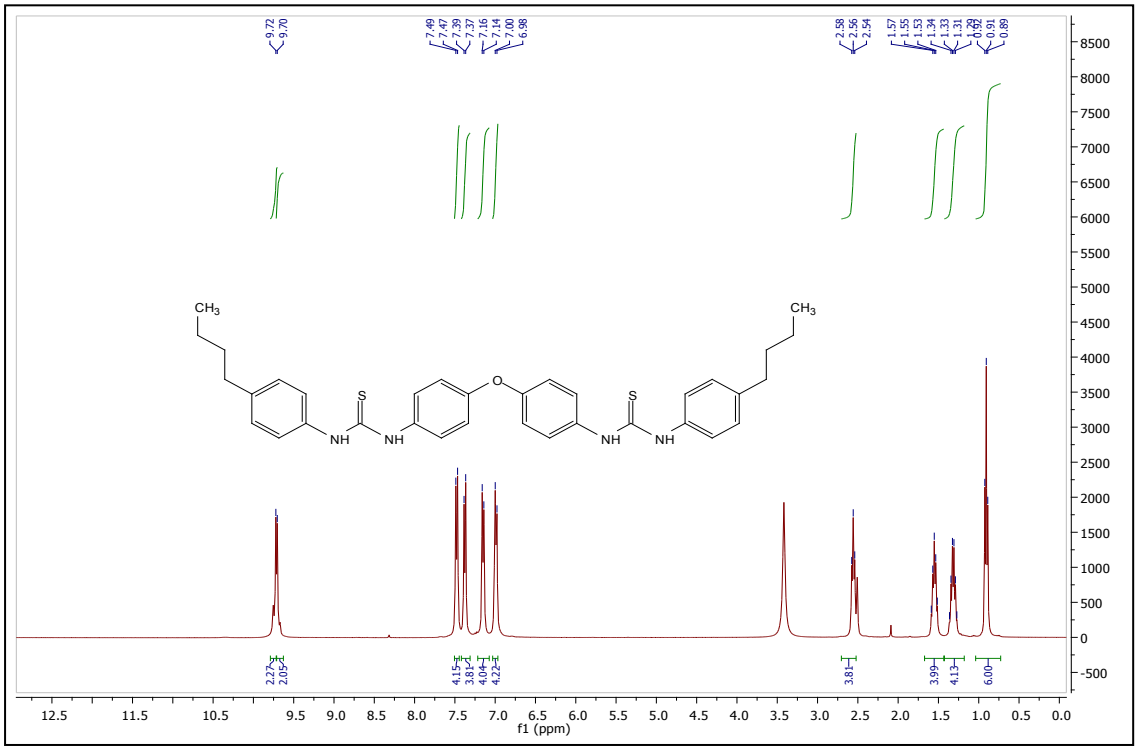

Figure S6. ^{1}H NMR spectrum of OPBT

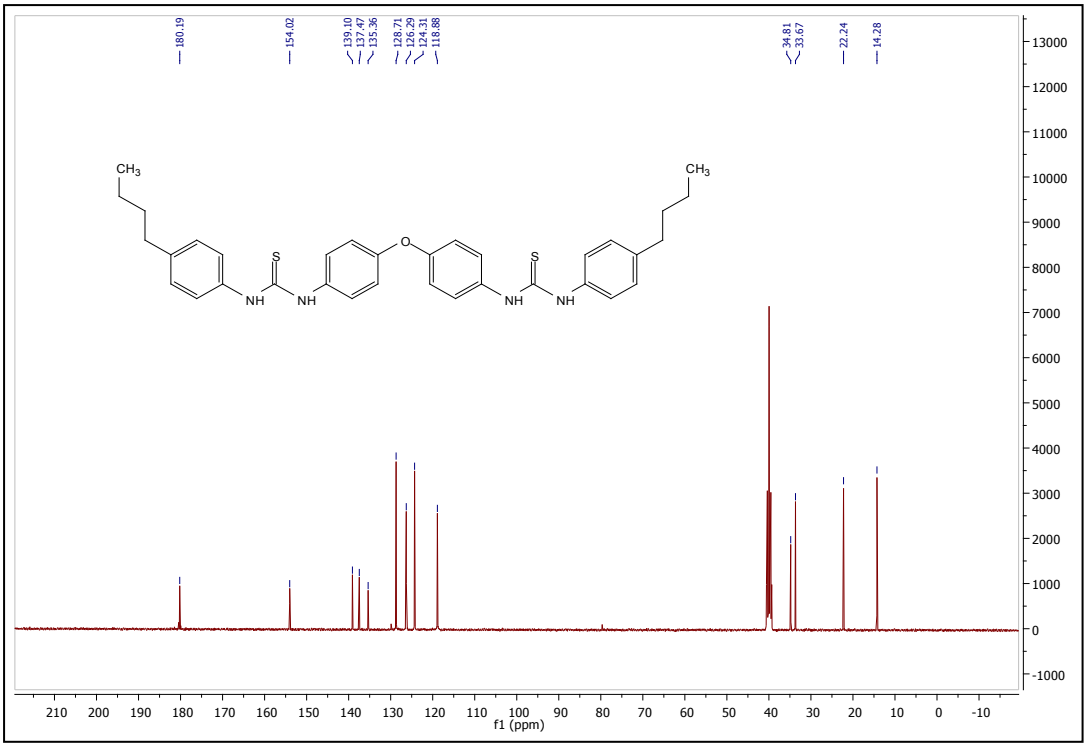

Figure S7. ^{13}C NMR spectrum of OPBT

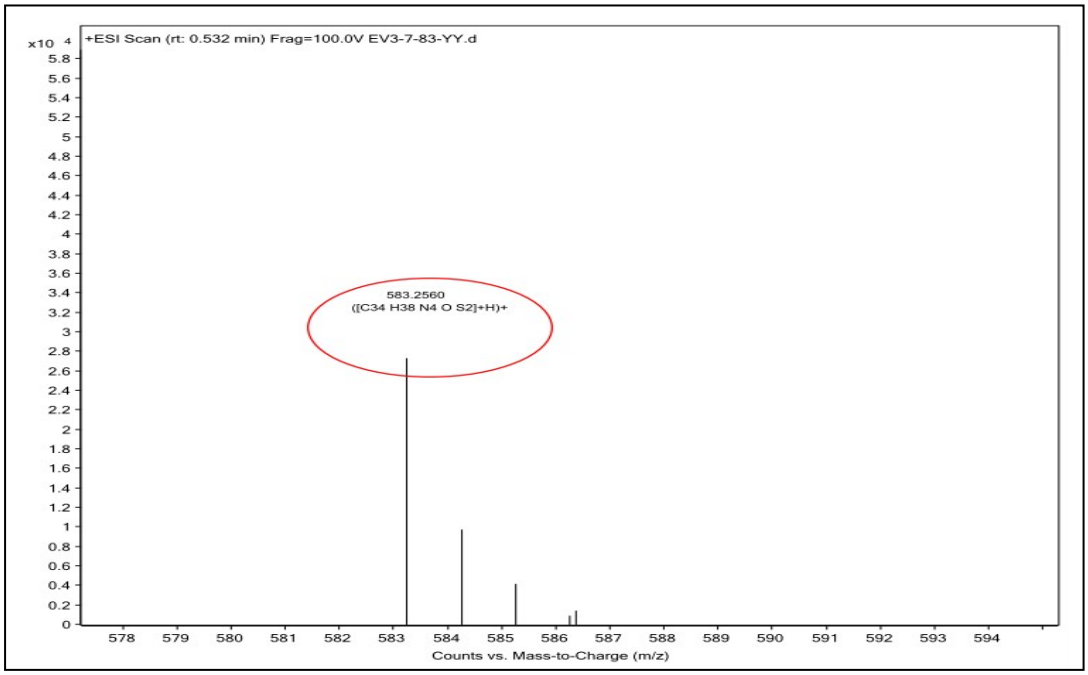

Figure S8. HRMS spectrum of OPBT

1.3. Synthesis of OPNT

1-Naphthyl isothiocyanate (2.2 mmol) with 4,4'-oxydianiline (1 mmol) was stirred in anhydrous methanol at room temperature for 2-3 days to obtain the product as white precipitate. It was then filtered, washed with methanol and dried under vacuum to obtain **1,1'-(oxybis(4,1-phenylene))bis(3-(naphthalen-1-yl)thiourea)(OPNT)** as white solid with 70% yield. The relevant scheme is given in the Figure S9. ^{1}H NMR (400 MHz, DMSO d_6) δ 9.88 (s, 2H), 9.77 (s, 2H), 8.05 – 7.99 (m, 2H), 7.98 (d, *J* = 7.9 Hz, 2H), 7.88 (d, *J* = 7.3 Hz, 2H), 7.62 (d, *J* = 9.8 Hz, 2H), 7.58-7.54 (m, 6H), 7.53-7.48 (m, 4H),7.02 (t, *J* = 7.3 Hz, 4H); ^{13}C NMR (100 MHz, DMSO d_6) δ 181.97, 154.26, 135.61, 134.42, 130.49, 128.63, 127.28, 126.88, 126.70, 126.62, 126.55, 126.17, 125.93, 123.63, 118.91; HRMS (ESI): calcd. For $C_{34}H_{26}N_4OS_2[M+H]^+$ 570.1548 found to be 571.1621.

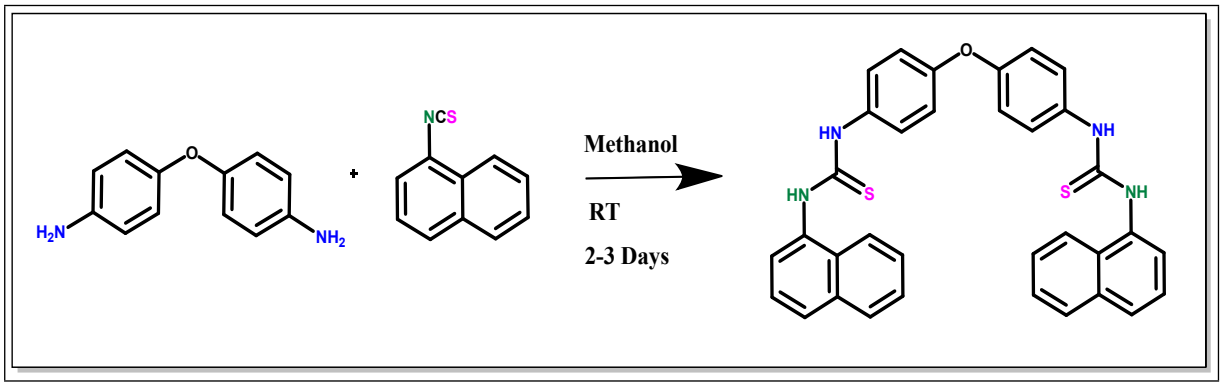

Figure S9. Scheme for the synthesis of organic additive-3 (OPNT)

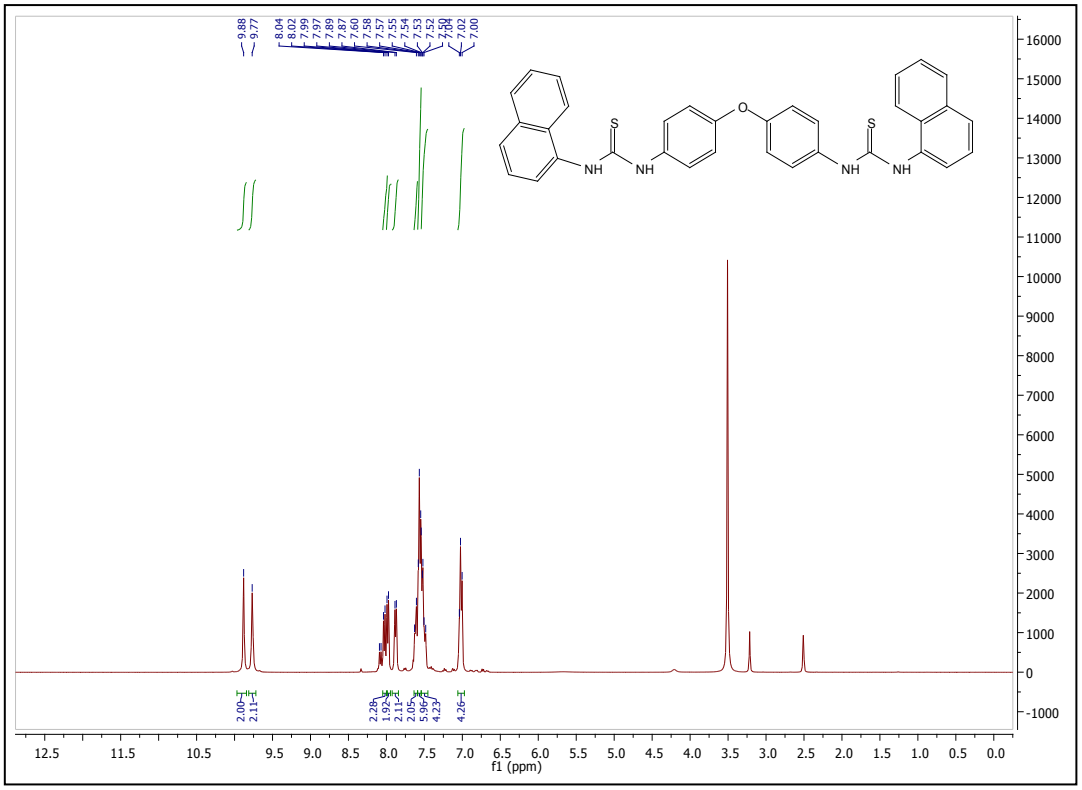

Figure S10. ^{1}H NMR spectrum of OPNT

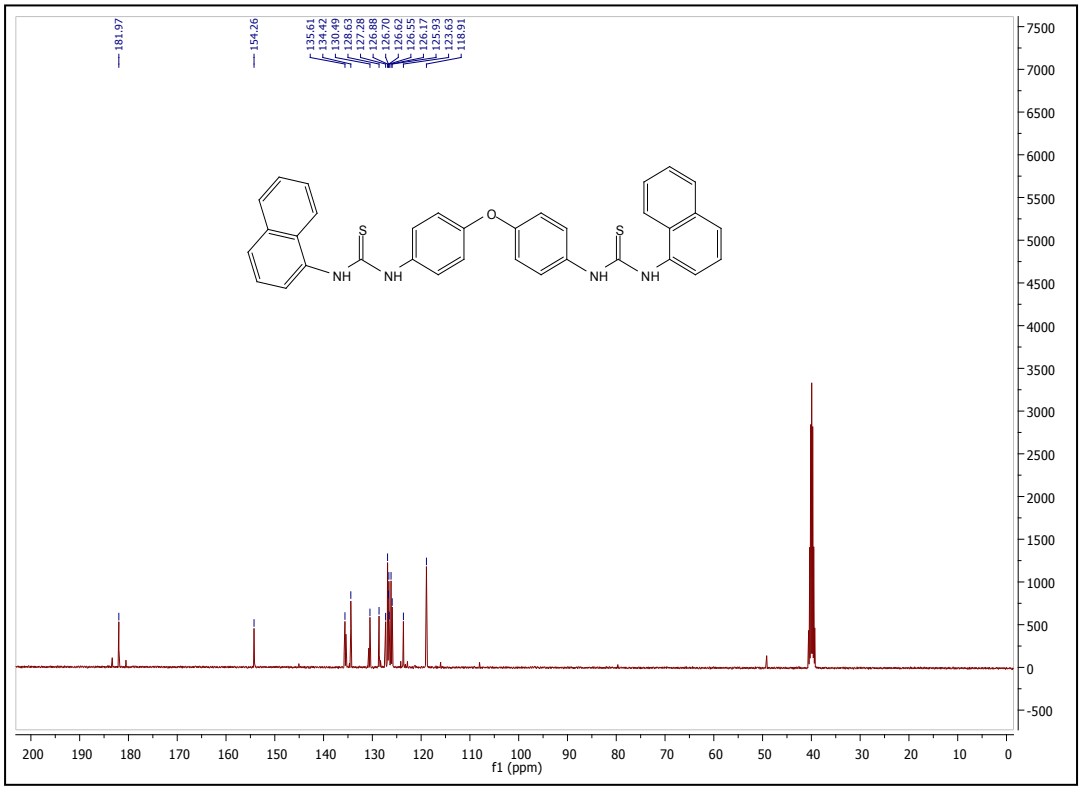

Figure S11. ^{13}C NMR spectrum of OPNT

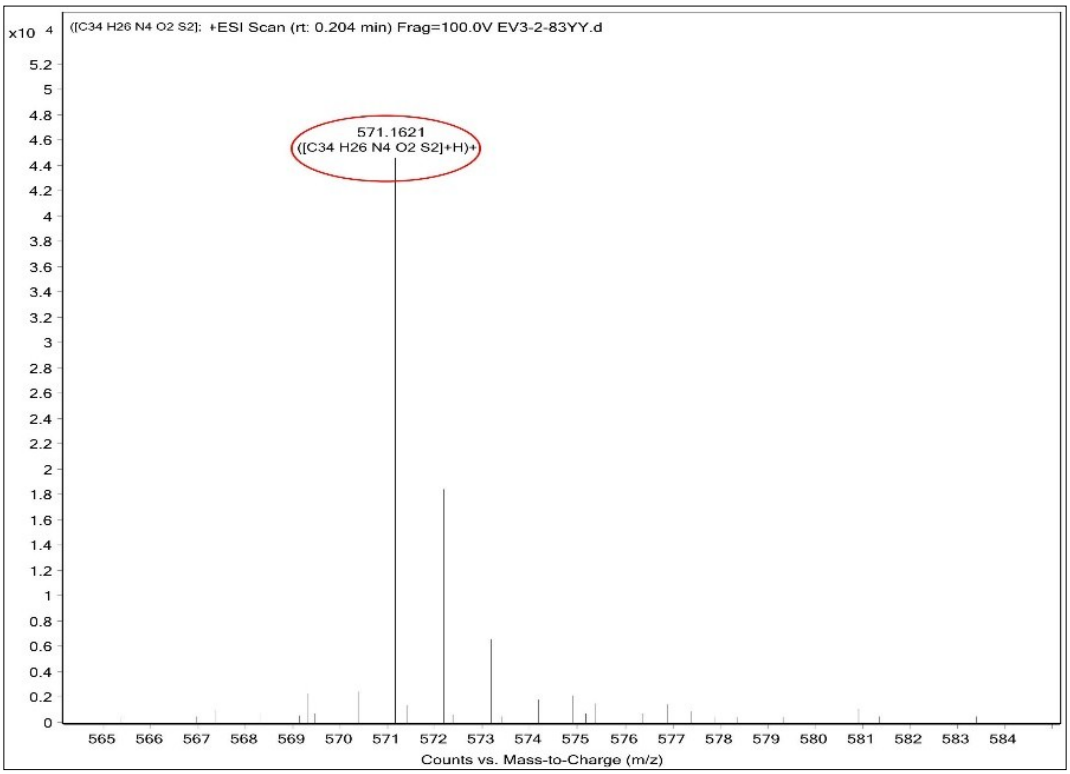

Figure S12. HRMS spectrum of OPNT

1.4. Synthesis of OPPT

2-isothiocyanato-6-methylpyridine was synthesized by already reported procedure.[48] 2-isothiocyanato-6-methylpyridine (2.2 mmol) with 4,4'-oxydianiline (1 mmol) was stirred in anhydrous methanol at room temperature for 2-3 days to obtain the product as white precipitate. It was then filtered, washed with methanol and dried under vacuum to obtain**1,1'-(oxybis(4,1-phenylene))bis(3-(6-methylpyridin-2-yl)thiourea) (OPPT)** with 85% yield. The relevant scheme is given in the Figure S13.^{1}H NMR (500 MHz, DMSO d_6) 14.14 (s,2H), 10.84 (s, 2H), 7.78 (d, J = 8.8 Hz, 4H), 7.73 (t, J = 7.9 Hz, 2H), 7.09-7.07 (m, 6H), 6.97 (d, J = 7.4 Hz, 2H), 2.48 (s,6H); ^{13}C NMR (125 MHz, DMSO d_6) δ 178.45, 154.87, 153.58, 140.11, 134.88, 125.76, 119.06, 117.76, 110.23, 54.38, 23.92. HRMS (ESI): calcd. For $C_{26}H_{24}N_6OS_2$ $[M+H]^+$ 500.1453 found to be 501.1526.

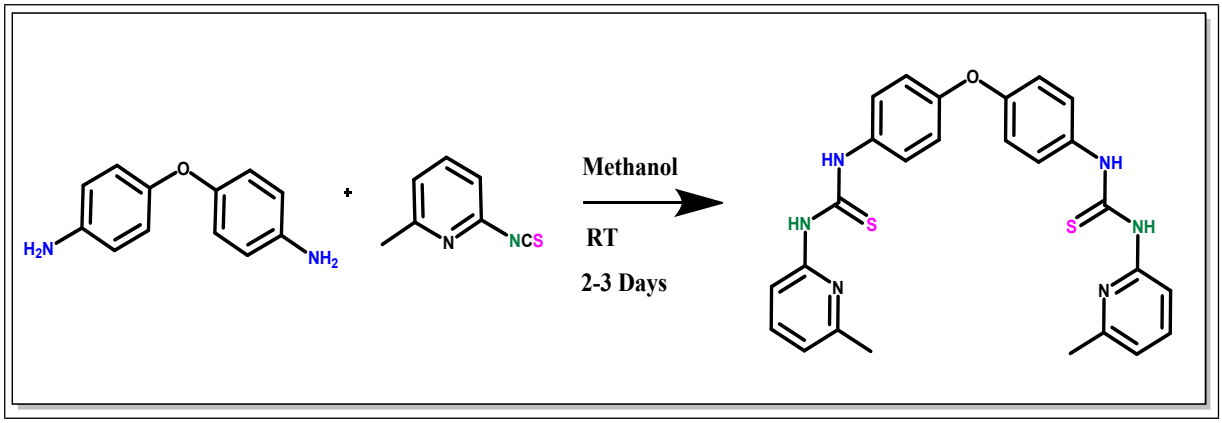

Figure S13. Scheme for the synthesis of organic additive-4 (OPPT)

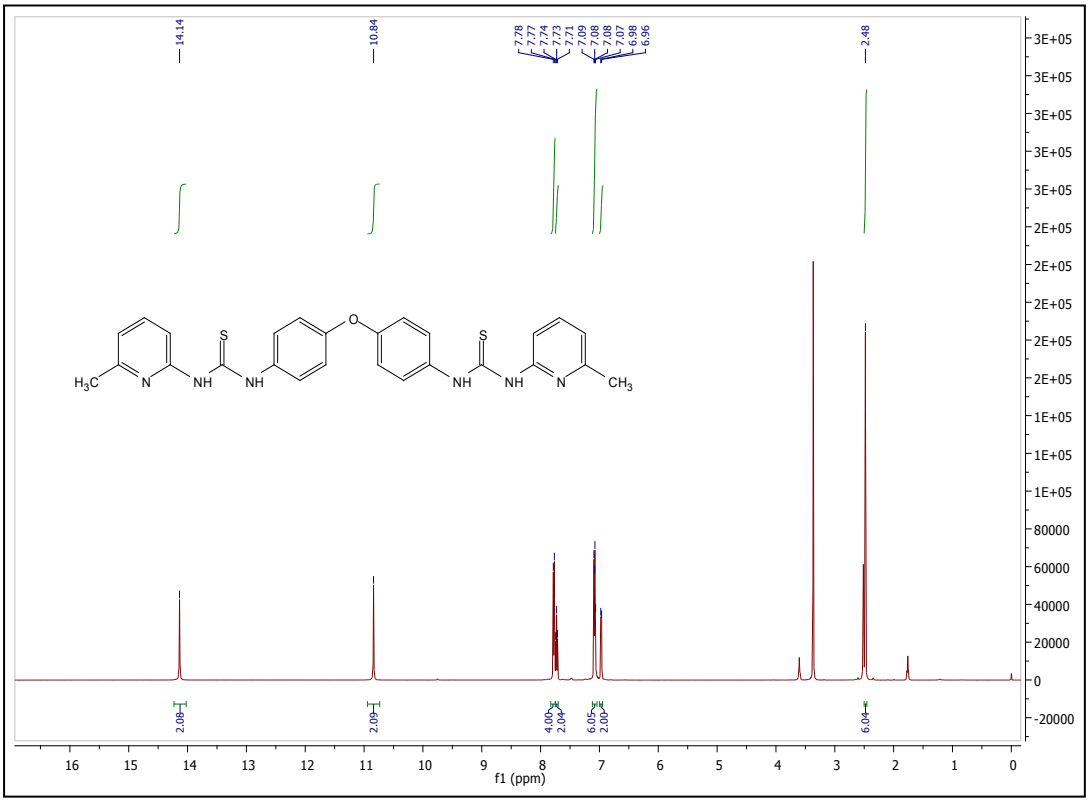

Figure S14. ^{1}H NMR spectrum of OPPT

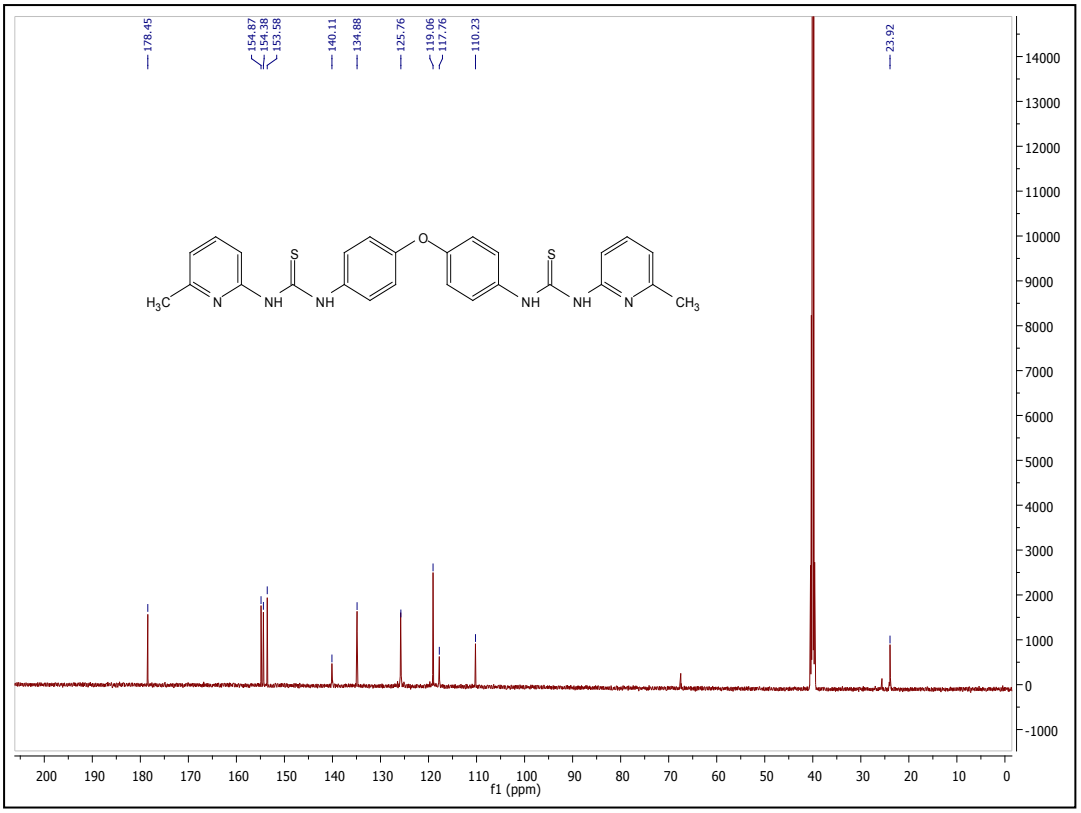

Figure S15. ^{13}C NMR spectrum of OPPT

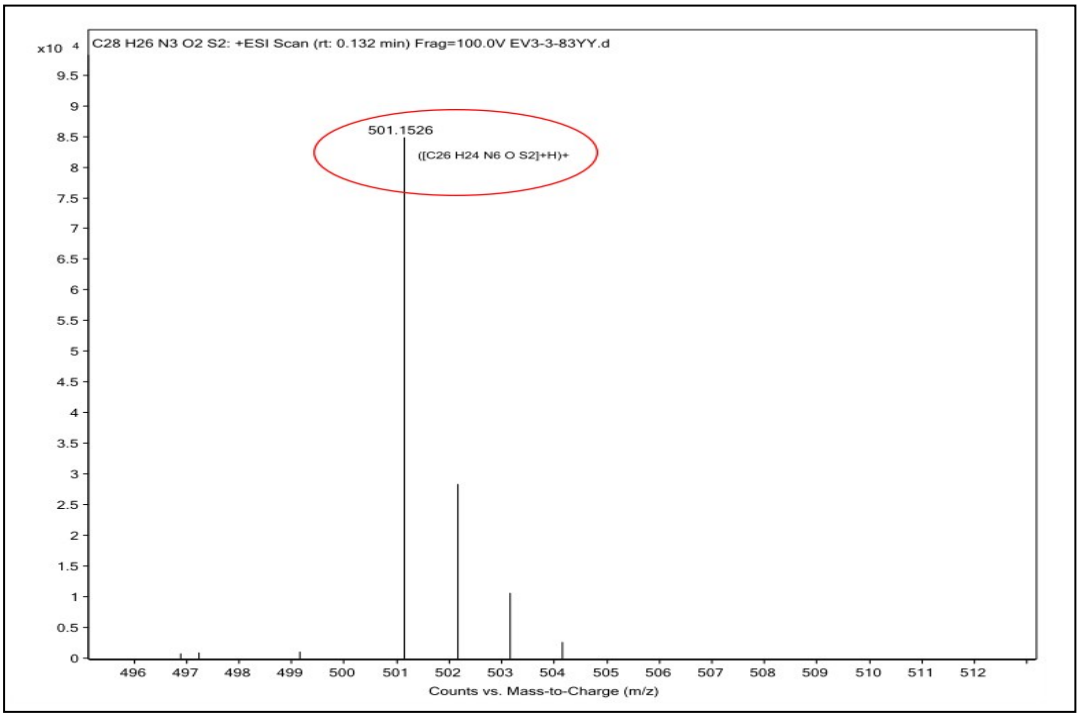

Figure S16. HRMS spectrum of OPPT

Table S1: Statistical analysis of the efficiency of the fabricated DSSC with gel polymer electrolyte system (Device-1)

S. No	Gel polymer electrolyte system	J_{sc}(mA cm^{-2})	V_{oc} (mV)	FF	η %
1	Device-1	10.4	755	0.51	4
2	Device-1	10.5	746	0.51	3.99
3	Device-1	10.3	756	0.51	3.97
Average(Mean)		10.4	752.3333	0.51	3.986667
Standard Deviation		0.08167	4.4969	0	0.0123

Table S2: Statistical analysis of the efficiency of the fabricated DSSC with gel polymer electrolyte system (Device-2)

S.No	Gel polymer electrolyte system	J_{sc}(mA cm^{-2})	V_{oc} (mV)	FF	η %
1	Device-2	13.6	785	0.52	5.55
2	Device-2	13.5	787	0.52	5.52
3	Device-2	13.7	778	0.52	5.54
Average(Mean)		13.6	783.3333	0.52	5.536667
Standard Deviation		0.08167	3.8584	0	0.0123

Table S3: Statistical analysis of the efficiency of the fabricated DSSC with gel polymer electrolyte system (Device-3)

S.No	Gel polymer electrolyte system	J_{sc}(mA cm^{-2})	V_{oc} (mV)	FF	η %
1	Device-3	11.1	765	0.51	4.33
2	Device-3	11.2	758	0.51	4.32
3	Device-3	11.3	752	0.51	4.33
Average(Mean)		11.2	758.3333	0.51	4.326667
Standard Deviation		0.08167	5.3124	0	0.00471

Table S4: Statistical analysis of the efficiency of the fabricated DSSC with gel polymer electrolyte system (Device-4)

S.No	Gel polymer electrolyte system	J_{sc}(mA cm^{-2})	V_{oc} (mV)	FF	η %
1	Device-4	12.2	775	0.52	4.91
2	Device-4	12.1	776	0.52	4.88
3	Device-4	12.1	773	0.52	4.86
Average(Mean)		12.13333	774.6667	0.52	4.883333
Standard Deviation		0.04711	1.2471	0	0.0649

Table S5: Statistical analysis of the efficiency of the fabricated DSSC with gel polymer electrolyte system (Device-5)

S.No	Gel polymer electrolyte system	J_{sc}(mA cm^{-2})	V_{oc} (mV)	FF	η %
1	Device-5	14	790	0.52	5.75
2	Device-5	14.1	784	0.52	5.74
3	Device-5	14.2	777	0.52	5.73
Average(Mean)		14.1	783.6667	0.52	5.74
Standard Deviation		0.08167	5.3124	0	0.00816

Table S6: Statistical analysis of the efficiency of the fabricated DSSC with gel polymer electrolyte system (Device-6)

S.No	Gel polymer electrolyte system	J_{sc}(mA cm^{-2})	V_{oc} (mV)	FF	η %
1	Device-6	12.6	780	0.51	5.01
2	Device-6	12.5	781	0.51	4.98
3	Device-6	12.4	782	0.51	4.94
Average(Mean)		12.5	781	0.51	4.976667
Standard Deviation		0.08167	0.8164	0	0.0283

Table S7: Photovoltaic parameters of the fabricated DSSC using prepared GPE's utilized for the stability test.

Device	J_{sc} (mA)	V_{oc} (mV)	ff	ŋ (%)
Device-1	10.4	755	0.51	4 %
Device-2	13.6	785	0.52	5.55%
Device-3	11.1	765	0.51	4.33%
Device-4	12.2	775	0.52	4.91%
Device-5	14	790	0.52	5.75%
Device-6	12.6	780	0.51	5.01%