# Supporting Information

# Self-Assembled Molecular Network with Waterwheel-like Architecture: Experimental and Theoretical Evaluation toward Electron Transport Capabilities for Optoelectronic Devices

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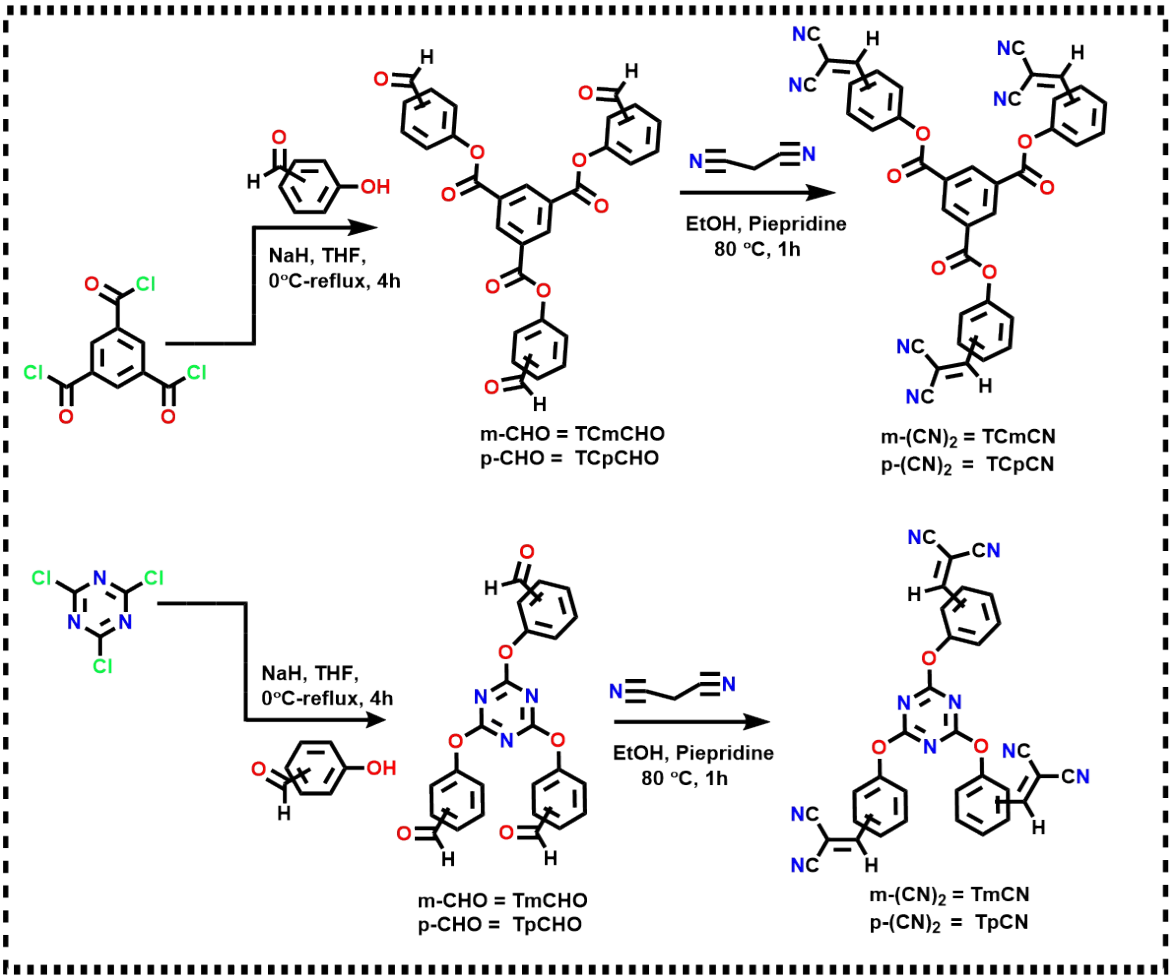
**General**

UV-vis absorption spectra were recorded on a Shimadzu UV-2450 spectrophotometer. Molecules were characterized by NMR spectroscopy on a Jeol EXC NMR spectrometer. Cyclic Voltammetry was performed on the Metrohm Auto lab instrument. The DSC analyses of the three compounds **TCpCN**, **TCmCN**, and **TmCN** were carried out on Mettler Toledo DSC 821 instruments under a nitrogen atmosphere at a scan rate of 10 °C/Min. The DSC analysis of **TpCN** was carried out on a Mettler Toledo DSC 1/700 instrument at a scan rate of 10 °C/min. The TGA analysis of the three compounds **TCpCN**, **TCmCN**, and **TmCN** was investigated on Shimadzu DTG-60 instrument at a heating scan rate of 10 °C/Min. Further, the TGA analysis of **TpCN** was carried out on the NETZSCH STA448 F1 JUPITOR instrument under a nitrogen atmosphere with a scan rate of 10 °C/min. Diffraction study for **TmCN** was done on Agilent Technologies X-ray diffractometer. For the UV-vis studies, 1mg/1ml stock solution was prepared in DCM solvent and experiments were done with 5μM concentration of the solution. The mentioned melting points were measured either from DSC studies or using melting point apparatus (Stone, Staffordshire, ST15 0SA, UK).




**Scheme S1**. Synthetic route for **TCpCN**, **TCmCN**, **TpCN,** and **TmCN**.

## 1. Experimental Section

*Procedure for the synthesis of* ***TCpCHO*** *[1]*

4-Hydroxy benzaldehyde (3.6 eq.) and NaH (3.6 eq.) were added to an anhydrous THF (50 mL) in a round bottom flask at 0°C temperature. Then the reaction mixture was allowed to stirr for 20 minutes at the same temperature. After that benzene-1,3,5-tricarbonyl trichloride (1 eq.) in 10 ml THF was added dropwise to the reaction mixture. Then the mixture was refluxed for an additional 4h. After that, the reaction mixture was allowed to come at room temperature and poured into water (50 mL), and extracted with the ethyl acetate. The

resulting crude product was purified by column chromatography (30% ethyl acetate-hexane). Light yellow solid. Yield 60%; $^{1}$H NMR (500 MHz, DMSO-*d6*): 10.06 (s, 3H), 9.07 (s, 3H), 8.06 (d, *J* = 8.95 Hz, 6H), 7.64 (d, *J* = 8.95 Hz, 6H); $^{13}$C NMR (125 MHz, DMSO-*d6*):191.41, 162.13, 154.51, 135.02, 134.14, 130.74, 130.45, 122.32.

*Procedure for the synthesis of* ***TCmCHO***

3-Hydroxy benzaldehyde (3.6 eq.) and NaH (3.6 eq.) were added to anhydrous THF (50 mL) in a round bottom flask at 0°C temperature. Then the reaction mixture was allowed to stirr for 20 minutes at the same temperature. After that, benzene-1,3,5-tricarbonyl trichloride (1 eq.) in 10 ml THF was added dropwise to the reaction mixture. Then the mixture was refluxed for an additional 4h. After that, the reaction mixture was allowed to come at the room temperature and poured into water (50 mL) and extracted with the ethyl acetate. The resulting crude product was purified by column chromatography (30% ethyl acetate-hexane). Light yellow solid. Yield 60%; $^{1}$H NMR (500 MHz, DMSO-*d6*): 10.06 (s, 3H), 9.07 (s, 3H), 7.91-7.92 (m, 6H), 7.74 (d, *J* = 4.85 Hz, 6H); $^{13}$C NMR (125 MHz, DMSO-*d6*):192.08, 162.68, 150.75, 137.60, 135.14, 130.61, 130.49, 127.80, 127.65, 121.88.

*Procedure for the synthesis of* ***TpCHO*** *[2]*

4-Hydroxy benzaldehyde (3.6 eq.) and NaH (3.6 eq.) were added to anhydrous THF (50 mL) in a round bottom flask at 0°C temperature. Then the reaction mixture was allowed to stirr for 20 minutes at the same temperature. After that, cyanuric chloride (1eq.) in 10 ml THF was added dropwise to the reaction mixture. Then the mixture was refluxed for an additional 4h. After that, the reaction mixture was allowed to come at room temperature and poured into water (50 mL), and extracted with the ethyl acetate. The resulting crude product was purified by column chromatography (37% ethyl acetate-hexane). White solid. Yield 55%; $^{1}$H NMR

(500 MHz, $CDCl_3$): 9.99 (s, 3H), 7.91 (d, *J* = 6.85 Hz, 6H), 7.32 (d, *J* = 7.55 Hz, 6H); $^{13}C$ NMR (125 MHz, $CDCl_3$): 190.58, 173.10, 155.56, 134.33, 131.25, 122.11.

*Procedure for the synthesis of* ***TmCHO*** [3]

3-Hydroxy benzaldehyde (3.6 eq.) and NaH (3.6 eq.) were added to anhydrous THF (50 mL) in a round bottom flask at 0°C temperature. Then the reaction mixture was allowed to stirr for 20 minutes at the same temperature. After that, cyanuric chloride (1 eq.) in 10 ml THF was added dropwise to the reaction mixture. Then the mixture was refluxed for an additional 4h. After that, the reaction mixture was allowed to come at room temperature and poured into water (50 mL) and extracted with ethyl acetate. The resulting crude product was purified by column chromatography (37% ethyl acetate-hexane). White solid. Yield 65%; $^1H$ NMR (500 MHz, DMSO-*d6*): 9.97 (s, 3H), 7.80-7.59 (m, 12H); $^{13}C$ NMR (125 MHz, DMSO-*d6*): 192.19, 172.91, 151.77, 137.57, 130.57, 127.63, 127.54, 121.65.

*Procedure for the synthesis of* ***TCpCN***

**TCpCHO** (1 eq.) and malononitrile (3.6 eq.) were added to EtOH (20 ml) in a round bottom flask at room temperature. After that, one drop of piperidine was added to the reaction mixture. Then the mixture was refluxed for 1 h at 80 °C. The yellow precipitate was filtered and washed with cold EtOH (3 times). Yellow solid. Yield 65%; $^1H$ NMR (500 MHz, DMSO-*d6*): 9.05 (s, 3H), 8.54 (s, 3H), 8.09 (d, *J* = 8.90 Hz, 6H), 7.65 (d, *J* = 8.25 Hz, 6H); $^{13}C$ NMR (125 MHz, DMSO-*d6*): 162.00, 160.01, 154.07, 135.11, 131.90, 130.36, 129.10, 122.65, 113.64, 112.67, 81.50; HRMS (ESI): Calculated for $C_{39}H_{18}N_6O_6Na$ $[M+Na]^+$: 689.1187, found: 689.1183; M.P: 198 °C (measured from DSC).

*Procedure for the synthesis of* ***TCmCN***

**TCmCHO** (1 eq.) and malononitrile (3.6 eq.) were added to EtOH (20 ml) in a round bottom flask at room temperature. After that, one drop of piperidine was added to the reaction mixture. Then the mixture was refluxed for 1 h at 80 °C. The yellow precipitate was filtered and washed with cold EtOH (3 times). Yellow solid. Yield 70%; $^{1}$H NMR (500 MHz, DMSO-*d6*): 9.05 (s, 3H), 8.59 (s, 3H), 7.92-7.95 (m, 6H), 7.76-7.73 (m, 6H);$^{13}$C NMR (125 MHz, DMSO-*d6*): 162.47, 160.15, 150.45, 135.21, 132.49, 130.74, 130.47, 128.09, 127.50, 123.27, 113.70, 112.66, 82.97. HRMS (ESI): Calculated for $C_{39}H_{18}N_6O_6Na$ $[M+Na]^+$: 689.1187, found: 689.1184; M.P: 195 °C (measured from DSC).

*Procedure for the synthesis of* ***TmCN***

**TmCHO** (1 eq.) and malononitrile (3.6 eq.) were added to EtOH (20 ml) in a round bottom flask at room temperature. After that, one drop of piperidine was added to the reaction mixture. Then the mixture was refluxed for 1 h at 80 °C. The white precipitate was filtered and washed with cold EtOH (3 times). White solid. Yield 65%; $^{1}$H NMR (500 MHz, DMSO-*d6*): 8.52 (s, 3H), 7.85 (d, $J$ = 7.60 Hz, 3H), 7.72 (s, 3H), 7.66 (t, $J$ = 8.25 Hz, 3H), 7.60-7.58 (m, 3H); $^{13}$C NMR (125 MHz, DMSO-*d6*): 172.83, 160.16, 151.42, 132.55, 130.84, 128.12, 127.61, 122.93, 113.93, 112.88, 83.09; HRMS (ESI): Calculated for $C_{33}H_{16}N_9O_3$ $[M+H]^+$: 586.1376, found: 586.1376; M.P: 211 °C (measured from DSC).

*Procedure for the synthesis of* ***TpCN***

**TpCHO** (1 eq.) and malononitrile (3.6 eq.) were added to EtOH (20 ml) in a round bottom flask at room temperature. After that, one drop of piperidine was added to the reaction mixture. Then the mixture was refluxed for 1 h at 80 °C. The white precipitate was filtered and washed with cold EtOH (3 times). White solid. Yield 60%; $^{1}$H NMR (500 MHz, DMSO-*d6*): 8.53 (s, 3H), 7.98 (d, $J$ = 8.25 Hz, 6H), 7.51 (d, $J$ = 8.25 Hz, 6H); $^{13}$C NMR (125 MHz, DMSO-*d6*): 172.62, 160.32, 155.09, 132.25, 129.20, 122.75, 114.14, 113.12, 81.64; HRMS

(ESI): Calculated for $C_{33}H_{16}N_9O_3$ $[M+H]^+$: 586.1376, found: 586.1371. M.P: 211 °C (measured using melting point apparatus).

**Electron-only device fabrication**

Electron-only devices were fabricated with **TmCN**, **TpCN**, **TCpCN**, and **TCmCN** as electron transport layer (ETL). The fabricated devices were comprised of ~125 nm ITO anode layer, ~10 nm ETL, ~1 nm LiF as EIL, and ~100 nm Al cathode layer.

The ITO-coated glass substrate was washed in acetone at 45°C for 30 minutes, 2-propanol at 60°C for 60 minutes (both in an ultrasonic bath), treated in an ozone environment under UV light for 15 minutes, and then used to fabricate the device. After cleaning, the substrate was put into a glove box filled with nitrogen to spin-coat the ETL. Tetrahydrofuran (THF) was used to dissolve the **TmCN**, **TpCN**, **TCpCN**, and **TCmCN** before they were spin-coated onto the specimen at 2,500 rpm for 20 seconds to coat the ETL. After the coating of ETL, the specimen was shifted to the high-vacuum chamber, which has a base pressure of ~7 X $10^{-6}$ torr. Thermal evaporation was used to successively deposit the subsequent layers (LiF and Al, respectively). The device's current density was measured using a Keithley 2400 electrometer.

## 2. Computed UV spectra with TD-DFT

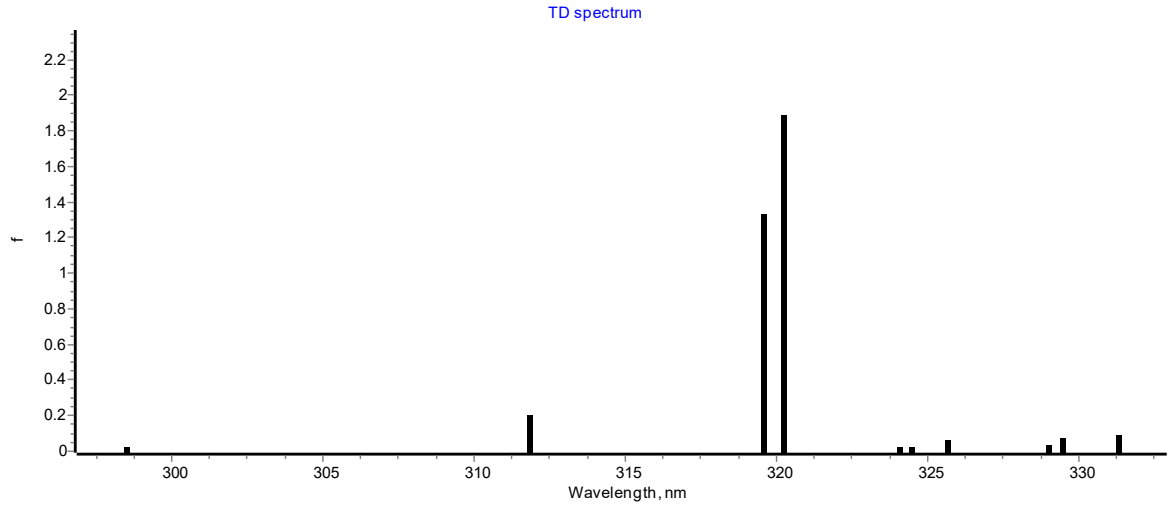



**Figure S1.** Computed UV spectrum with TD-DFT of **TCpCN**

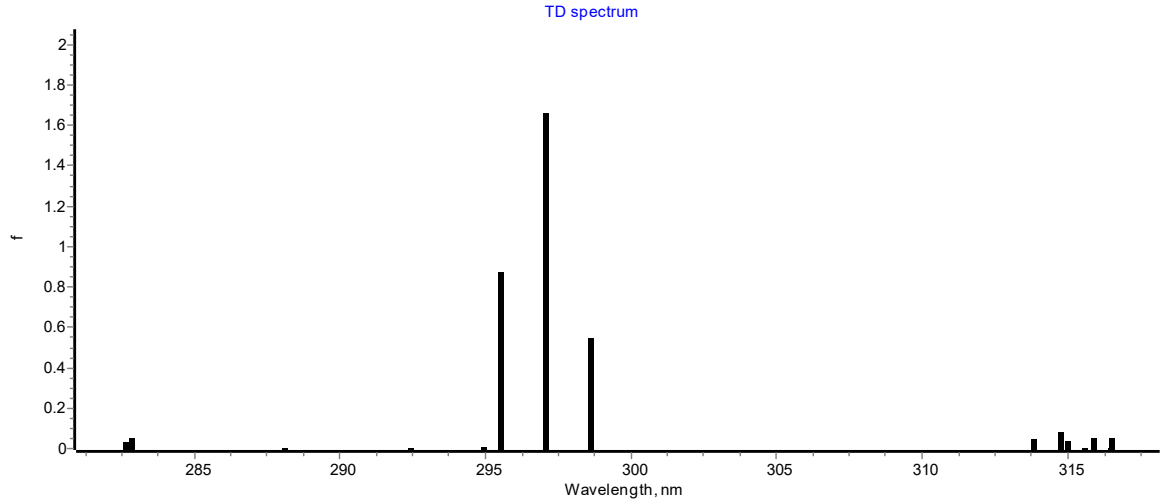



**Fig. S2.** Computed UV spectrum with TD-DFT of **TCmCN**

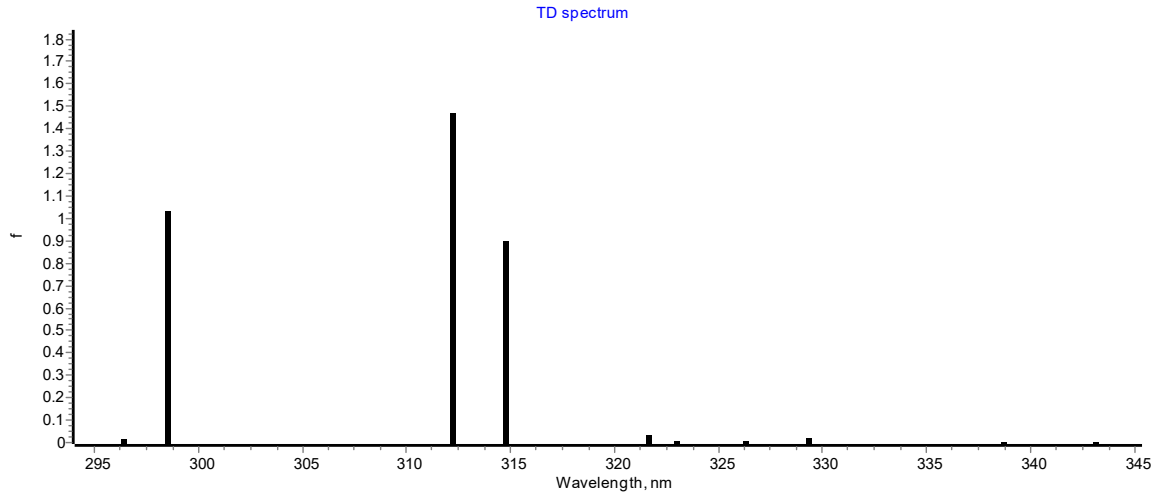



**Fig. S3.** Computed UV spectrum with TD-DFT of **TpCN**

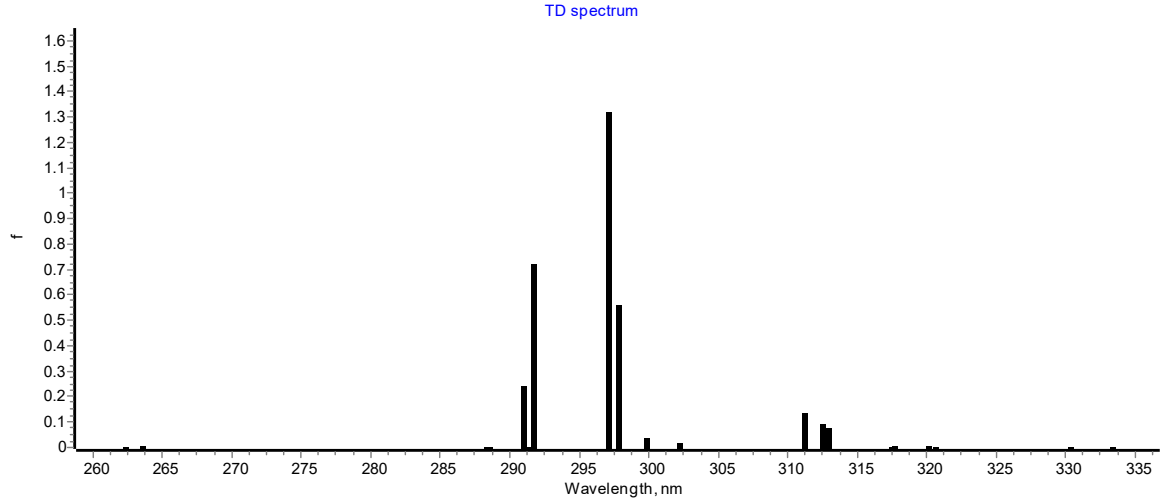



**Fig. S4.** Computed UV spectrum with TD-DFT of **TmCN**

**Table S1**. TD-DFT analysis of **TCpCN**. Oscillator strengths (f) and coefficients >0.20 are tabulated here.

| S.No. | Transition Energy (eV)/Wavelength (nm) | MOs involved Coefficients | f |
|---|---|---|---|
| 1. | 3.8718/320.22 | 171 -> 173 0.24592<br>171 -> 174 0.39264 | 1.8941 |
| 2. | 3.8802 / 319.53 | 169 -> 172 0.24332<br>170 -> 174 0.47321 | 1.3298 |
| 3. | 3.9763 / 311.81 | 169 -> 174 0.44488<br>170 -> 172 0.29417 | 0.2020 |

**Table S2**. TD-DFT analysis of **TCmCN**. Oscillator strengths (f) and coefficients >0.20 are tabulated here.

| S.No. | Transition Energy (eV)/Wavelength (nm) | MOs involved | Coefficients | f |
|---|---|---|---|---|
| 1. | 4.1521/ 298.61 | 169 -> 172 | 0.44335 | 0.5446 |
| 2. | 4.1740 / 297.04 | 169 -> 172<br>170 -> 173 | 0.30382<br>0.36154 | 1.6614 |
| 3. | 4.1962 / 295.47 | 169 -> 172<br>170 -> 173<br>171 -> 174 | 0.23620<br>0.24687<br>0.45325 | 0.8726 |

**Table S3**. TD-DFT analysis of **TpCN**. Oscillator strengths (f) and coefficients >0.20 are tabulated here.

| S.No. | Transition Energy (eV)/Wavelength (nm) | MOs involved Coefficients | f |
|---|---|---|---|
| 1. | 3.9393 / 314.74 | 149 ->152 0.51287 | 0.8963 |
| 2. | 3.9720 / 312.14 | 150 ->153 0.45177 | 1.4733 |
| 3. | 4.1543 / 298.45 | 148 ->151 0.42844<br>149 ->152 0.32161<br>150 ->153 0.25762 | 1.0298 |

**Table S4**. TD-DFT analysis of **TmCN.** Oscillator strengths (f) and coefficients >0.20 are tabulated here.

| S.No. | Transition Energy(eV)/Wavelength (nm) | MOs involved | Coefficients | f |
|---|---|---|---|---|
| 1. | 4.1638 / 297.77 | 149 ->153<br>150 ->152 | 0.37561<br>0.30735 | 0.5567 |
| 2. | 4.1751 / 296.96 | 148 ->151 | 0.35407 | 1.3203 |
| 3. | 4.2519 / 291.60 | 147 ->152<br>148 ->151<br>149 ->152<br>150 ->153 | 0.22479<br>0.33137<br>0.21316<br>0.26469 | 0.7205 |

**Table S5.** Charge transfer distances and magnitude of charge transfer for all the compounds computed using wB97XD functional and 6-311G(d,p) basis set.

| Molecule | $S_1$ | | $S_2$ | | $T_1$ | | $T_2$ | |
|---|---|---|---|---|---|---|---|---|
| | $D_{CT}$ (Å) | $Q_{CT}$ | $D_{CT}$ (Å) | $Q_{CT}$ | $D_{CT}$ (Å) | $Q_{CT}$ | $D_{CT}$ (Å) | $Q_{CT}$ |
| **TCmCN** | 1.45 | 0.45 | 1.28 | 0.44 | 0.31 | 0.39 | 0.32 | 0.40 |
| **TCpCN** | 1.00 | 0.45 | 0.44 | 0.45 | 0.23 | 0.41 | 0.22 | 0.41 |
| **TpCN** | 1.21 | 0.43 | 0.68 | 0.43 | 0.33 | 0.39 | 0.32 | 0.39 |
| **TmCN** | 0.92 | 0.45 | 1.07 | 0.44 | 0.25 | 0.41 | 0.25 | 0.41 |

### 3. Reorganization energy:

The intramolecular electron/hole reorganization energies are very crucial to examine the electron transfer ability of the developed molecules, which were calculated using Nelsen's four-point formula[4-8] equations as depicted below:

$$\lambda_h = \lambda_1 + \lambda_2$$

$$\lambda_1 = E^+(M^0) - E^+(M^+)$$

$$\lambda_2 = E^0(M^+) - E^0(M^0)$$

$$\lambda_e = \lambda_3 + \lambda_4$$

$$\lambda_3 = E^-(M^0) - E^-(M^-)$$

$$\lambda_4 = E^0(M^-) - E^0(M^0)$$

where [4-8]

**(a)** $E^+(M^+)$ belongs to the energy of the cation set at its optimized geometry of $M^+$ and $E^0(M^0)$ belongs to the energy of the neutral set.

**(b)** $E^-(M^-)$ belongs to the energy of anion set and $E^0(M^-)$ belongs to the energy of neutral sets at its optimized geometry $M^-$.

**(c)** $E^-(M^0)$ belongs to the energies of the anion set at its optimized geometry $M^0$ and $E^0(M^+)$ is the energy of the neutral set at its optimized geometries of $M^+$.

**(d)** $E^+(M^0)$ is the energy of the cation set at the optimized geometries of $M^0$.

## 4. Cyclic Voltammetry

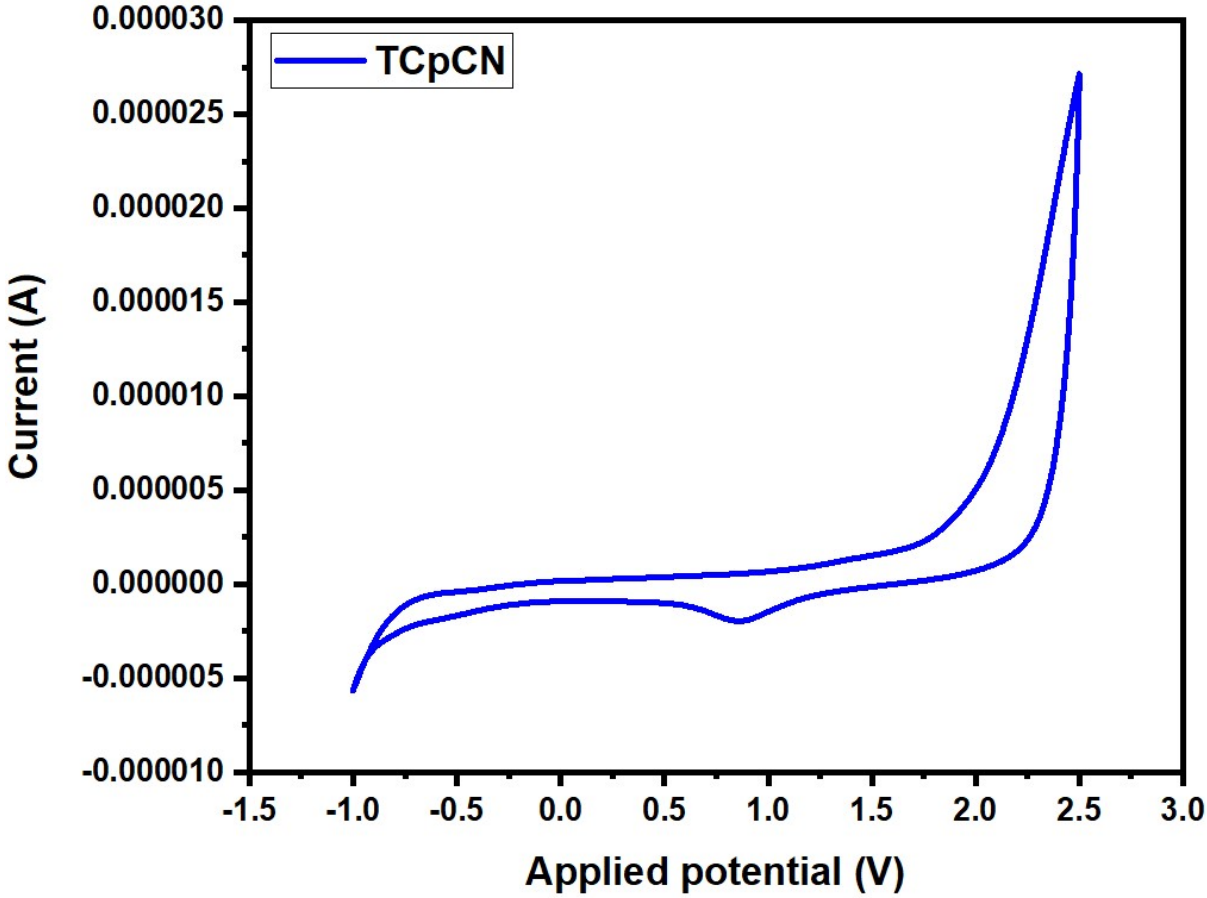



**Fig. S5.** Cyclic voltammetry graph of **TCpCN** (Ag/AgCl reference electrode, 0.1 M tetrabutylammonium hexafluorophosphate (electrolyte)).

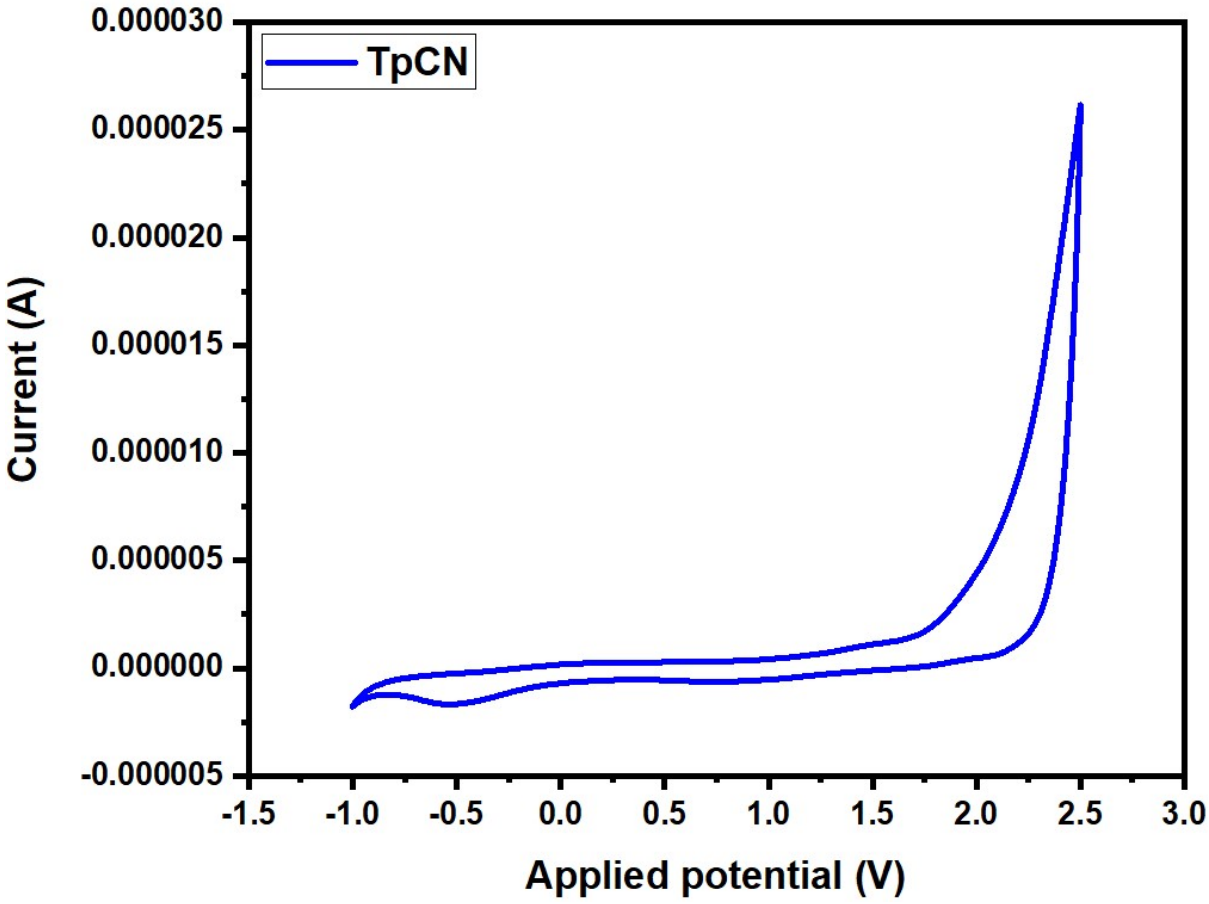



**Fig. S6.** Cyclic voltammetry graph of **TpCN** (Ag/AgCl reference electrode, 0.1 M tetrabutylammonium hexafluorophosphate (electrolyte)).

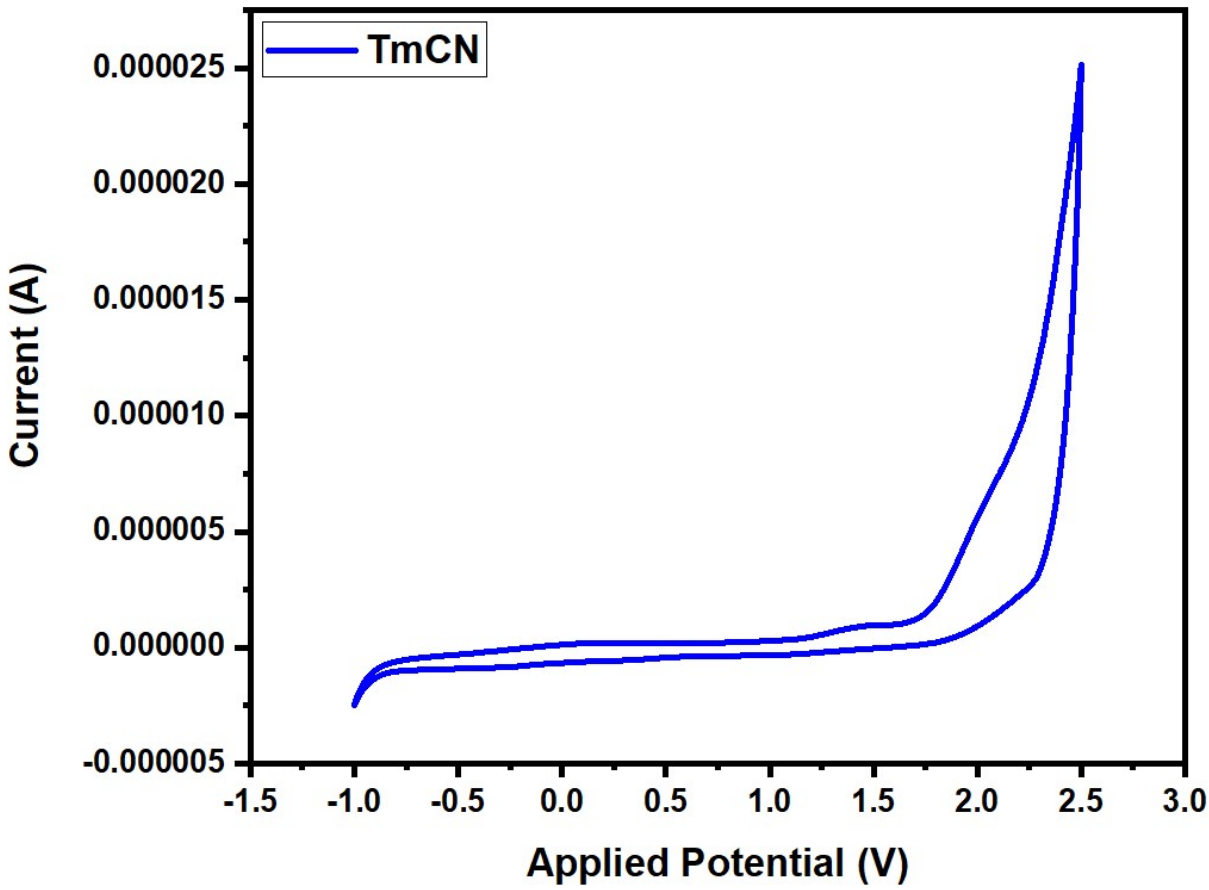



**Fig. S7.** Cyclic voltammetry graph of **TmCN** (Ag/AgCl reference electrode, 0.1 M tetrabutylammonium hexafluorophosphate (electrolyte)).

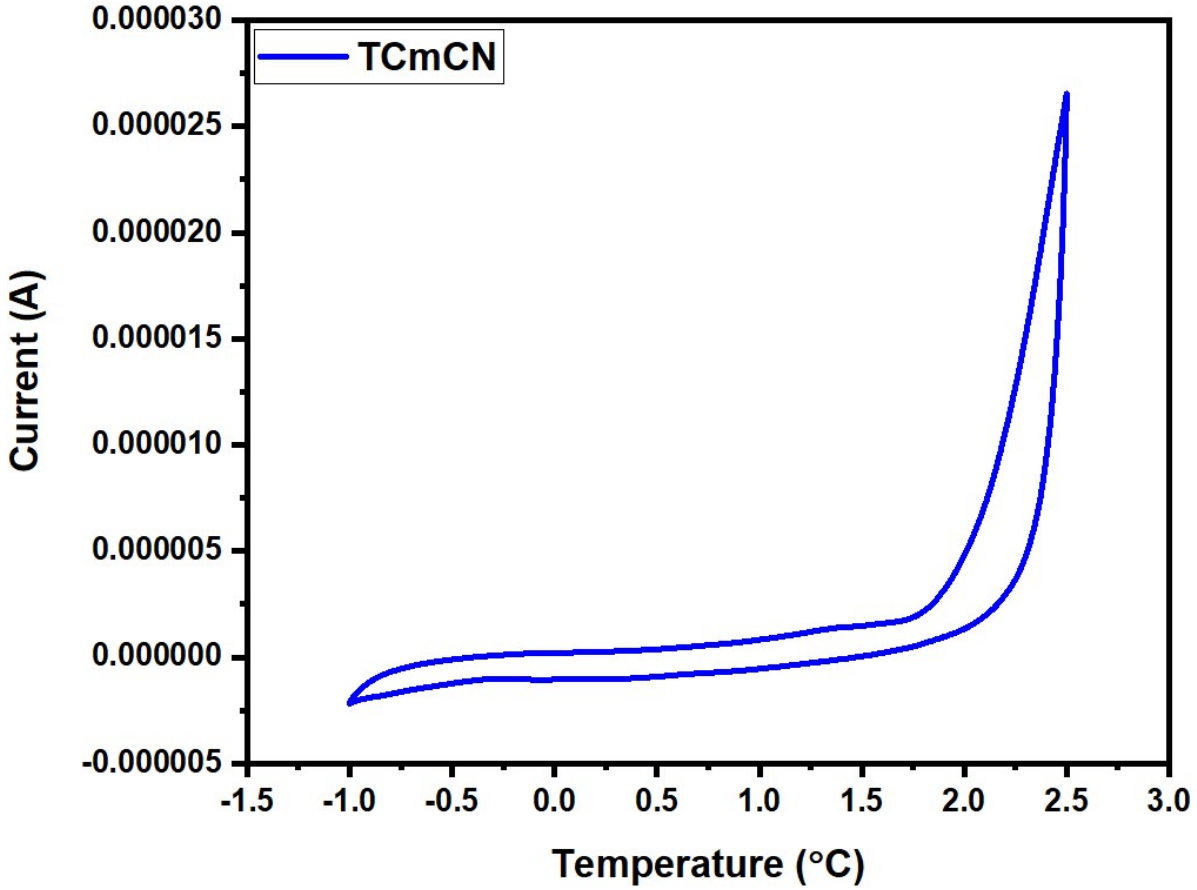



**Fig. S8.** Cyclic voltammetry graph of **TCmCN** (Ag/AgCl reference electrode, 0.1 M tetrabutylammonium hexafluorophosphate (electrolyte)).

## 5. TGA analysis

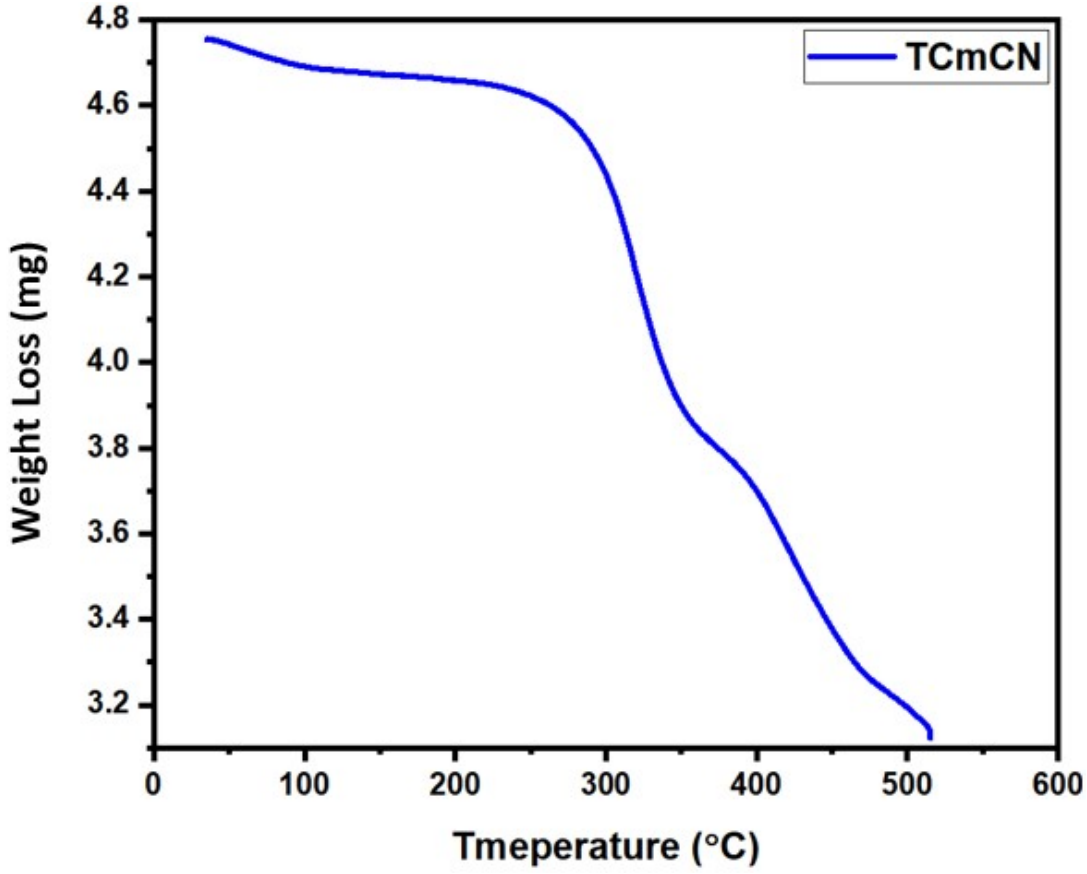



**Fig. S9.** TGA graph of **TCmCN**.

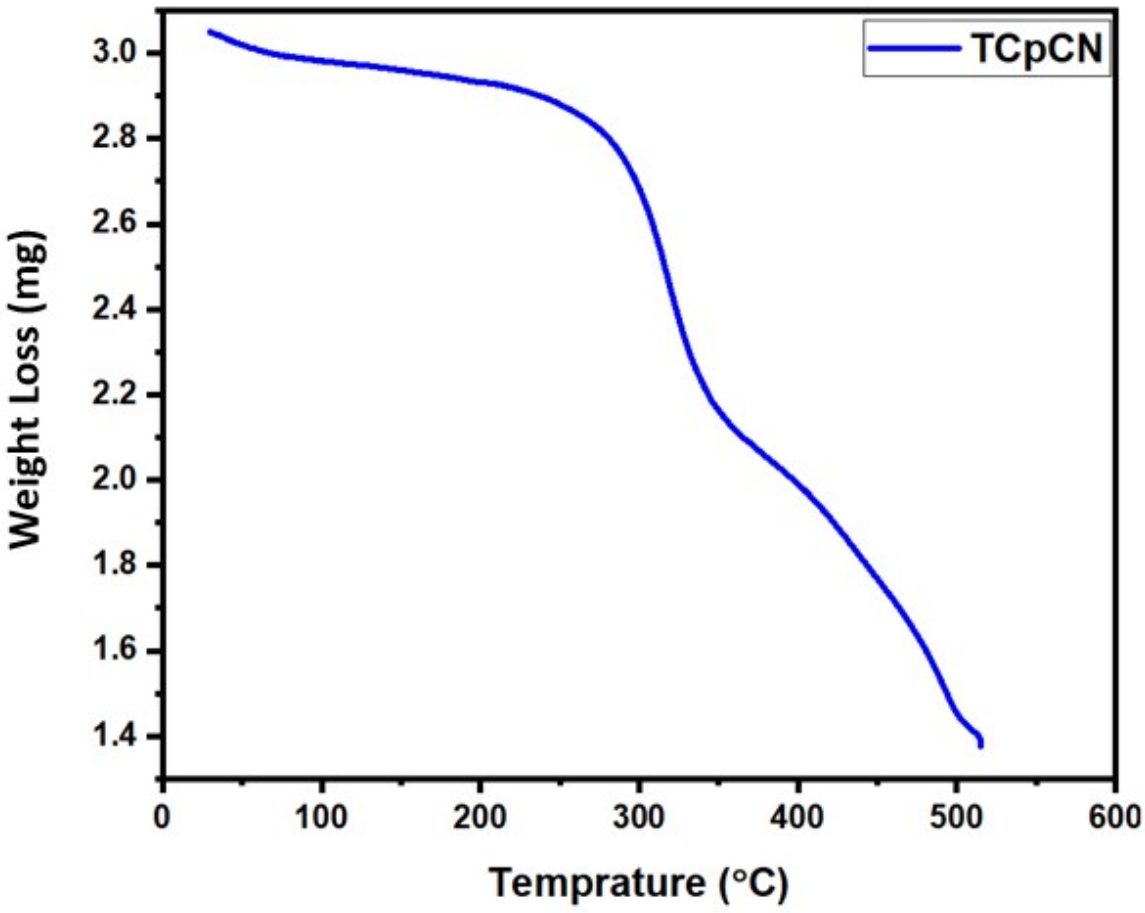



**Fig. S10.** TGA graph of **TCpCN**.

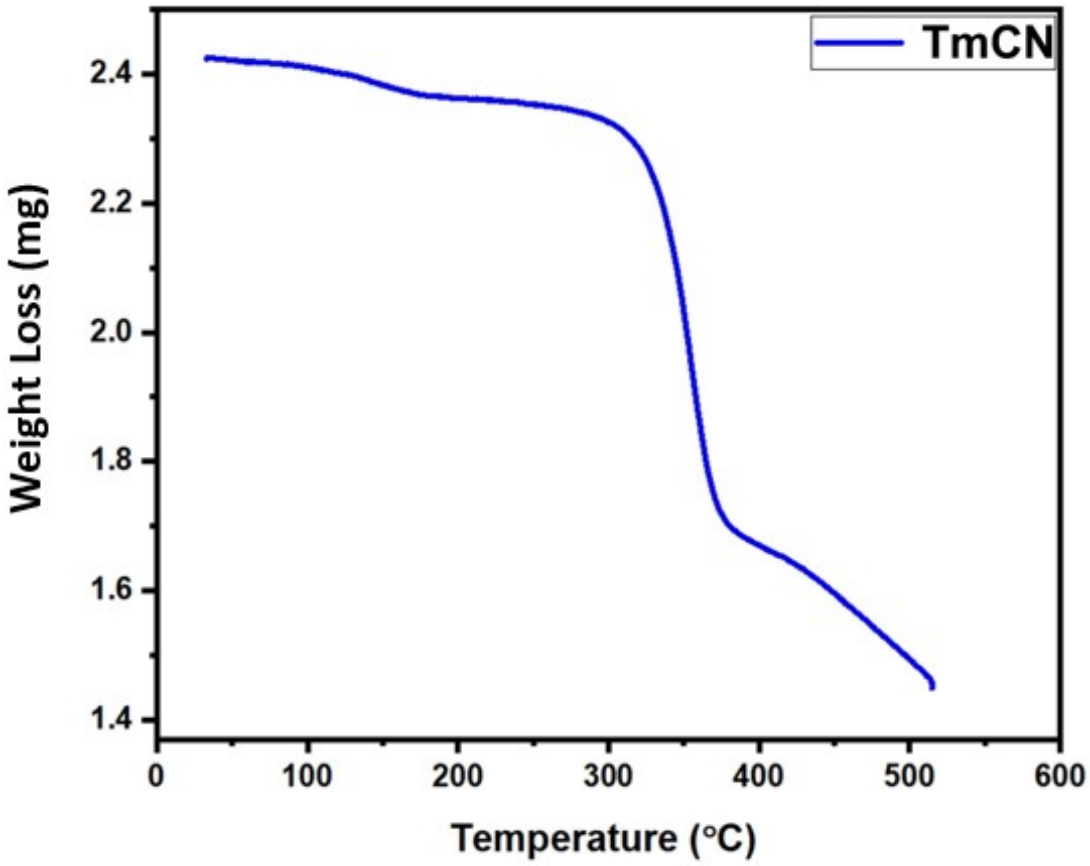



**Fig. S11.** TGA graph of **TmCN**.

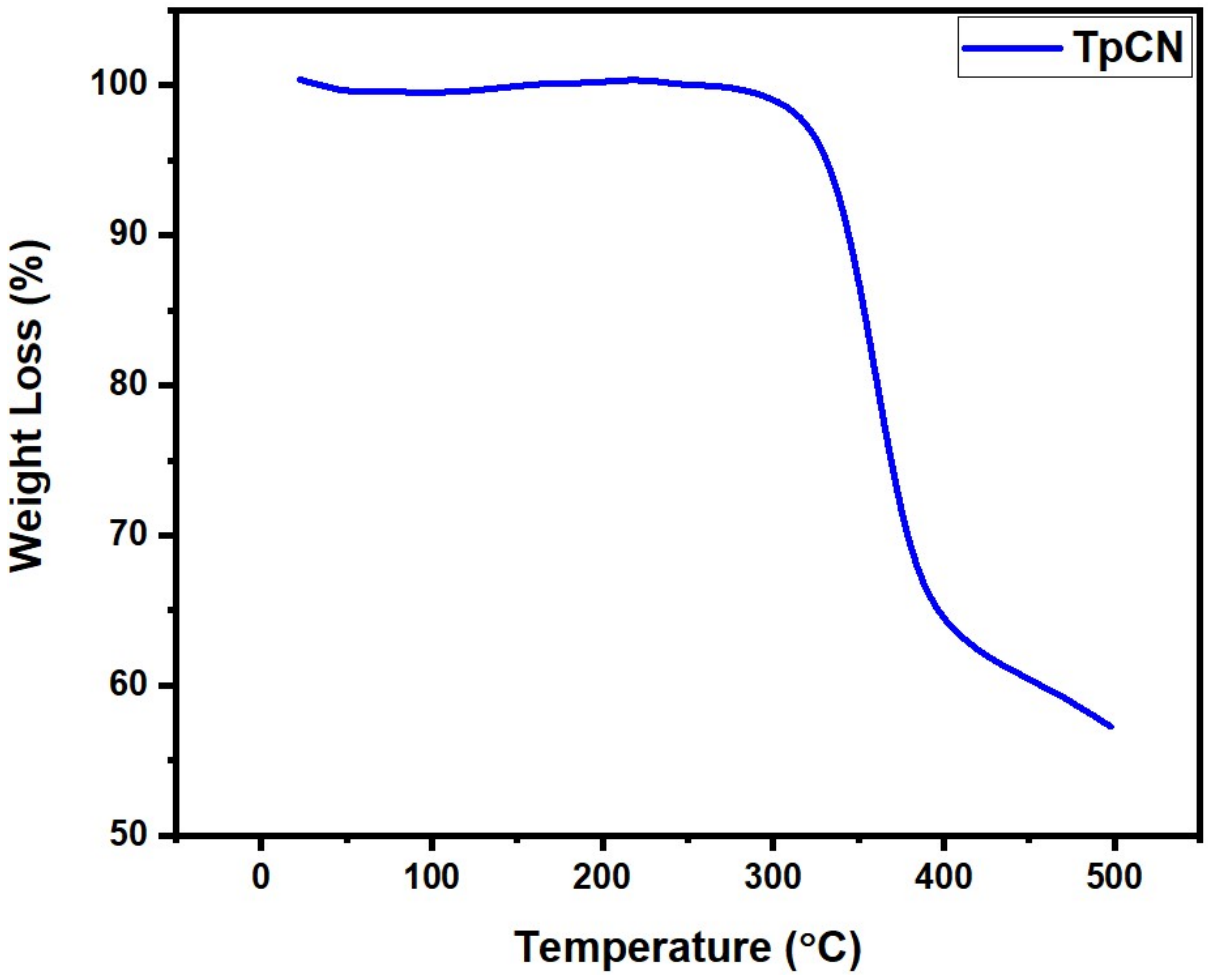



**Fig. S12.** TGA graph of **TpCN**.

## 6. DSC analysis:

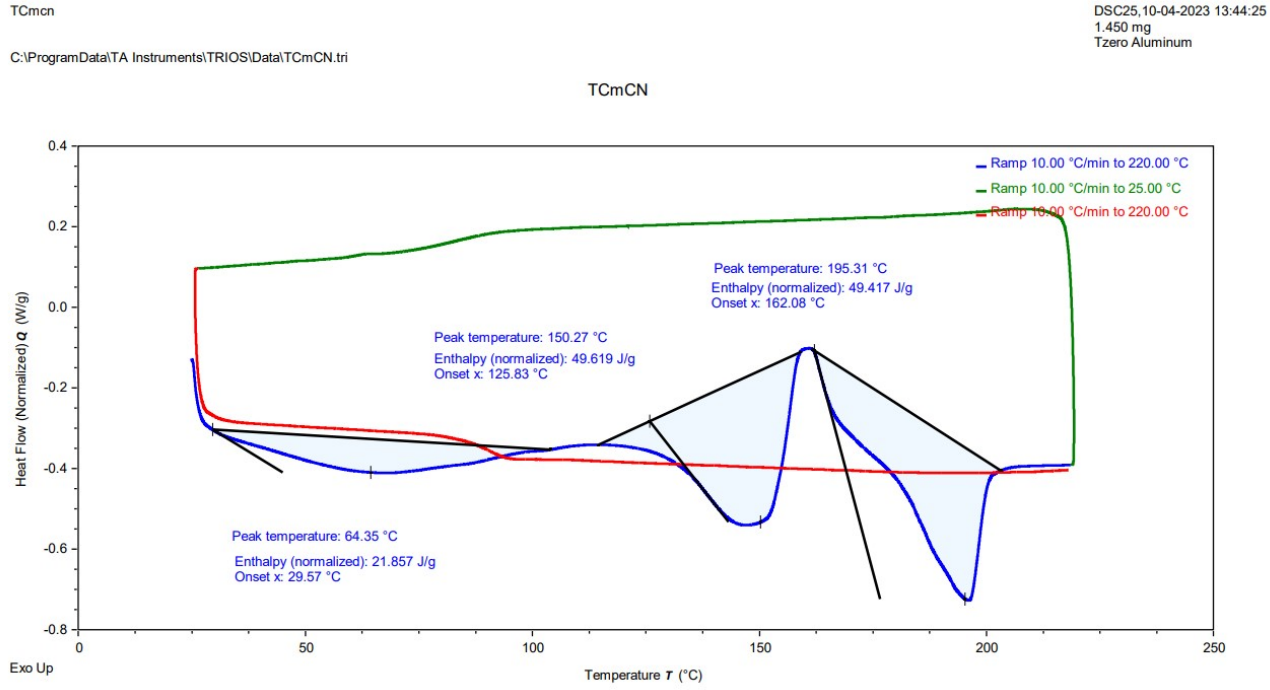



**Fig. S13.** DSC graph of **TCmCN**.

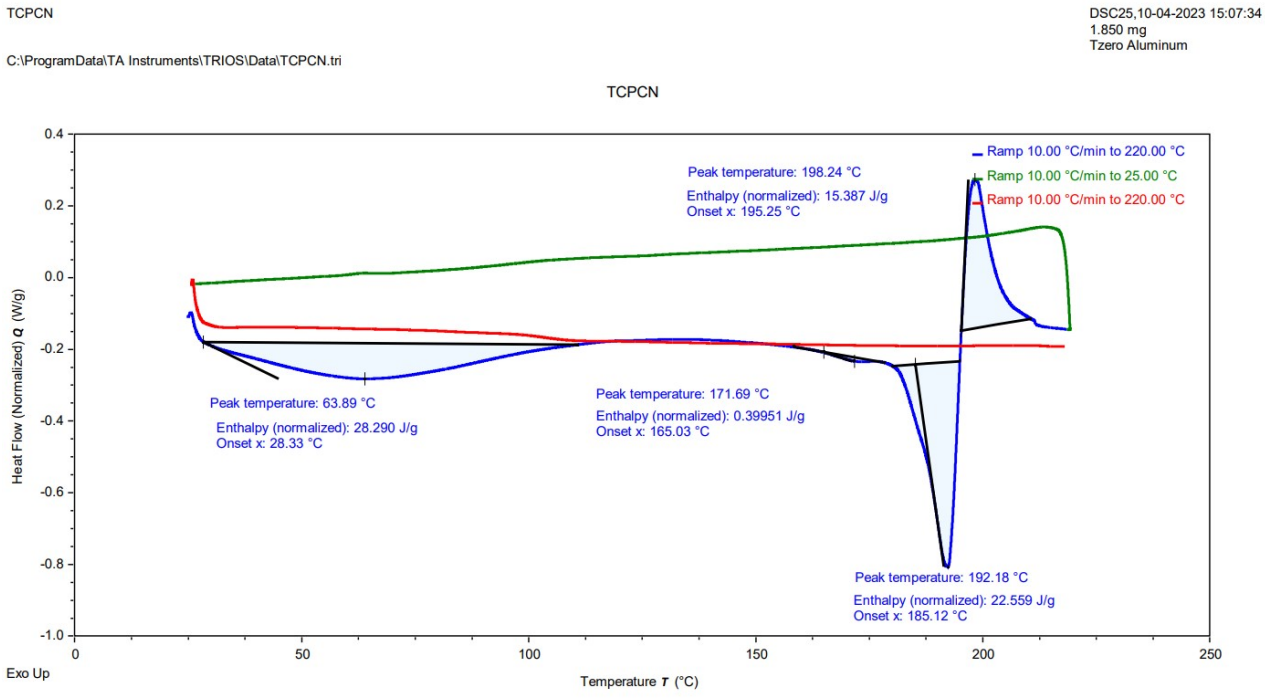



**Fig. S14.** DSC graph of **TCpCN**.

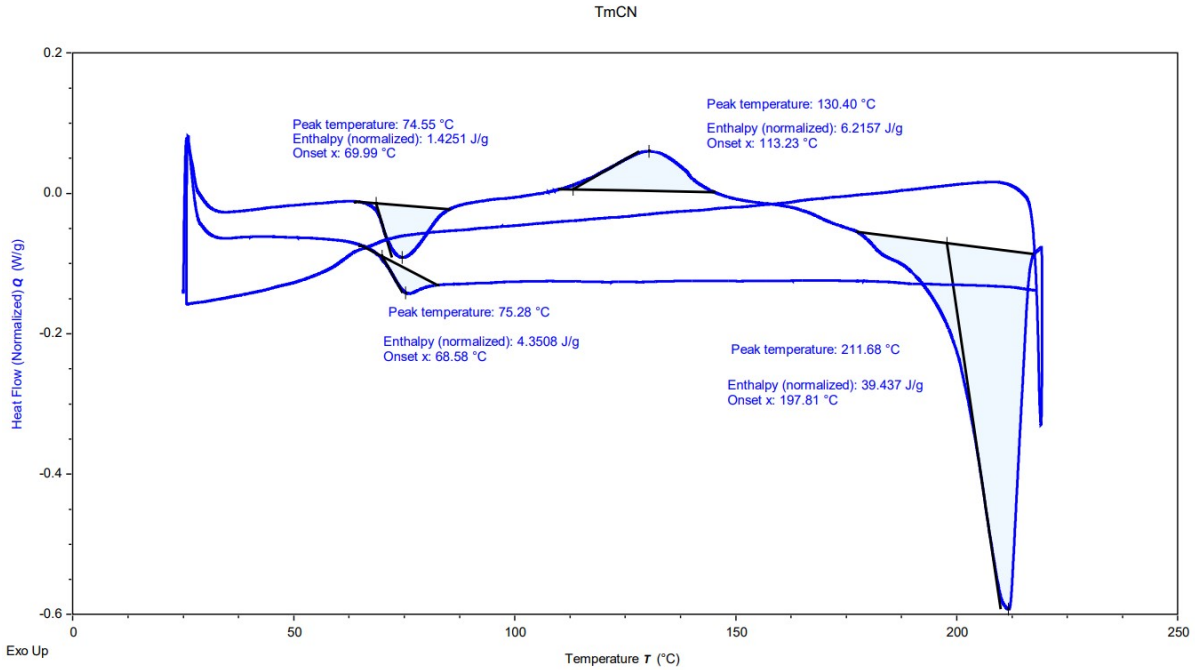



**Fig. S15.** DSC graph of **TmCN**.

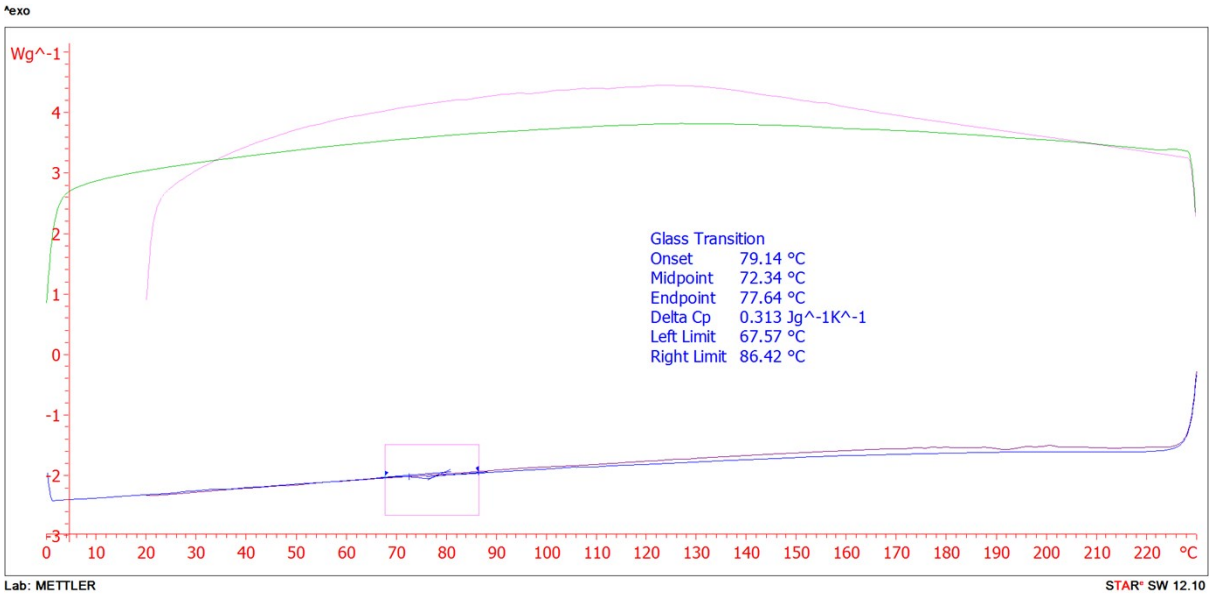
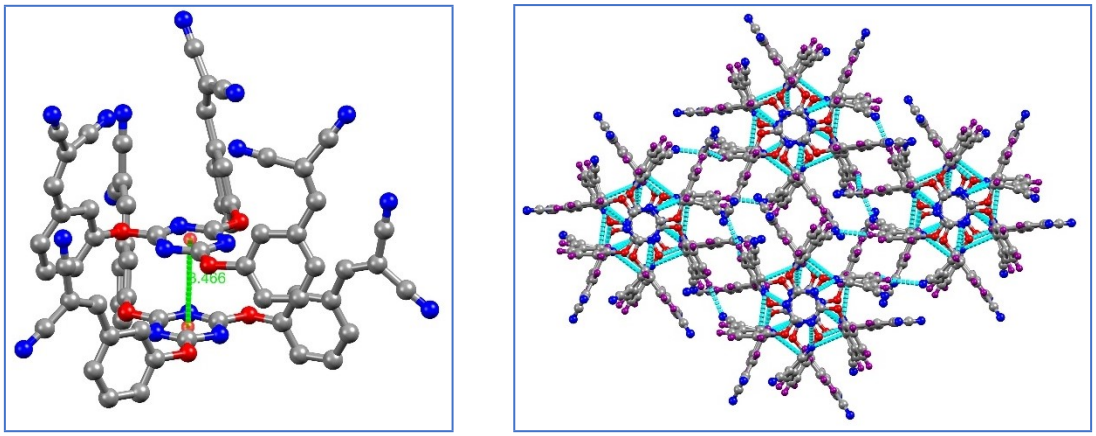



**Fig. S16.** DSC graph of **TpCN**.

## 7. Single Crystal analysis.

| Table S6 Crystal data and structure refinement for TmCN_Rt_Cu. | |
|---|---|
| Identification code | TmCN_ |
| Empirical formula | $C_{33}H_{15}N_9O_3$ |
| Formula weight | 585.54 |
| Temperature/K | 293(2) |
| Crystal system | trigonal |
| Space group | R-3 |
| a/Å | 23.8757(6) |
| b/Å | 23.8757(6) |
| c/Å | 17.6972(5) |
| α/° | 90 |
| β/° | 90 |
| γ/° | 120 |
| Volume/Å$^3$ | 8736.7(5) |
| Z | 12 |
| $\rho_{calc}$g/cm$^3$ | 1.335 |
| μ/mm$^{-1}$ | 0.748 |
| F(000) | 3600.0 |
| Radiation | Cu Kα (λ = 1.54184) |
| 2Θ range for data collection/° | 6.574 to 134.182 |
| Index ranges | -28 ≤ h ≤ 28, -28 ≤ k ≤ 19, -20 ≤ l ≤ 21 |
| Reflections collected | 9227 |
| Independent reflections | 3327 [$R_{int}$ = 0.0509, $R_{sigma}$ = 0.0465] |
| Data/restraints/parameters | 3327/0/271 |
| Goodness-of-fit on F$^2$ | 0.984 |
| Final R indexes [I>=2σ (I)] | $R_1$ = 0.0682, $wR_2$ = 0.1993 |
| Final R indexes [all data] | $R_1$ = 0.0830, $wR_2$ = 0.2149 |
| CCDC No | 2285795 |

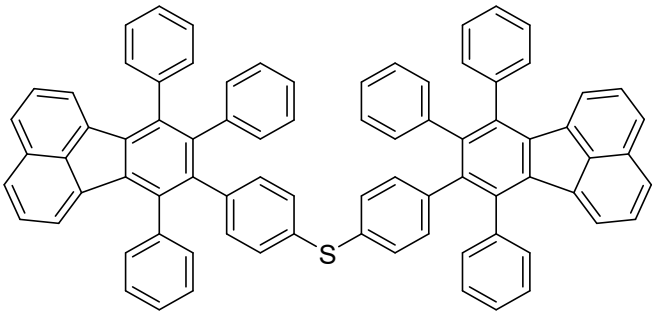
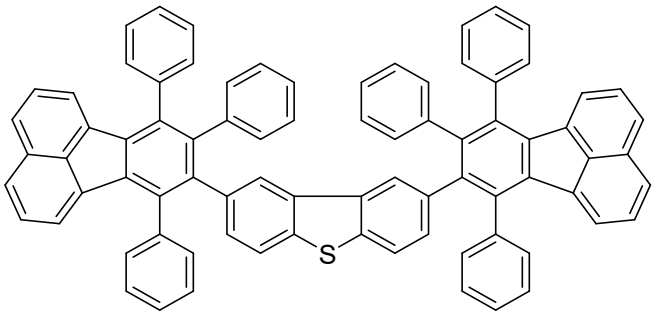
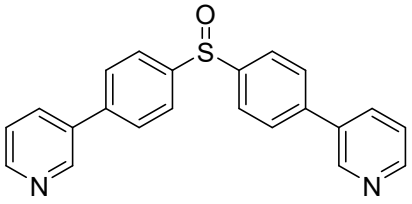


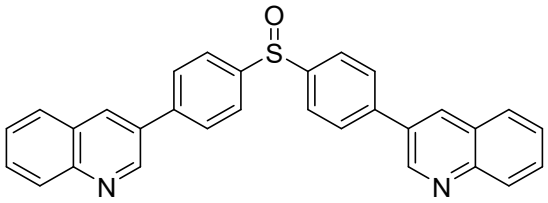
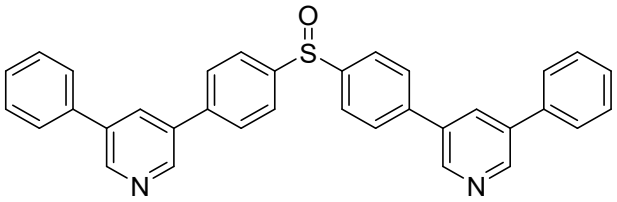
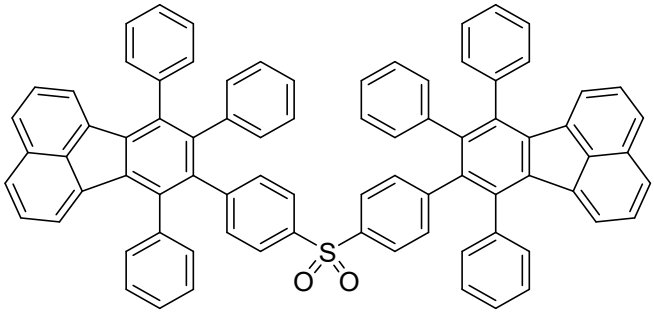
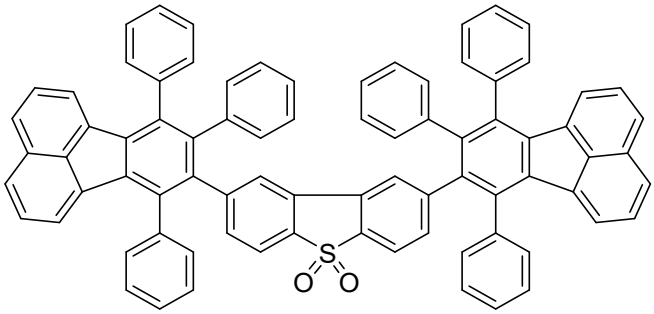

A B

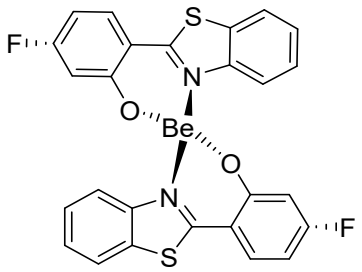
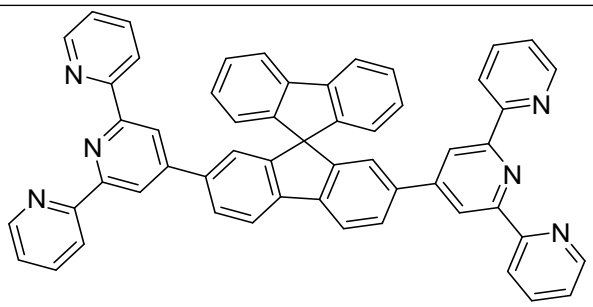
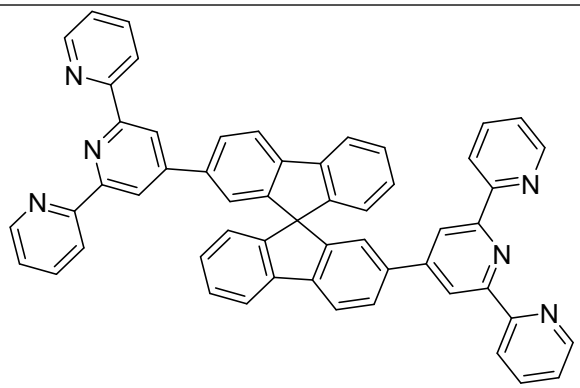
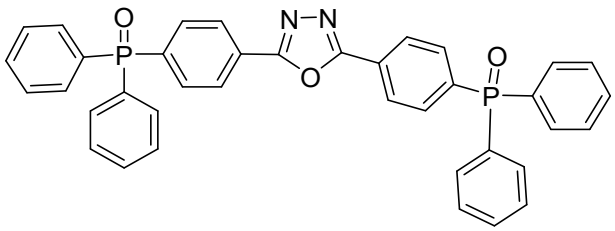
**Fig. S17** (A) π-π interaction between the two **TmCN** molecules. (B) Three-dimensional hydrogen-boned assembly of **TmCN** (the H-bonding interactions are represented as cyan color).

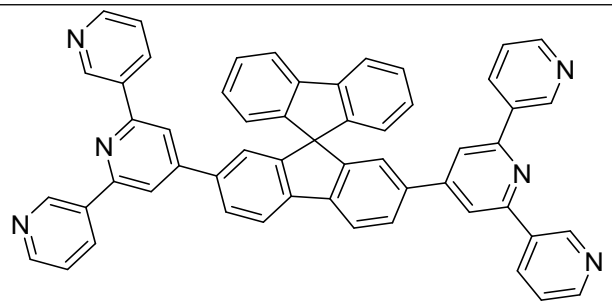
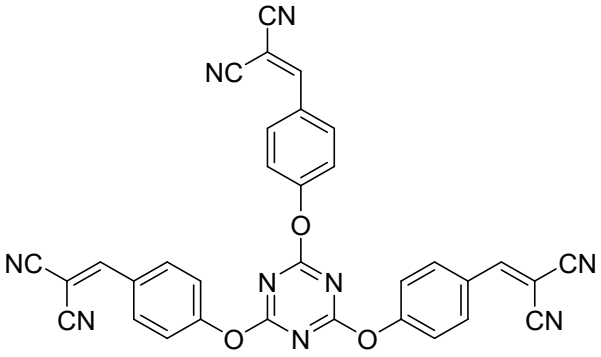
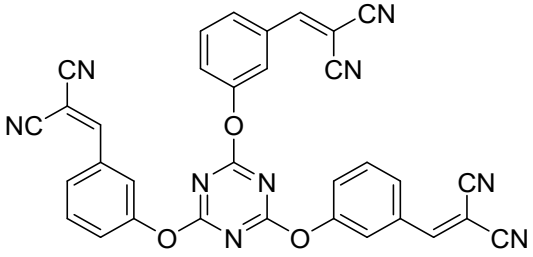
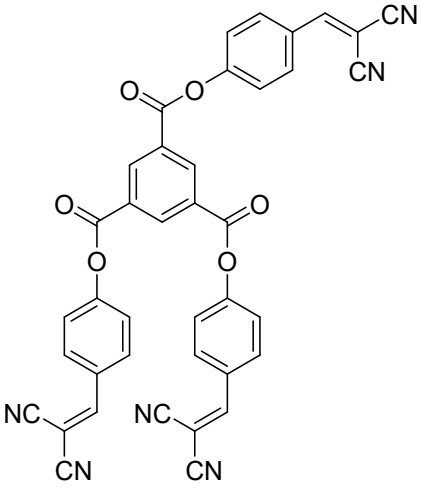
## 8. A comparison of HOMO/LUMO energy levels:

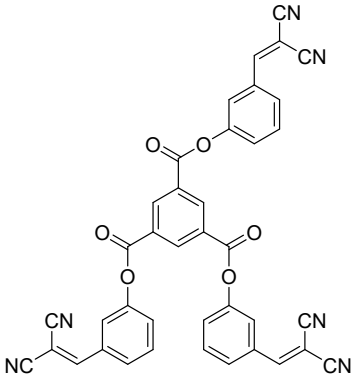
**Table S7.** A comparison between HOMO/LUMO energy levels of some reported and the present materials. (Measured using Ag/AgCl, Pt-wire, and Pt-disc electrodes, and tetrabutylammonium hexafluorophosphate as supporting electrolyte).

| S.No. | Structure of ETM | HOMO/LUMO (eV) | Reference |
|---|---|---|---|
| 1. | TPFDPS  | -6.1/-2.8 | New J. Chem., 2015, 39, 6351-6357 |
| 2. | TPFDBT  | -6.2/-2.8 | New J. Chem., 2015, 39, 6351-6357 |
| 3. |  SPDP | -6.17/-2.57 | J. Mater. Chem. C, 2014,2,10129-10137 |

| | | | |
|---|---|---|---|
| 4 |  <br>SPDQ | -6.32/-2.7 | J. Mater. Chem. C, 2014,2,10129-10137 |
| 5. |  <br>SPPP | -6.16/-2.37 | J. Mater. Chem. C, 2014,2,10129-10137 |
| 6. |  <br>TPFDBSO2 | -6.1/-2.9 | J. Phys. Chem. C 2015, 119, 19297−19304 |
| 7 |  <br>TPFDBTO2 | -6.2/-2.8 | J. Phys. Chem. C 2015, 119, 19297−19304 |

| | | | |
|---|---|---|---|
| 8 |  <br>Be(FBTZ)2 | -6.14/-3.14 | RSC Adv., 2016, 6, 5008-5015 |
| 9 |  <br>27-TPSF | -5.9/-2.7 | J. Mater. Chem. C, 2018, 6, 10276-10283 |
| 10 |  <br>22-TPSF | -5.9/-2.6 | J. Mater. Chem. C, 2018, 6, 10276-10283 |
| 11 |  <br>OXDPPO | -6.42/-2.83 | RSC Adv., 2015, 5, 36568-36574 |

| | | | |
|---|---|---|---|
| 12 |  <br>27-mTPSF | -6.0/-2.5 | J. Mater. Chem. C, 2019, 7, 11581-11587 |
| 13 |  <br>TpCN | -6.15/-2.82 | This work |
| 14 |  <br>TmCN | -6.15/-2.71 | This work |
| 15 |  <br>TCpCN | -6.05/-2.55 | This work |

| | | | |
|---|---|---|---|
| 16. |  <br>TCmCN | -6.15/-2.76 | This work |

**9.** Computed electron reorganization energy comparison with some reported ETMs:[9]

| Table S8 Electron reorganization energies | |
|---|---|
| Compound | Electron reorganization energies |
| Phen | 0.109 |
| 4-PPhen | 0.172 |
| 2-PPhen | 0.106 |
| 2,9-DPPhen | 0.084 |
| 2,4-DPPhen | 0.107 |
| 4,7-DPPhen(Bphen) | 0.120 |
| p-bPPhenB | 0.056 |
| m-bPPhenB | 0.069 |
| TmPyPB | 0.071 |
| **TCpCN** | 0.20 |
| **TCmCN** | 0.25 |
| **TpCN** | 0.20 |
| **TmCN** | 0.22 |

## 10. $^{1}H$ and $^{13}C$ NMR

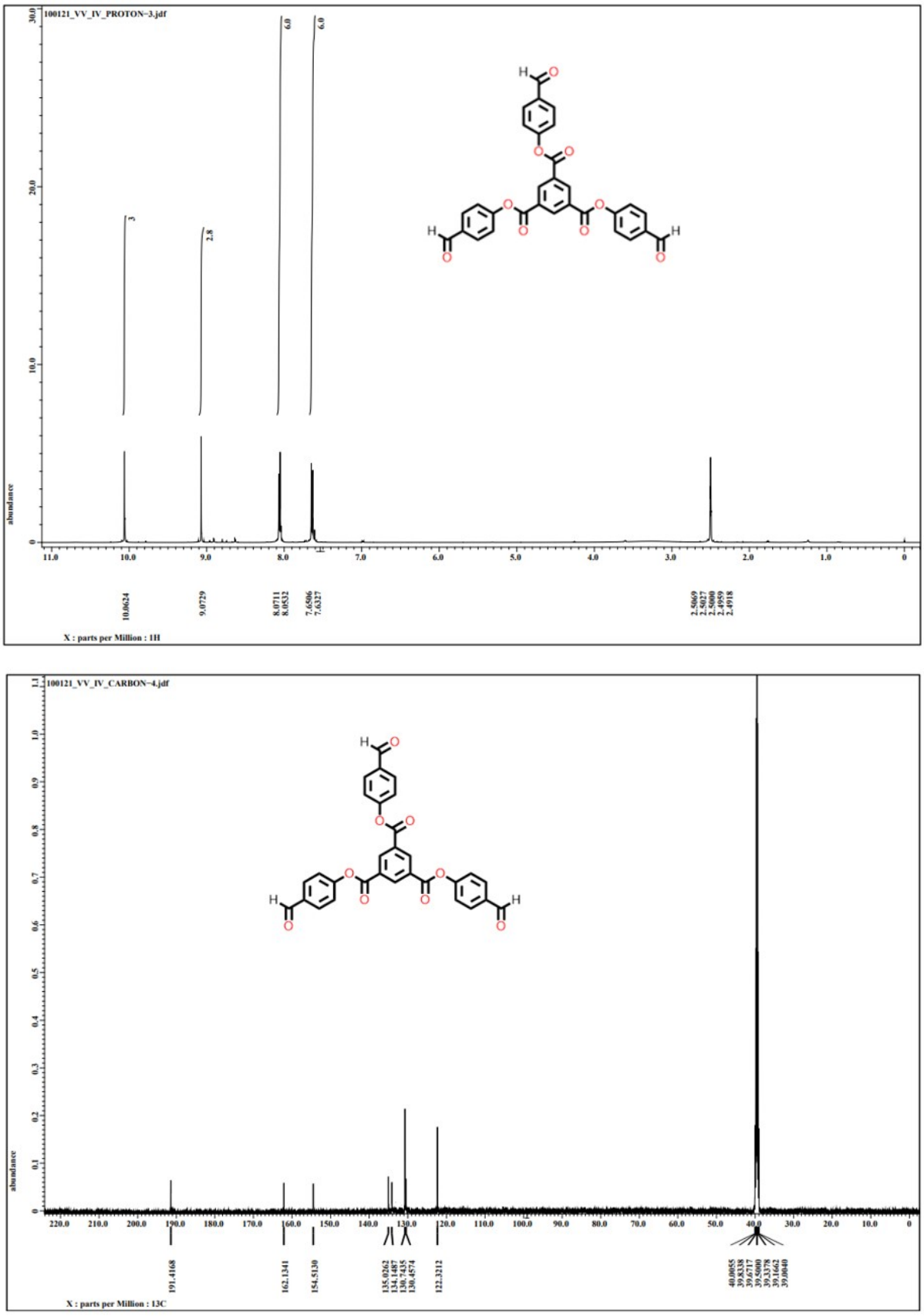
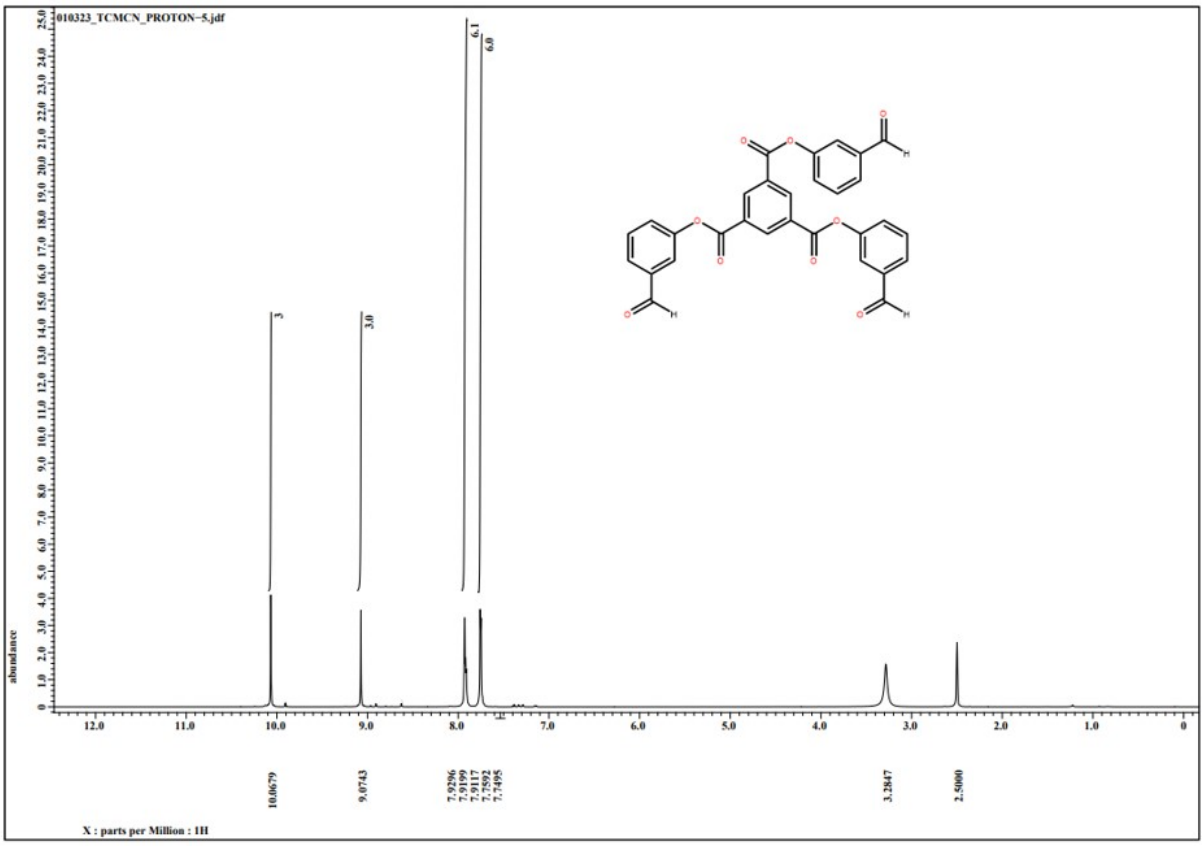



**Fig. S18.** $^{1}H$ and $^{13}C$ NMR of **TCpCHO.**



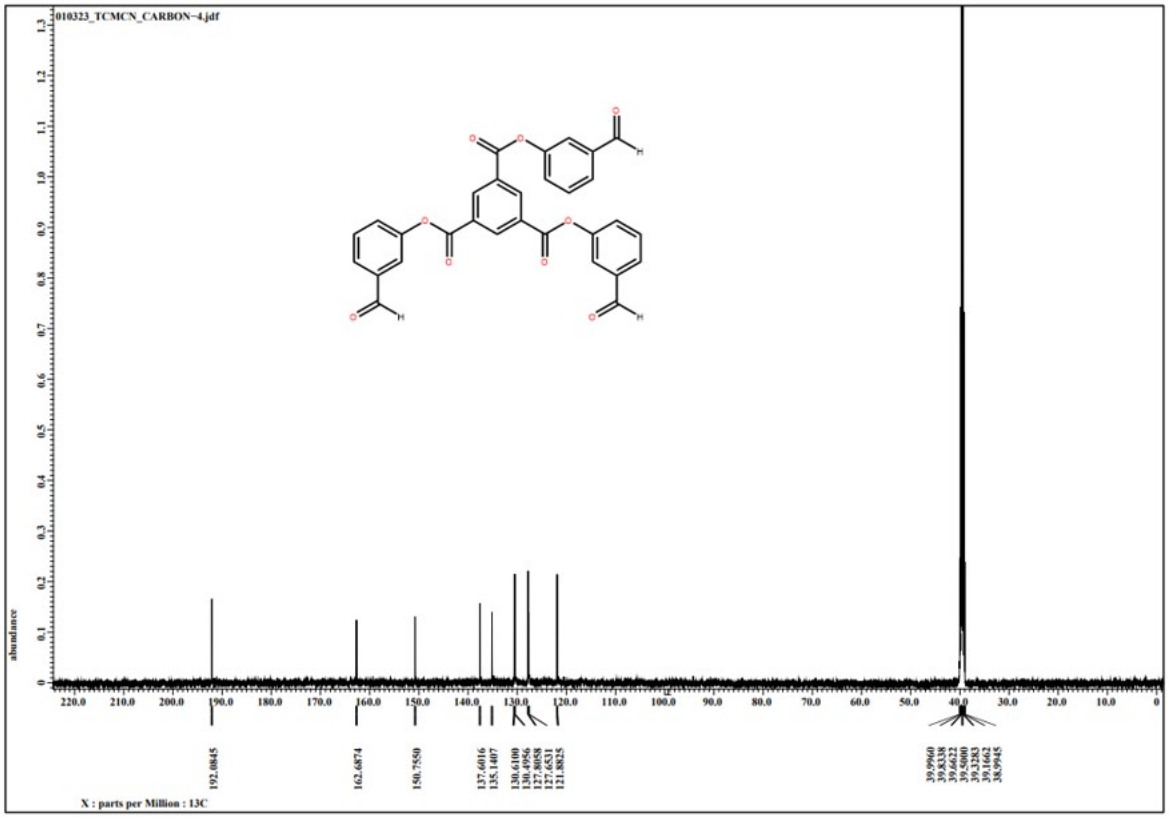




**Fig. S19.** $^{1}H$ and $^{13}C$ NMR of **TCmCHO**

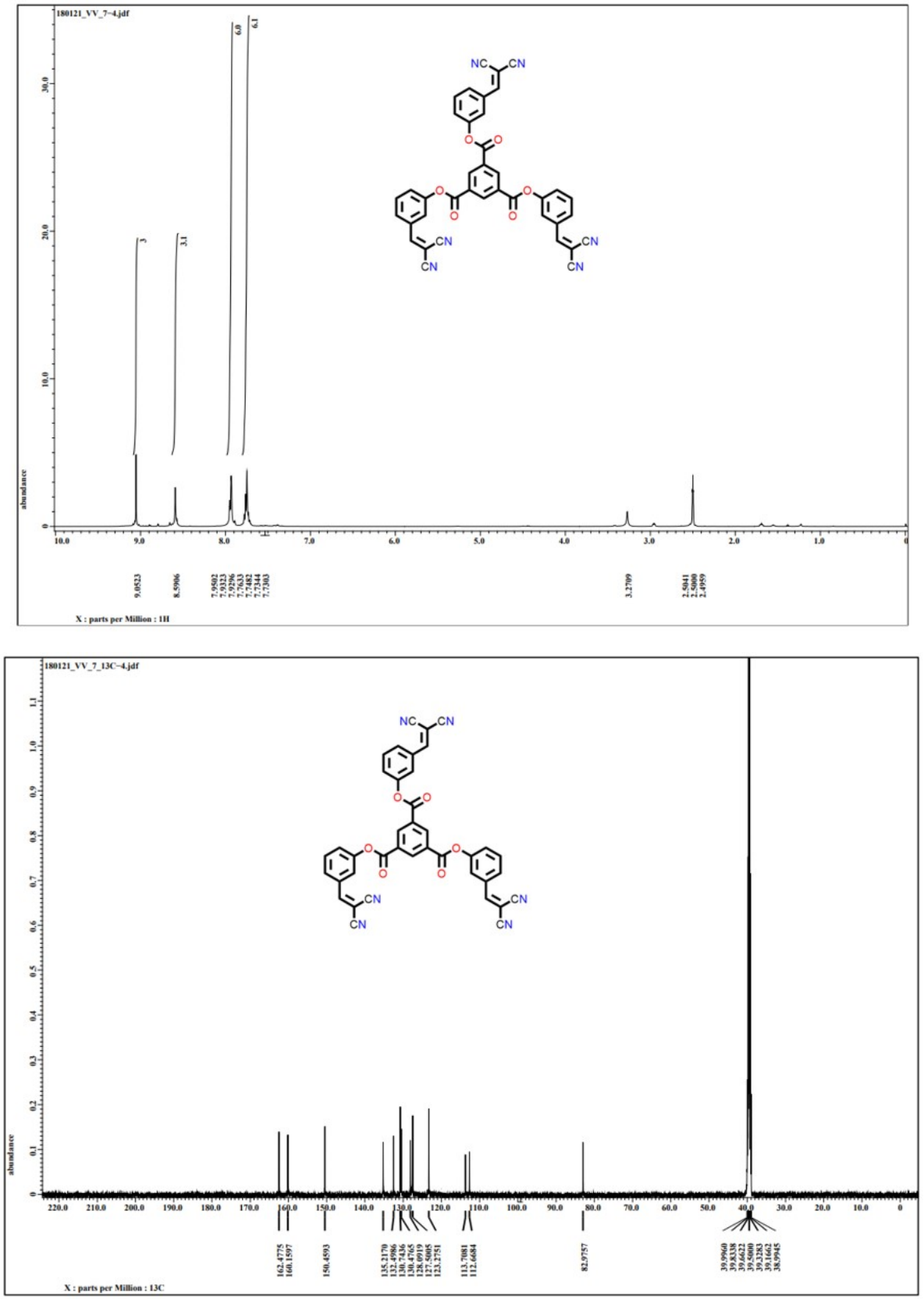



**Fig. S20.** $^{1}H$ and $^{13}C$ NMR of **TCmCN.**

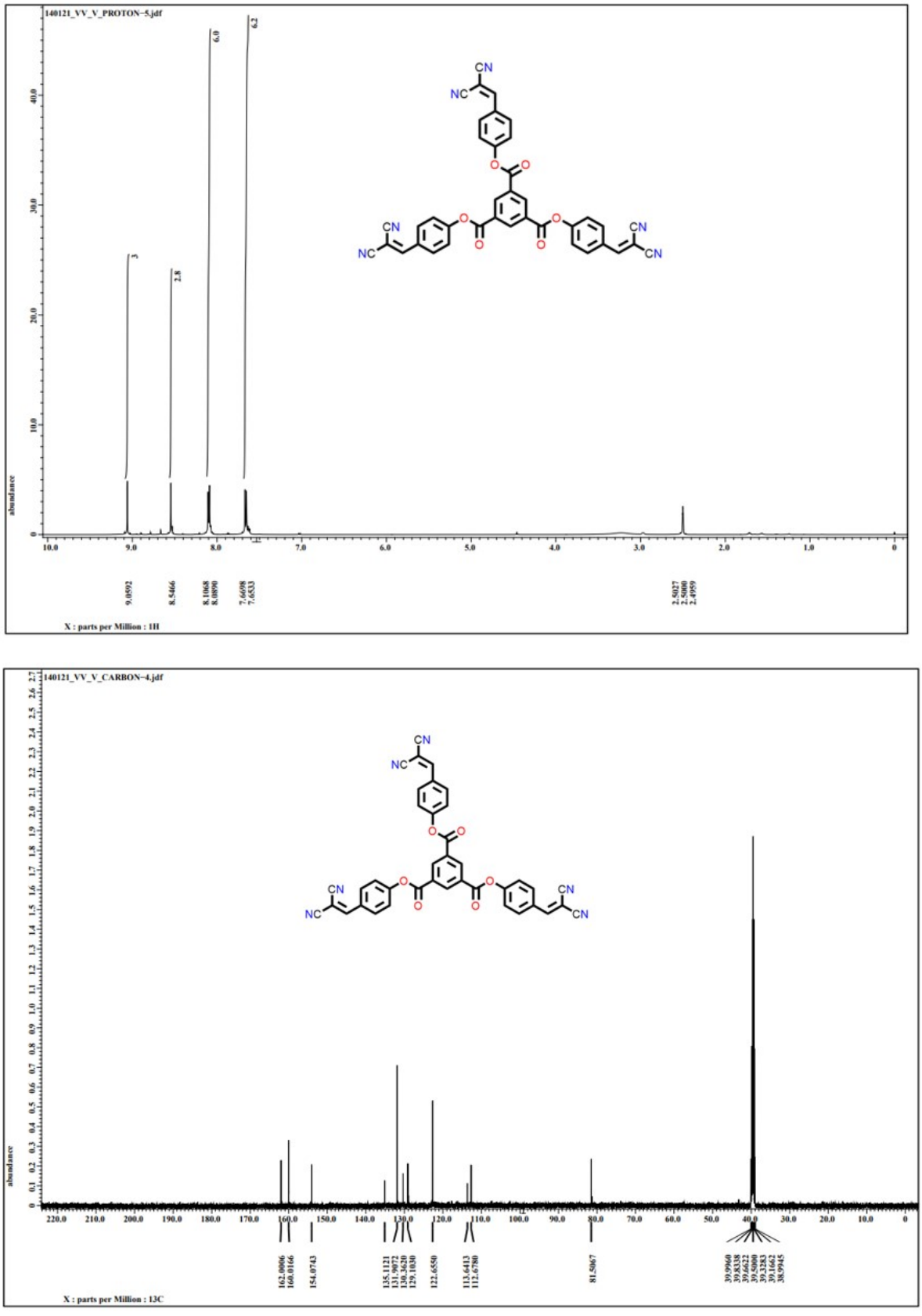



**Fig. S21.** $^{1}H$ and $^{13}C$ NMR of **TCpCN.**

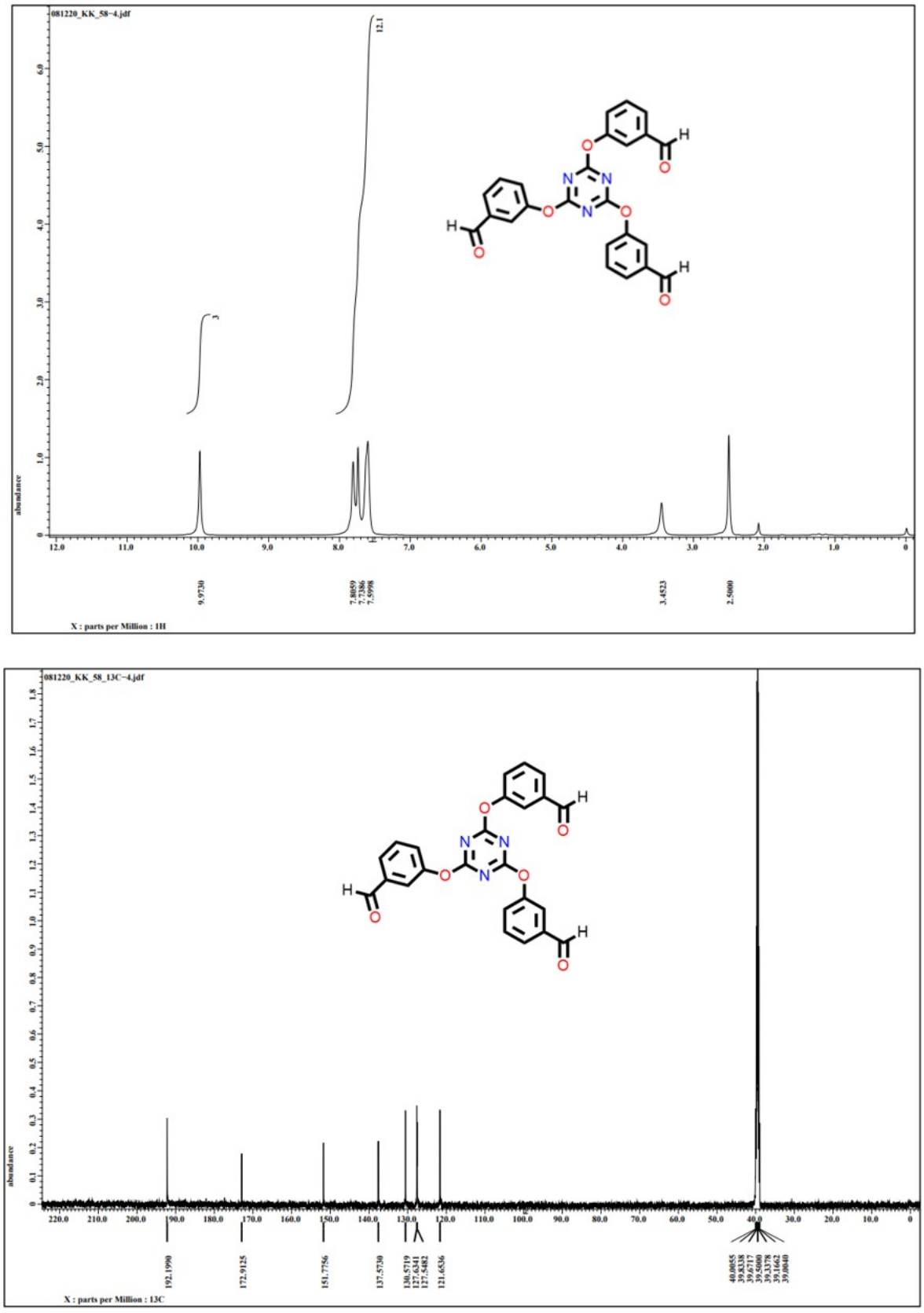



**Fig. S22.** $^{1}H$ and $^{13}C$ NMR of **TmCHO**

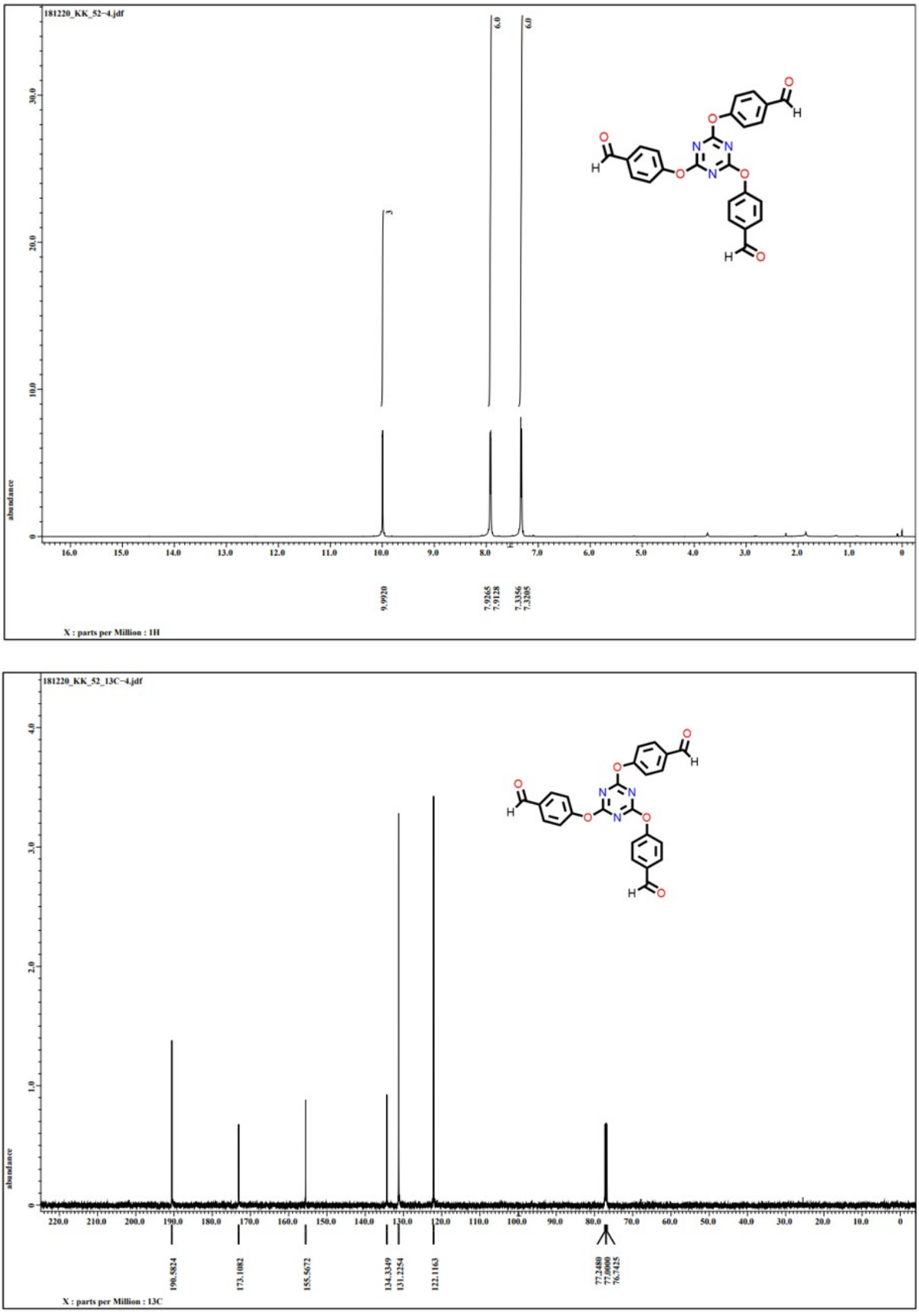



**Fig. S23.** $^{1}H$ and $^{13}C$ NMR of **TpCHO.**

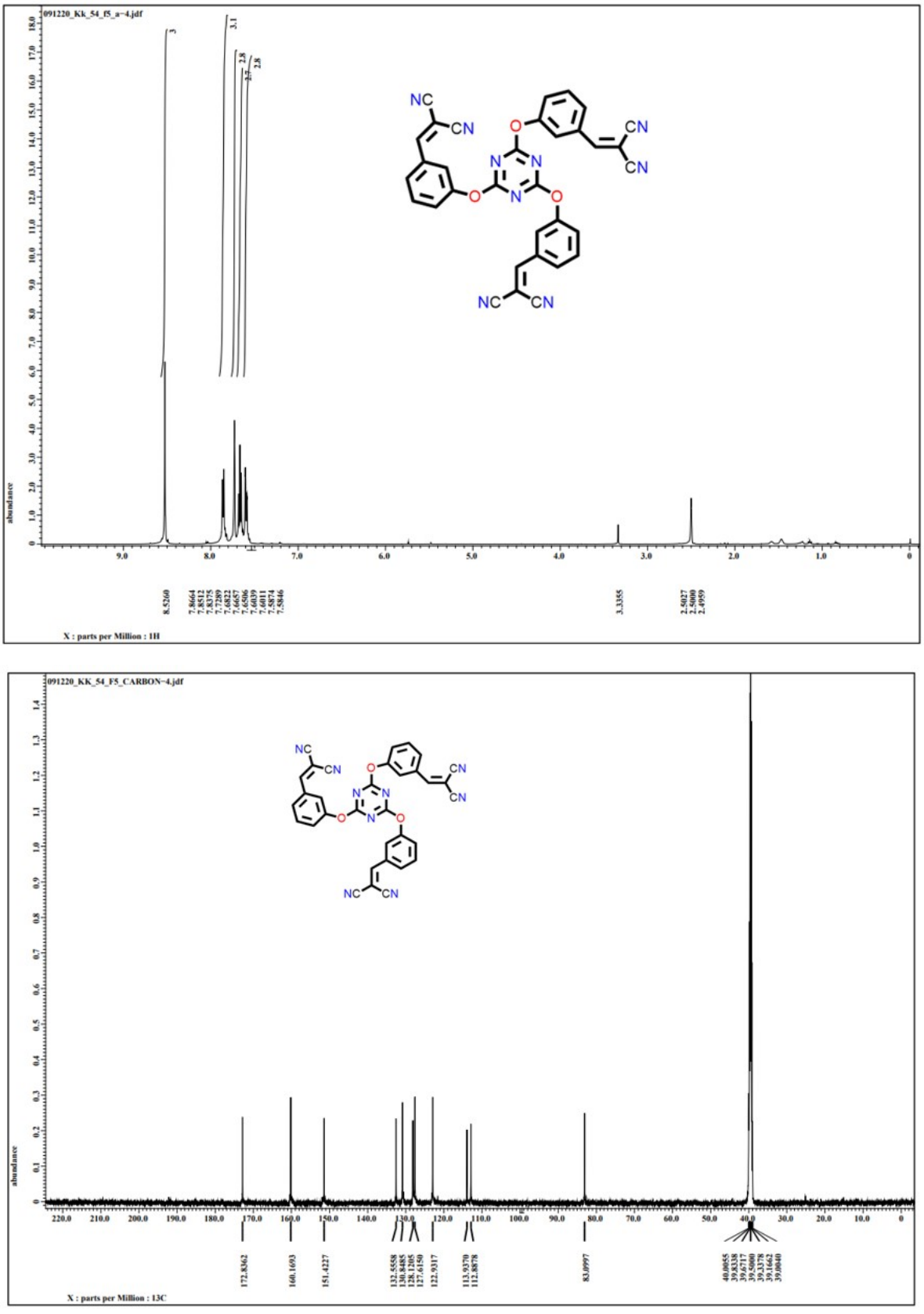



**Fig. S24.** $^{1}H$ and $^{13}C$ NMR of **TmCN.**

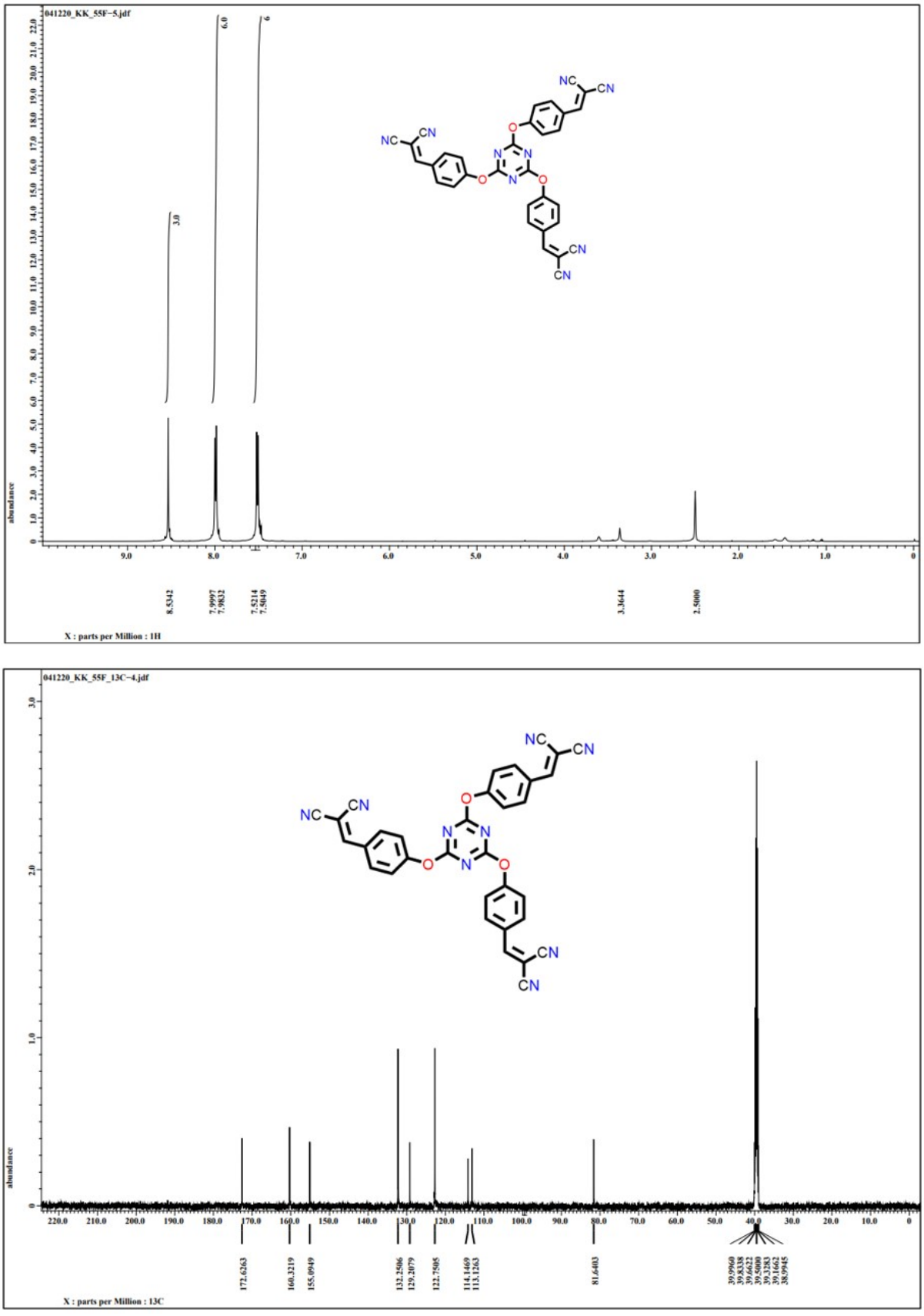



**Fig. S25.** $^{1}$H and $^{13}$C NMR of **TpCN.**

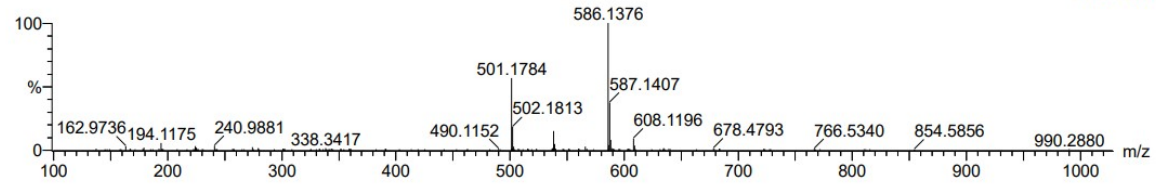
## 11. HRMS

**Elemental Composition Report** **Page 1**

**Single Mass Analysis**
Tolerance = 20.0 PPM / DBE: min = -1.5, max = 50.0
Element prediction: Off
Number of isotope peaks used for i-FIT = 3

Monoisotopic Mass, Even Electron Ions
48 formula(e) evaluated with 1 results within limits (up to 50 closest results for each mass)
Elements Used:
C: 0-35 H: 0-50 N: 0-9 O: 0-3

060423_SRY_TMCN 73 (0.757) IITRPR XEVO G2-XS QTOF
Test Name : 060423_SRY_TMCN
1: TOF MS ES+
2.65e+006




| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| Minimum: | | | | -1.5 | | | | |
| Maximum: | | 5.0 | 20.0 | 50.0 | | | | |
| Mass | Calc. Mass | mDa | PPM | DBE | i-FIT | Norm | Conf(%) | Formula |
| 586.1376 | 586.1376 | 0.0 | 0.0 | 30.5 | 643.3 | n/a | n/a | C33 H16 N9 O3 |

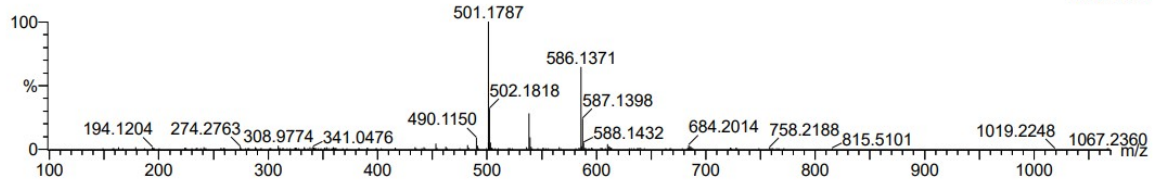
**Fig. S26.** HRMS of **TmCN**

**Elemental Composition Report** **Page 1**

**Single Mass Analysis**
Tolerance = 20.0 PPM / DBE: min = -1.5, max = 50.0
Element prediction: Off
Number of isotope peaks used for i-FIT = 3

Monoisotopic Mass, Even Electron Ions
62 formula(e) evaluated with 1 results within limits (up to 50 closest results for each mass)
Elements Used:
C: 0-33 H: 0-100 N: 0-9 O: 0-3

130423_SRY_TPCN 19 (0.214) IITRPR XEVO G2-XS QTOF
Test Name : 130423_SRY_TPCN
1: TOF MS ES+
2.35e+006




| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| Minimum: | | | | -1.5 | | | | |
| Maximum: | | 5.0 | 20.0 | 50.0 | | | | |
| Mass | Calc. Mass | mDa | PPM | DBE | i-FIT | Norm | Conf(%) | Formula |
| 586.1371 | 586.1376 | -0.5 | -0.9 | 30.5 | 557.0 | n/a | n/a | C33 H16 N9 O3 |

**Fig. S27.** HRMS of **TpCN.**

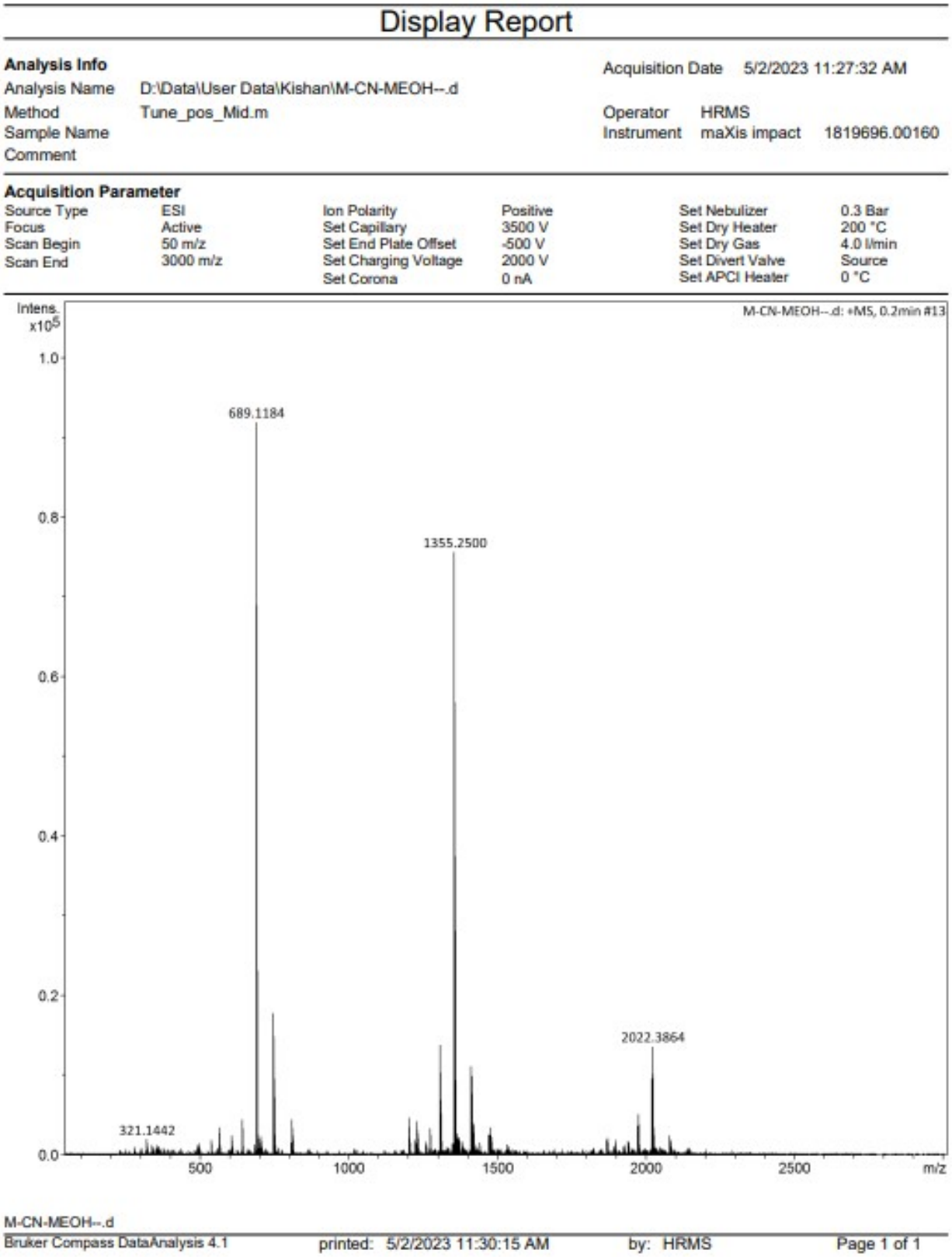



**Fig. S28.** HRMS of **TCmCN.**

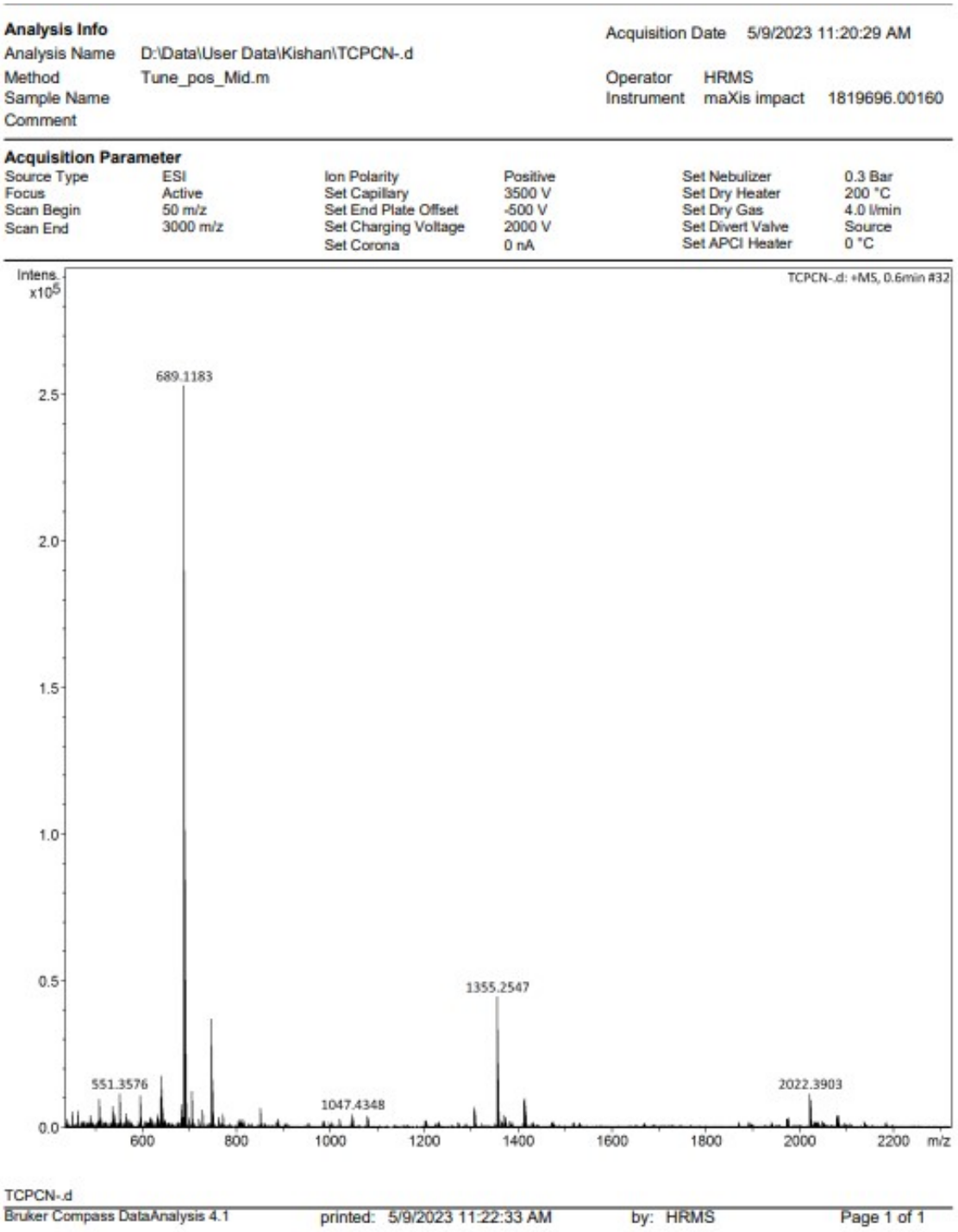



**Fig. S29.** HRMS of **TCpCN.**

**12. Cartesian Co-ordinates:**

**Cartesian coordinates of the optimized structures of the TCpCN molecule.**

| atomic number | X-coordinate | Y-coordinate | Z-coordinate |
|---|---|---|---|
| 6 | 0.958035000 | -2.068398000 | 0.302998000 |
| 6 | -0.363980000 | -1.623897000 | 0.312653000 |
| 6 | -0.640225000 | -0.254445000 | 0.295326000 |
| 6 | 0.410596000 | 0.664756000 | 0.268864000 |
| 6 | 1.733750000 | 0.217817000 | 0.258216000 |
| 6 | 2.008483000 | -1.150024000 | 0.275423000 |
| 6 | 3.397135000 | -1.689273000 | 0.268325000 |
| 6 | 0.183517000 | 2.136879000 | 0.253807000 |
| 6 | -1.438707000 | -2.655289000 | 0.339719000 |
| 6 | 5.680526000 | -0.942884000 | 0.233403000 |
| 6 | -1.610207000 | 3.739763000 | 0.269776000 |
| 6 | -3.833843000 | -2.844074000 | 0.349393000 |
| 6 | -4.041439000 | -3.859592000 | -0.585266000 |
| 6 | -5.251700000 | -4.532849000 | -0.595219000 |
| 6 | -6.272746000 | -4.204432000 | 0.320954000 |
| 6 | -6.029215000 | -3.168144000 | 1.246282000 |

| | | | |
|---|---|---|---|
| 6 | -4.821198000 | -2.490690000 | 1.263919000 |
| 6 | 6.455067000 | -0.249012000 | -0.695715000 |
| 6 | 7.830917000 | -0.400003000 | -0.693466000 |
| 6 | 8.456297000 | -1.250530000 | 0.241917000 |
| 6 | 7.642266000 | -1.930812000 | 1.171877000 |
| 6 | 6.264261000 | -1.784290000 | 1.177440000 |
| 6 | -1.130147000 | 4.684852000 | 1.173510000 |
| 6 | -1.704755000 | 5.946051000 | 1.180439000 |
| 6 | -2.756760000 | 6.286837000 | 0.304649000 |
| 6 | -3.220890000 | 5.302554000 | -0.592701000 |
| 6 | -2.650925000 | 4.041495000 | -0.608313000 |
| 6 | 9.881056000 | -1.486012000 | 0.334514000 |
| 6 | 10.928327000 | -1.008585000 | -0.400345000 |
| 6 | 12.257346000 | -1.424079000 | -0.074550000 |
| 6 | 10.831256000 | -0.106007000 | -1.501123000 |
| 6 | -3.276232000 | 7.633474000 | 0.408646000 |
| 6 | -4.279477000 | 8.271263000 | -0.263158000 |
| 6 | -4.579508000 | 9.632478000 | 0.056099000 |
| 6 | -5.091627000 | 7.698921000 | -1.287134000 |
| 6 | -7.568389000 | -4.845340000 | 0.391290000 |

| | | | |
|---|---|---|---|
| 6 | -8.122823000 | -5.852226000 | -0.345716000 |
| 6 | -7.493407000 | -6.537666000 | -1.427302000 |
| 6 | -9.450411000 | -6.289910000 | -0.043799000 |
| 8 | 3.680658000 | -2.857102000 | 0.298747000 |
| 8 | -1.252722000 | -3.842832000 | 0.348438000 |
| 8 | 1.053612000 | 2.966612000 | 0.243380000 |
| 8 | -1.152975000 | 2.430133000 | 0.240587000 |
| 8 | 4.318246000 | -0.679454000 | 0.209287000 |
| 8 | -2.678107000 | -2.075654000 | 0.366827000 |
| 7 | -7.011327000 | -7.110742000 | -2.307544000 |
| 7 | -10.522817000 | -6.647447000 | 0.195707000 |
| 7 | -5.765962000 | 7.262926000 | -2.118324000 |
| 7 | -4.826415000 | 10.732347000 | 0.310210000 |
| 7 | 13.333369000 | -1.756426000 | 0.184294000 |
| 7 | 10.784671000 | 0.624263000 | -2.395679000 |
| 1 | 1.169661000 | -3.129258000 | 0.316567000 |
| 1 | -1.662636000 | 0.091995000 | 0.302904000 |
| 1 | 2.535752000 | 0.941957000 | 0.237499000 |
| 1 | -3.268553000 | -4.119179000 | -1.294082000 |
| 1 | -5.395043000 | -5.314834000 | -1.326235000 |

| | | | |
|---|---|---|---|
| 1 | -6.800039000 | -2.896488000 | 1.957662000 |
| 1 | -4.633035000 | -1.694367000 | 1.972265000 |
| 1 | 5.967642000 | 0.402313000 | -1.409664000 |
| 1 | 8.410698000 | 0.145247000 | -1.423448000 |
| 1 | 8.104150000 | -2.583922000 | 1.902899000 |
| 1 | 5.657682000 | -2.313227000 | 1.897727000 |
| 1 | -0.327091000 | 4.443383000 | 1.854049000 |
| 1 | -1.337714000 | 6.686581000 | 1.881238000 |
| 1 | -4.024242000 | 5.513729000 | -1.282830000 |
| 1 | -3.001614000 | 3.280676000 | -1.293687000 |
| 1 | 10.169074000 | -2.165462000 | 1.130088000 |
| 1 | -2.780366000 | 8.245955000 | 1.154758000 |
| 1 | -8.215992000 | -4.455219000 | 1.169748000 |

**Cartesian coordinates of the optimized structures of the TCmCN molecule.**

| atomic number | X-coordinate | Y-coordinate | Z-coordinate |
|---|---|---|---|
| 6 | -1.338322000 | -2.054546000 | -1.310021000 |
| 6 | 0.022638000 | -1.847067000 | -1.086004000 |
| 6 | 0.478706000 | -0.592500000 | -0.673733000 |
| 6 | -0.431639000 | 0.449921000 | -0.488364000 |
| 6 | -1.794173000 | 0.239478000 | -0.711222000 |

| | | | |
|---|---|---|---|
| 6 | -2.248677000 | -1.013753000 | -1.122943000 |
| 6 | -3.689225000 | -1.299101000 | -1.375411000 |
| 6 | -0.007381000 | 1.809289000 | -0.049726000 |
| 6 | 0.942109000 | -2.999723000 | -1.300702000 |
| 6 | -5.846018000 | -0.289186000 | -1.318245000 |
| 6 | 1.918402000 | 3.083703000 | 0.534696000 |
| 6 | 3.243706000 | -3.616618000 | -1.183539000 |
| 6 | 4.009861000 | -3.893009000 | -0.066662000 |
| 6 | 5.088568000 | -4.793050000 | -0.171575000 |
| 6 | 5.348353000 | -5.392156000 | -1.420156000 |
| 6 | 4.565262000 | -5.097267000 | -2.528704000 |
| 6 | 3.505106000 | -4.202196000 | -2.421335000 |
| 6 | -6.644693000 | -0.081610000 | -0.209556000 |
| 6 | -8.045898000 | -0.082870000 | -0.355767000 |
| 6 | -8.590462000 | -0.301947000 | -1.636735000 |
| 6 | -7.766853000 | -0.506888000 | -2.736180000 |
| 6 | -6.383302000 | -0.499637000 | -2.586915000 |
| 6 | 2.810467000 | 3.700552000 | -0.323719000 |
| 6 | 3.478859000 | 4.875398000 | 0.075704000 |
| 6 | 3.214302000 | 5.399984000 | 1.356107000 |

| | | | |
|---|---|---|---|
| 6 | 2.315209000 | 4.762386000 | 2.199626000 |
| 6 | 1.659766000 | 3.597919000 | 1.800655000 |
| 6 | -8.989947000 | 0.123701000 | 0.725325000 |
| 6 | -8.802299000 | 0.359045000 | 2.055591000 |
| 6 | -9.941706000 | 0.523490000 | 2.904749000 |
| 6 | -7.540226000 | 0.462148000 | 2.714319000 |
| 6 | 4.397265000 | 5.448050000 | -0.890026000 |
| 6 | 5.180899000 | 6.563060000 | -0.838859000 |
| 6 | 6.002504000 | 6.890695000 | -1.963253000 |
| 6 | 5.271426000 | 7.472807000 | 0.257236000 |
| 6 | 5.967761000 | -5.161097000 | 0.921259000 |
| 6 | 6.006720000 | -4.758694000 | 2.223839000 |
| 6 | 5.123605000 | -3.816505000 | 2.831886000 |
| 6 | 7.005014000 | -5.299689000 | 3.093987000 |
| 8 | -4.126243000 | -2.358255000 | -1.742678000 |
| 8 | 0.598319000 | -4.096309000 | -1.656896000 |
| 8 | -0.752186000 | 2.735782000 | 0.135907000 |
| 8 | 1.346896000 | 1.881404000 | 0.112235000 |
| 8 | -4.464454000 | -0.202172000 | -1.131137000 |
| 8 | 2.237947000 | -2.659125000 | -1.036138000 |

| | | | |
|---|---|---|---|
| 7 | 4.424822000 | -3.056663000 | 3.351071000 |
| 7 | 7.809287000 | -5.732716000 | 3.801351000 |
| 7 | 5.369035000 | 8.229073000 | 1.125364000 |
| 7 | 6.667976000 | 7.160043000 | -2.868448000 |
| 7 | -10.858034000 | 0.656925000 | 3.595613000 |
| 7 | -6.534194000 | 0.550952000 | 3.275739000 |
| 1 | -1.689571000 | -3.026794000 | -1.629003000 |
| 1 | 1.531330000 | -0.429708000 | -0.497423000 |
| 1 | -2.486646000 | 1.055442000 | -0.560129000 |
| 1 | 3.756373000 | -3.404344000 | 0.862254000 |
| 1 | 6.173303000 | -6.088168000 | -1.513445000 |
| 1 | 4.780112000 | -5.562152000 | -3.482634000 |
| 1 | 2.891934000 | -3.960674000 | -3.279066000 |
| 1 | -6.165732000 | 0.080234000 | 0.744530000 |
| 1 | -9.666495000 | -0.306713000 | -1.762452000 |
| 1 | -8.200481000 | -0.669810000 | -3.714773000 |
| 1 | -5.730389000 | -0.650134000 | -3.436422000 |
| 1 | 2.990835000 | 3.268019000 | -1.300032000 |
| 1 | 3.706882000 | 6.297348000 | 1.699457000 |
| 1 | 2.121429000 | 5.171115000 | 3.183390000 |

| | | | |
|---|---|---|---|
| 1 | 0.964992000 | 3.097411000 | 2.462096000 |
| 1 | -10.030419000 | 0.082645000 | 0.420513000 |
| 1 | 4.473467000 | 4.891779000 | -1.818551000 |
| 1 | 6.730513000 | -5.884503000 | 0.652766000 |

**Cartesian coordinates of the optimized structures of the TmCN molecule.**

| atomic number | X-coordinate | Y-coordinate | Z-coordinate |
|---|---|---|---|
| 7 | 1.553603000 | -2.829618000 | -0.726267000 |
| 6 | 0.335309000 | -2.323022000 | -0.592225000 |
| 7 | 0.020973000 | -1.045866000 | -0.390075000 |
| 6 | 1.078554000 | -0.246284000 | -0.327484000 |
| 7 | 2.352470000 | -0.607746000 | -0.445498000 |
| 6 | 2.511224000 | -1.910555000 | -0.640713000 |
| 8 | 3.749534000 | -2.408850000 | -0.795863000 |
| 8 | 0.901443000 | 1.066874000 | -0.116970000 |
| 8 | -0.647068000 | -3.233165000 | -0.695799000 |
| 6 | 4.880030000 | -1.614081000 | -0.630295000 |
| 6 | -0.382328000 | 1.611424000 | -0.070904000 |
| 6 | -1.976067000 | -2.896054000 | -0.452770000 |
| 6 | -2.395372000 | -2.498498000 | 0.815720000 |
| 6 | -3.740921000 | -2.267181000 | 1.042816000 |

| | | | |
|---|---|---|---|
| 6 | -4.686746000 | -2.435194000 | 0.008913000 |
| 6 | -4.226288000 | -2.853457000 | -1.256171000 |
| 6 | -2.878971000 | -3.082912000 | -1.491403000 |
| 6 | 5.177217000 | -1.023633000 | 0.597218000 |
| 6 | 6.362774000 | -0.324601000 | 0.743133000 |
| 6 | 7.269866000 | -0.208305000 | -0.331805000 |
| 6 | 6.940881000 | -0.830615000 | -1.553336000 |
| 6 | 5.753887000 | -1.530598000 | -1.706839000 |
| 6 | -0.839723000 | 2.092647000 | 1.148205000 |
| 6 | -2.072266000 | 2.727293000 | 1.194649000 |
| 6 | -2.857082000 | 2.886484000 | 0.034470000 |
| 6 | -2.354265000 | 2.391659000 | -1.187598000 |
| 6 | -1.123603000 | 1.759716000 | -1.240904000 |
| 6 | 8.531295000 | 0.500354000 | -0.285697000 |
| 6 | 9.135275000 | 1.211637000 | 0.710818000 |
| 6 | 10.407654000 | 1.816535000 | 0.464983000 |
| 6 | 8.609540000 | 1.424632000 | 2.020149000 |
| 6 | -4.126300000 | 3.564927000 | 0.200082000 |
| 6 | -5.122420000 | 3.849661000 | -0.688270000 |
| 6 | -6.285354000 | 4.548733000 | -0.235819000 |

| | | | |
|---|---|---|---|
| 6 | -5.127211000 | 3.505779000 | -2.073279000 |
| 6 | -6.112644000 | -2.220922000 | 0.141436000 |
| 6 | -6.868998000 | -1.787693000 | 1.191968000 |
| 6 | -6.374736000 | -1.418266000 | 2.478658000 |
| 6 | -8.285703000 | -1.672864000 | 1.033163000 |
| 7 | -6.004268000 | -1.109510000 | 3.528909000 |
| 7 | -9.430806000 | -1.578800000 | 0.910967000 |
| 7 | -5.161395000 | 3.238883000 | -3.197152000 |
| 7 | -7.226961000 | 5.114031000 | 0.123007000 |
| 7 | 11.435678000 | 2.307489000 | 0.271852000 |
| 7 | 8.210316000 | 1.614879000 | 3.087872000 |
| 1 | -1.675309000 | -2.377248000 | 1.613743000 |
| 1 | -4.049308000 | -1.963362000 | 2.032164000 |
| 1 | -4.936754000 | -2.995821000 | -2.061722000 |
| 1 | -2.523358000 | -3.406954000 | -2.460743000 |
| 1 | 4.488916000 | -1.114319000 | 1.426425000 |
| 1 | 6.578473000 | 0.124850000 | 1.701165000 |
| 1 | 7.626163000 | -0.758497000 | -2.389593000 |
| 1 | 5.500204000 | -2.012045000 | -2.642213000 |
| 1 | -0.233482000 | 1.974365000 | 2.036771000 |

| | | | |
|---|---|---|---|
| 1 | -2.438230000 | 3.106354000 | 2.141337000 |
| 1 | -2.915188000 | 2.502622000 | -2.103686000 |
| 1 | -0.734707000 | 1.388464000 | -2.180075000 |
| 1 | 9.096976000 | 0.460963000 | -1.210823000 |
| 1 | -4.320487000 | 3.905235000 | 1.212029000 |
| 1 | -6.681530000 | -2.440157000 | -0.756240000 |

**Cartesian coordinates of the optimized structures of the TpCN molecule.**

| atomic number | X-coordinate | Y-coordinate | Z-coordinate |
|---|---|---|---|
| 7 | 1.693806000 | -1.210764000 | 2.289292000 |
| 6 | 0.456031000 | -1.054424000 | 1.839102000 |
| 7 | 0.045220000 | -0.181208000 | 0.921297000 |
| 6 | 1.022354000 | 0.581528000 | 0.445808000 |
| 7 | 2.304117000 | 0.538537000 | 0.797444000 |
| 6 | 2.563250000 | -0.380042000 | 1.720594000 |
| 8 | 3.823067000 | -0.528545000 | 2.160731000 |
| 8 | 0.744709000 | 1.507914000 | -0.484220000 |
| 8 | -0.436868000 | -1.888358000 | 2.395142000 |
| 6 | 4.836269000 | 0.295303000 | 1.654832000 |
| 6 | -0.565166000 | 1.655453000 | -0.957100000 |
| 6 | -1.784749000 | -1.823368000 | 2.022904000 |

| | | | |
|---|---|---|---|
| 6 | -2.264974000 | -2.762183000 | 1.129047000 |
| 6 | -3.637174000 | -2.790181000 | 0.807505000 |
| 6 | -4.492207000 | -1.849911000 | 1.415458000 |
| 6 | -3.985184000 | -0.921340000 | 2.314111000 |
| 6 | -2.626824000 | -0.899765000 | 2.628859000 |
| 6 | 5.600111000 | -0.168237000 | 0.601508000 |
| 6 | 6.683135000 | 0.607602000 | 0.143143000 |
| 6 | 6.954556000 | 1.834030000 | 0.782503000 |
| 6 | 6.173312000 | 2.275163000 | 1.842552000 |
| 6 | 5.103432000 | 1.504888000 | 2.288954000 |
| 6 | -1.339363000 | 2.677157000 | -0.439945000 |
| 6 | -2.625999000 | 2.918251000 | -0.962650000 |
| 6 | -3.092092000 | 2.101904000 | -2.012041000 |
| 6 | -2.291839000 | 1.086365000 | -2.517029000 |
| 6 | -1.019181000 | 0.853558000 | -1.996200000 |
| 6 | 7.558919000 | 0.239322000 | -0.952750000 |
| 6 | 7.555517000 | -0.838151000 | -1.788497000 |
| 6 | 8.560366000 | -0.941381000 | -2.801532000 |
| 6 | 6.616239000 | -1.912431000 | -1.759569000 |
| 6 | -3.366472000 | 4.010924000 | -0.360056000 |

| | | | |
|---|---|---|---|
| 6 | -4.612898000 | 4.503399000 | -0.611182000 |
| 6 | -5.092462000 | 5.611428000 | 0.156563000 |
| 6 | -5.523872000 | 4.012981000 | -1.594661000 |
| 6 | -4.052404000 | -3.810416000 | -0.137266000 |
| 6 | -5.270818000 | -4.096260000 | -0.678503000 |
| 6 | -6.488409000 | -3.400270000 | -0.412831000 |
| 6 | -5.386789000 | -5.181381000 | -1.603859000 |
| 7 | 9.369111000 | -1.030288000 | -3.621715000 |
| 7 | 5.869546000 | -2.794252000 | -1.761584000 |
| 7 | -6.284938000 | 3.641311000 | -2.380551000 |
| 7 | -5.484454000 | 6.505510000 | 0.774306000 |
| 7 | -7.490907000 | -2.858798000 | -0.220916000 |
| 7 | -5.486582000 | -6.055767000 | -2.352279000 |
| 1 | -1.579214000 | -3.474544000 | 0.687263000 |
| 1 | -5.549897000 | -1.842430000 | 1.198632000 |
| 1 | -4.652374000 | -0.207500000 | 2.780668000 |
| 1 | -2.227250000 | -0.185401000 | 3.337485000 |
| 1 | 5.348646000 | -1.121229000 | 0.160475000 |
| 1 | 7.787253000 | 2.436837000 | 0.440430000 |
| 1 | 6.396683000 | 3.218134000 | 2.325341000 |

| | | | |
|---|---|---|---|
| 1 | 4.486220000 | 1.825679000 | 3.118585000 |
| 1 | -0.944150000 | 3.290206000 | 0.360530000 |
| 1 | -4.070930000 | 2.257396000 | -2.440349000 |
| 1 | -2.657412000 | 0.468824000 | -3.327758000 |
| 1 | -0.386525000 | 0.069658000 | -2.392854000 |
| 1 | 8.355288000 | 0.951184000 | -1.142659000 |
| 1 | -2.834563000 | 4.527611000 | 0.431890000 |
| 1 | -3.251135000 | -4.459564000 | -0.474590000 |

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