**Electronic Supporting Information**

**For**

# Polypyridyl Co Complex-based Water Reduction Catalysts: Why Replace a Pyridine Group with Isoquinoline rather than Quinoline?

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# Experimental section

## Materials

Cobalt(II) chloride hexahydrate, trimethylamine (TEA), extra dry acetonitrile, *N, N*-bis(2-pyridinylmethyl) amine, 2-pyridinecarboxaldehyde, 2-quinolinecarboxaldehyde, 1-isoquinolinecarboxaldehyde, and 3-isoquinolinecarboxaldehyde were purchased from Innochem. Milli-Q ultrapure water was used in all experiments. $TBAPF_6$ (tetrabutylammonium hexafluorophosphate) was purchased from Adamas-beta and was recrystallized from ethanol and dried under vacuum overnight before use. All other materials were used as received without further purification.

## Instruments

$^1$H NMR spectra were obtained on a Bruker DMX-400 MHz spectrophotometer. ESI-MS spectra were taken on a Q-Exactive mass spectrometer and a Bruker solariX mass spectrometer. Elemental analysis was performed on vario MICRO cube. Single-crystal X-ray diffraction data for complexes **C3** and **C4** were collected on Bruker D8. UV-vis absorption spectra were recorded on a Shimadzu UV-2450 spectrophotometer. Fluorescence emission spectra were run on a Hitachi F-4600 fluorescence spectrophotometer.

## Synthesis

**L1: TPA (tris(2-pyridinylmethyl)-amine)**

In a two-neck-flask, 2-pyridinecarboxaldehyde (321 mg, 3 mmol) and *N, N*-bis(2-pyridinylmethyl) amine (600 mg, 3 mmol) were combined and dissolved in 25 mL 1,2-dichloroethane. Then sodium triacetoxyborohydride (1016 mg, 4.8 mmol) was added into the solution. The mixture was bubbled with argon slowly and stirred for 2 days at room temperature. After removing the solvent by vacuum rotary evaporation, the crude product was dissolved in dichloromethane and washed with saturated $NaCO_3$ aqueous solution to neutralize the excess acid. After drying the solution by anhydrous $MgSO_4$, dichloromethane was removed by vacuum rotary evaporation. Further purification was made by alumina column chromatography. The obtained product was yellow oil (661 mg, 75.9%). $^1$H NMR (400 MHz, chloroform-d) δ 8.54 (d, $J$ = 5.0 Hz, 3H), 7.74-7.62 (m, 3H), 7.58 (d, $J$ = 7.8 Hz, 3H), 7.15 (dd, $J$ = 7.3, 5.0 Hz, 3H), 3.93 (s, 6H). ESI-MS: m/z = 291.1593, m/z = 313.1410 (calcd m/z = 291.16 for $[M + H]^+$, m/z = 313.14 for $[M + Na]^+$).

**L2: BPQA (*N, N*-bis(2-pyridinylmethyl)-quinoline-6-methanamine)**

Following the synthetic method of TPA except for substituting 2-quinolinecarboxaldehyde (500 mg, 3.2 mmol) for 2-pyridinecarboxaldehyde, BPQA was obtained as yellow oil. (701.5 mg, 64.8%). $^1$H NMR (400 MHz, chloroform-d) δ 8.54 (d, $J$ = 4.9 Hz, 2H), 8.13 (d, $J$ = 8.5 Hz, 1H), 8.06 (d, $J$ = 8.5 Hz, 1H), 7.77 (dd, $J$ = 10.6, 8.4 Hz, 2H), 7.72-7.62 (m, 3H), 7.59 (d, $J$ = 7.8 Hz,

2H), 7.50 (t, *J* = 7.5 Hz, 1H), 7.14 (dd, *J* = 7.2, 5.2 Hz, 2H), 4.07 (s, 2H), 3.95 (s, 4H). ESI-MS: m/z = 341.1753 (calcd m/z = 341.18 for $[M + H]^+$ ).

**L3: DPA-3-IQA (*N, N*-bis(2-pyridinylmethyl)-isoquinoline-3-methanamine)**

Following the synthetic method of TPA except for substituting 3-isoquinolinecarboxaldehyde (250 mg, 1.6 mmol) for 2-pyridinecarboxaldehyde, DPA-3-IQA was obtained as yellow oil. (372 mg, 63.7%). $^1$H NMR (400 MHz, chloroform-d) δ 9.22 (s, 1H), 8.57-8.49 (m, 2H), 7.95 (d, *J* = 8.2 Hz, 1H), 7.88 (s, 1H), 7.82 (d, *J* = 8.3 Hz, 1H), 7.68-7.63 (m, 5H), 7.56 (t, *J* = 7.5 Hz, 1H), 7.13 (q, *J* = 4.5 Hz, 2H), 4.09 (s, 2H), 3.98 (s, 4H). ESI-MS: m/z = 341.1752 (calcd m/z = 341.18 for $[M + H]^+$ ).

**L4: DPA-1-IQA (*N, N*-bis(2-pyridinylmethyl)-isoquinoline-1-methanamine)**

Following the synthetic method of TPA except for substituting 1-isoquinolinecarboxaldehyde (250 mg, 1.6 mmol) for 2-pyridinecarboxaldehyde, the DPA-1-IQA was obtained as yellow oil. (307 mg, 52.6%). $^1$H NMR (400 MHz, chloroform-d) δ 8.54 (d, J = 4.9 Hz, 2H), 8.43 (d, J = 5.7 Hz, 1H), 8.29 (d, J = 8.5 Hz, 1H), 7.77 (d, *J* = 8.2 Hz, 1H), 7.62 (dt, *J* = 19.0, 7.5 Hz, 3H), 7.52 (dd, *J* = 10.4, 6.7 Hz, 2H), 7.40 (d, *J* = 7.8 Hz, 2H), 7.13 (dd, *J* = 7.2, 5.3 Hz, 2H), 4.38 (s, 2H), 3.95 (s, 4H). ESI-MS: m/z = 341.1749, m/z = 363.1566 (calcd m/z = 341.18 for $[M + H]^+$, m/z = 363.16 for $[M + Na]^+$).

**C1: [Co(TPA)Cl]Cl**

Under an argon atmosphere, TPA (162 mg, 0.56 mmol) in dry acetonitrile was combined with 1 equivalent $CoCl_2 \cdot 6H_2O$ (133 mg, 0.56 mmol) which was also dissolved in dry acetonitrile. The mixture was stirred for 4 h at room temperature. Then the crude product was precipitated by adding diethyl ether into the mixture. After filtering and washing by diethyl ether, the solid was dried under vacuum to afford green crystals (194.5 mg, 83.1%). ESI-MS: m/z = 384.0639 (calcd m/z = 384.06 for $[Co(TPA)Cl]^+$). Elemental analysis: anal. calcd for $C_{18}H_{18}Cl_2CoN_4$: C, 51.45; H, 4.32; N, 13.33. Found: C, 51.12; H, 4.37; N, 13.50.

**C2: [Co(BPQA)Cl]Cl**

Following the synthetic method of [Co(TPA)Cl]Cl except for substituting BPQA (340 mg, 1 mmol) for TPA, [Co(BPQA)Cl]Cl was got as green crystals (377 mg, 80.2%). ESI-MS: m/z = 434.0691 (calcd m/z = 434.07 for $[Co(BPQA)Cl]^+$). Elemental analysis: anal. calcd for $C_{22}H_{20}Cl_2CoN_4$: C, 56.19; H, 4.29; N, 11.91. Found: C, 56.51; H, 4.31; N, 11.85.

**C3: [Co(DPA-3-IQA)Cl]Cl**

Following the synthetic method of [Co(TPA)Cl]Cl except for substituting DPA-3-IQA (170 mg, 0.5 mmol) for TPA, [Co(DPA-3-IQA)Cl]Cl was got as green crystals (148.7 mg, 63.3%). The single crystal of **C3** suitable for X-ray crystallographic analysis were obtained by slow diffusion of tetrahydrofuran into concentrated acetonitrile solution of **C3**.

ESI-MS: m/z = 434.0704 (calcd m/z = 434.07 for $[Co(DPA-3-IQA)Cl]^+$). Elemental analysis: anal. calcd for $C_{22}H_{20}Cl_2CoN_4$: C, 56.19; H, 4.29; N, 11.91. Found: C, 56.42; H, 4.28; N, 11.66.

**C4: [Co(DPA-1-IQA)Cl]Cl**

Following the synthetic method of [Co(TPA)Cl]Cl except for substituting DPA-1-IQA (179 mg, 0.5 mmol) for TPA, [Co(DPA-1-IQA)Cl]Cl was got as green crystals (160.9 mg, 68.5%). The single crystal of **C4** suitable for X-ray crystallographic analysis were obtained by slow diffusion of tetrahydrofuran into concentrated acetonitrile solution of **C4**.

ESI-MS: m/z = 434.0703 (calcd m/z = 434.07 for $[Co(DPA\text{-}1\text{-}IQA)Cl]^+$). Elemental analysis: anal. calcd for $C_{22}H_{20}Cl_2CoN_4$: C, 56.19; H, 4.29; N, 11.91. Found: C, 55.98; H, 4.26; N, 11.73.

## Photocatalytic $H_2$ evolution

5 mL of $CH_3CN/H_2O$ (8:2, v/v) solution containing 0.2 mM $[Ir(ppy)_2(dtbpy)]PF_6$, 10 μM cobalt complexes, and 0.3 M TEA was put into a 25 mL home-made glass vial equipped with a rubber-septum-sealed outlet. After bubbling with argon for 25 min to remove the oxygen, 3 mL of gas in the vial was removed by a syringe and 3 mL of methane was injected into the reaction vessel to serve as the internal standard for $H_2$ quantification. Then the vial was irradiated by an LED light (λ = 455 nm ± 10 nm, 40 W), The production of $H_2$ was monitored and quantified by gas chromatography on a Shimadzu GC-2014 equipped with thermal conductivity detector (TCD), 5 Å molecular sieves 80/100 mesh packed column, $N_2$ gas as carrier.

## Electrochemistry

Electrochemical measurements were carried out using an EG&G model 283 potentiostat/galvanostat in a four-neck electrode cell with a three-electrode system, in which there were a 3 mm glassy carbon working electrode, a 1×1×0.1 cm platinum-plate counter electrode, and a SCE (saturated calomel electrode) reference electrode. Cyclic voltammetry was conducted at a scan rate of 50 mV/s 1 in 10 mL argon-saturated anhydrous MeCN containing 0.1 M n-$Bu_4NPF_6$ as the supporting electrolyte. Controlled potential electrolysis (CPE) were performed under stirring in the same kind of equipment.

## Luminescence quenching experiments

A solution of 10 μM $[Ir(ppy)_2(dtbpy)]Cl$ in $MeCN/H_2O$ (8:2, v/v) was prepared and degassed with argon for 25 min. Then 50 to 250 μL of solutions containing a quencher was added. Steady-state luminescence spectra were collected subsequently. The luminescence intensities with and without a quencher were used for the calculation of the bimolecular quenching rate constant $k_q$ by the Stern-Volmer equation.

$$I_0/I = 1 + k_q\tau_0[Q] \quad (1)$$

In this equation, $I_0$ and $I$ are emission intensities of the photosensitizer without or with the quencher. $\tau_0$ is the triplet excited state lifetime of the photosensitizer which is 0.557 μs[1], [Q] is the concentration of the quencher.

## DFT calculations

The cobalt complex geometry optimizations and Mulliken spin population analysis were performed with the Gaussian 09 (G09) program package 3[2] employing the density function theory (DFT) method with B3LYP.[3, 4] The LANL2DZ[5, 6, 7] basis set was used for the Co atom and 6-31+g(d) basis set was applied for all the other atoms. In single-point calculations, 6-311+g(d, p) basis set was used for the C, N, H, Cl atoms for more accurate results. The subsequent frequency analysis calculations were carried out on the optimized structures at the same level of theory to identify imaginary frequency. The solvation effect of water was taken into account using the SMD continuum solution model.[8] The Spin Density Distribution were analyzed and visualized by the Multiwfn program.[9]

The Gibbs free energy change was calculated following the equation (2), where the $G_{sol}$ is the Gibbs free energy in the aqueous sphere. The $G_{gas}$ is Gibbs free energy in the gas sphere. The $\Delta G_{solv}$ is solvation free energies from 1 M gas sphere (24.5 L/mol at 1 atm, 298.15 K) to 1 M in water solution. The $\Delta G^{0/*}$ is the correction for the free energy change from the standard state gas phase (1 atm, 298.15 K) to the standard state aqueous phase. It can be calculated from the equation (3), we got $\Delta G^{0/*}$ =1.89 kcal/mol. The Gibbs free energies of proton and electron were obtained from previously reported values.[10]

$$G_{sol} = G_{gas} + \Delta G_{solv} + \Delta G^{0/*} \quad (2)$$

$$\Delta G^{0/*} = -T\Delta S^{0/*} = RT\ln\left(V^0/V^*\right) = RT\ln(24.46) = 1.89 kcal/mol (T = 298.15K) \quad (3)$$

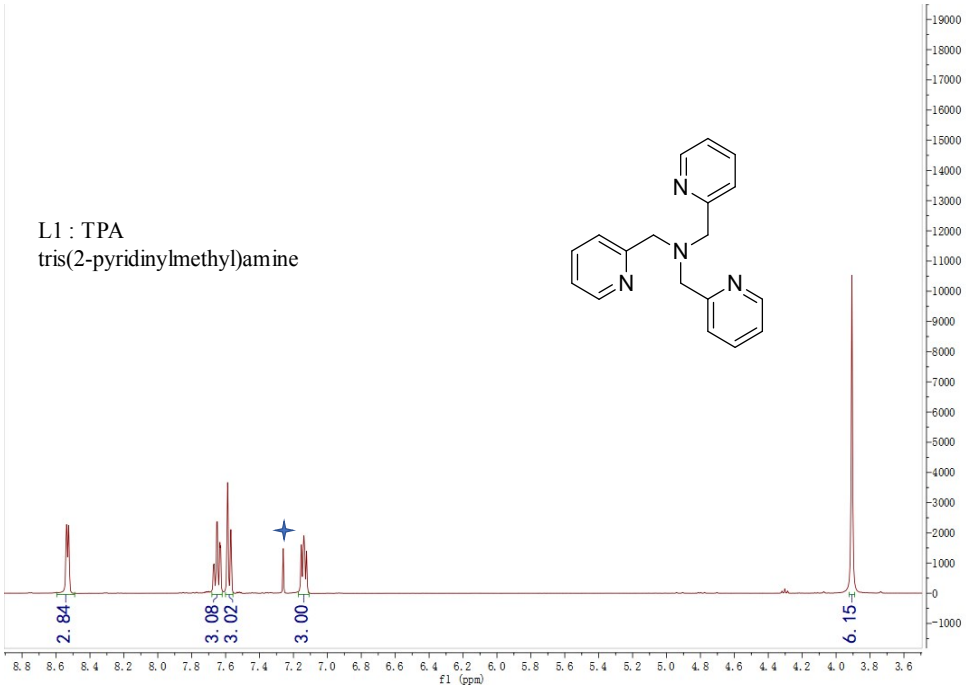


**Figure S1**. $^1$H NMR spectrum of **L1** in $CDCl_3$.

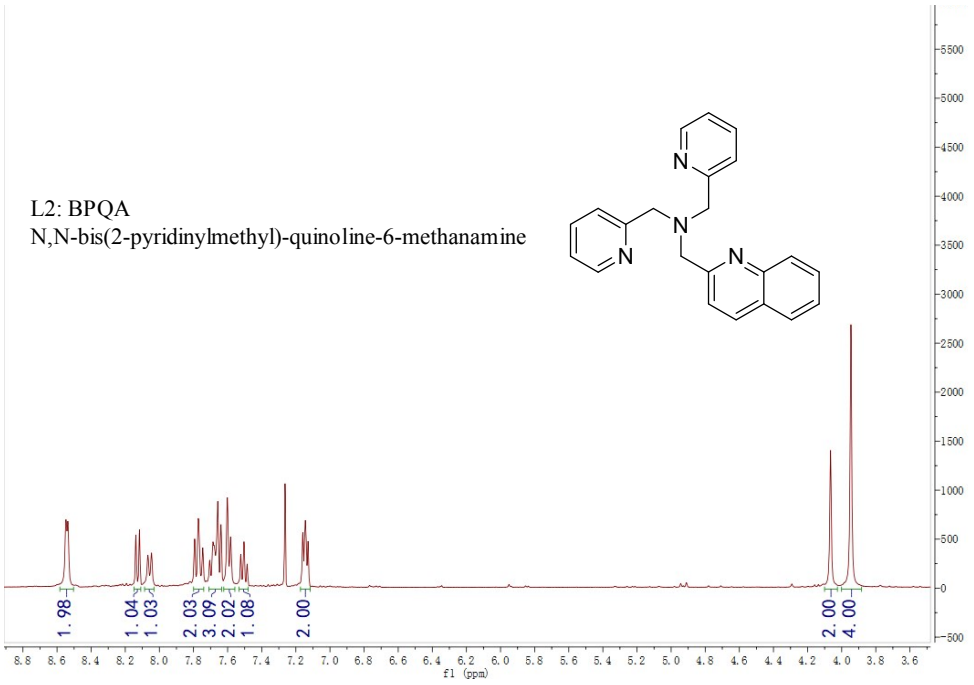


**Figure S2**. $^{1}$H NMR spectrum of **L2** in $CDCl_3$.

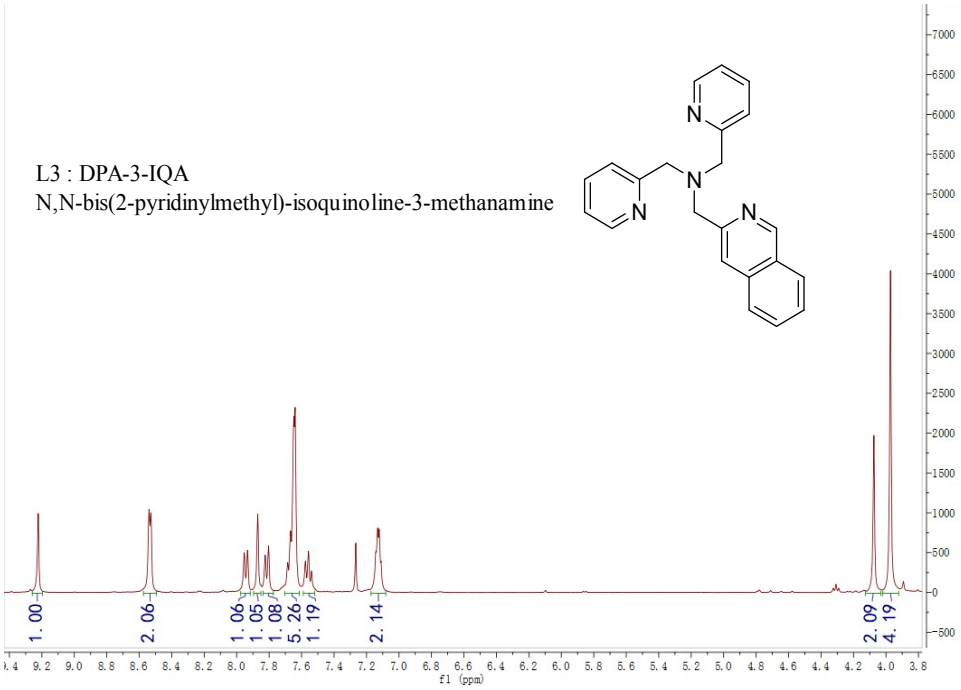


**Figure S3**. $^{1}$H NMR spectrum of **L3** in $CDCl_3$.

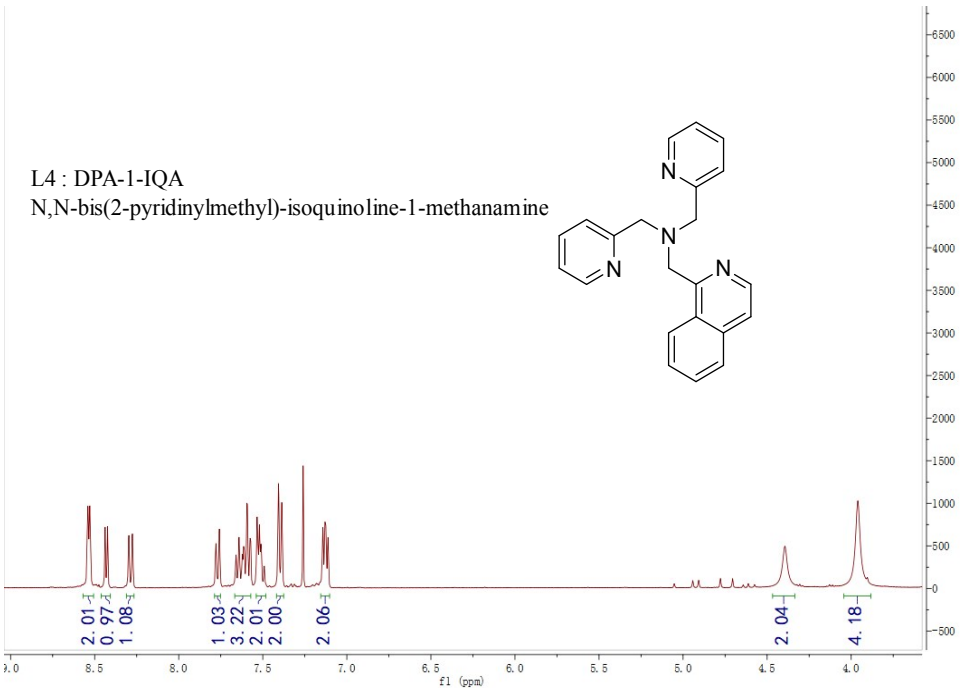


**Figure S4**. $^{1}H$ NMR spectrum of **L4** in $CDCl_3$.

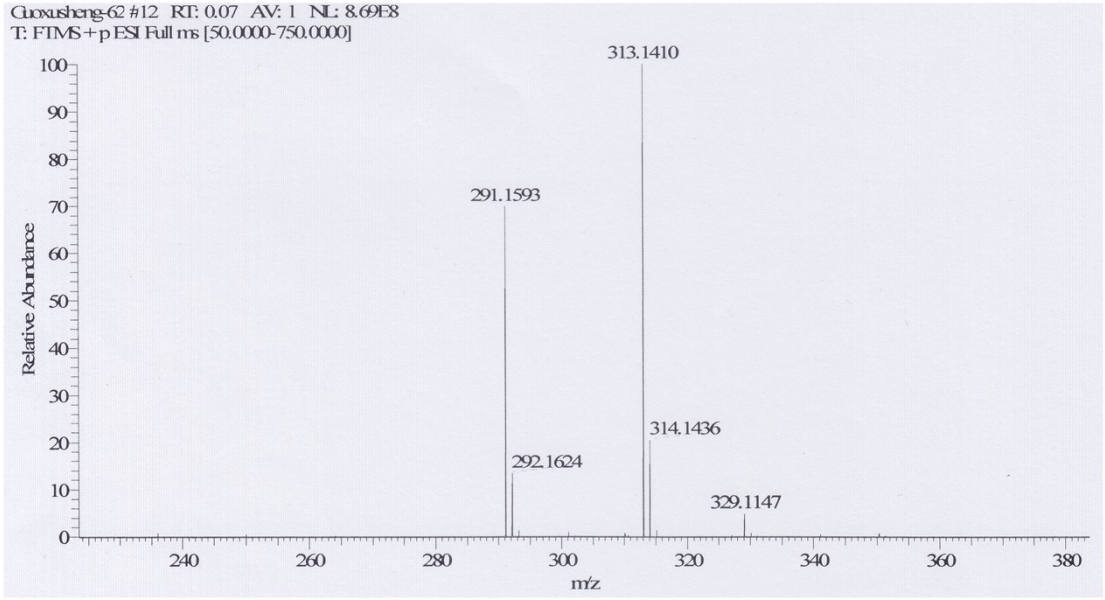


**Figure S5**. ESI-MS spectrum of **L1**.

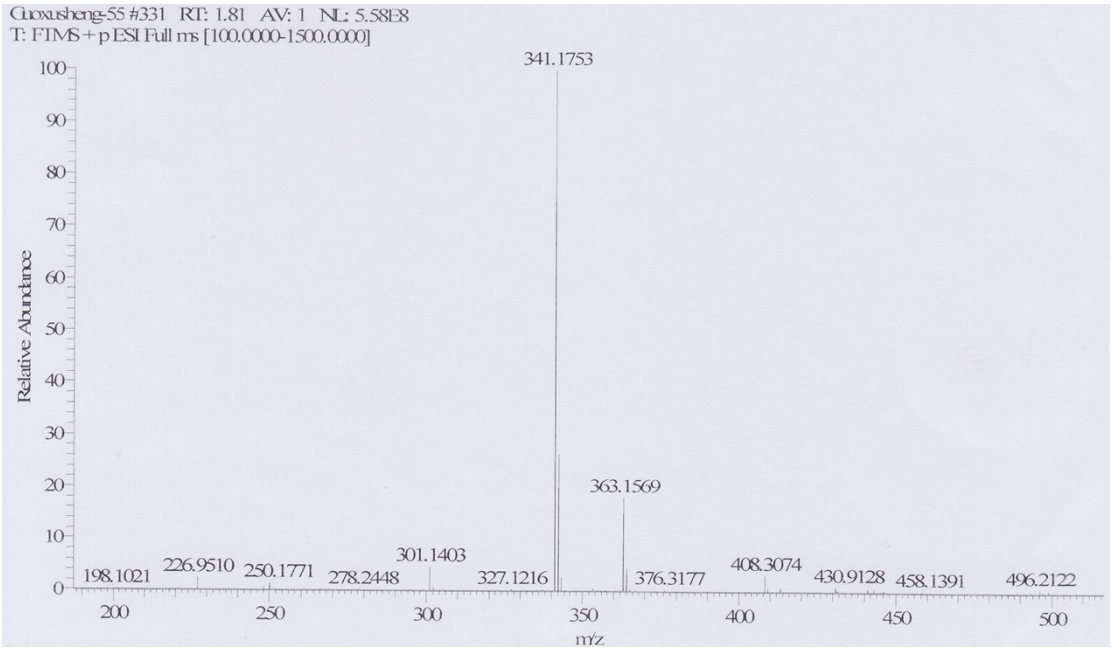


**Figure S6**. ESI-MS spectrum of **L2**.

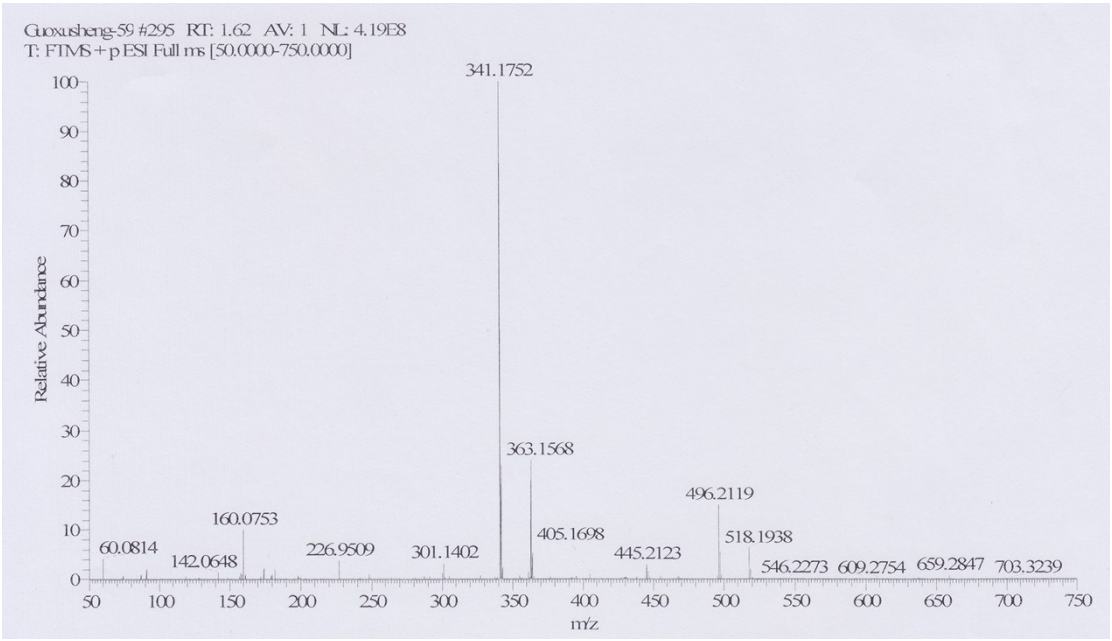


**Figure S7**. ESI-MS spectrum of **L3**.

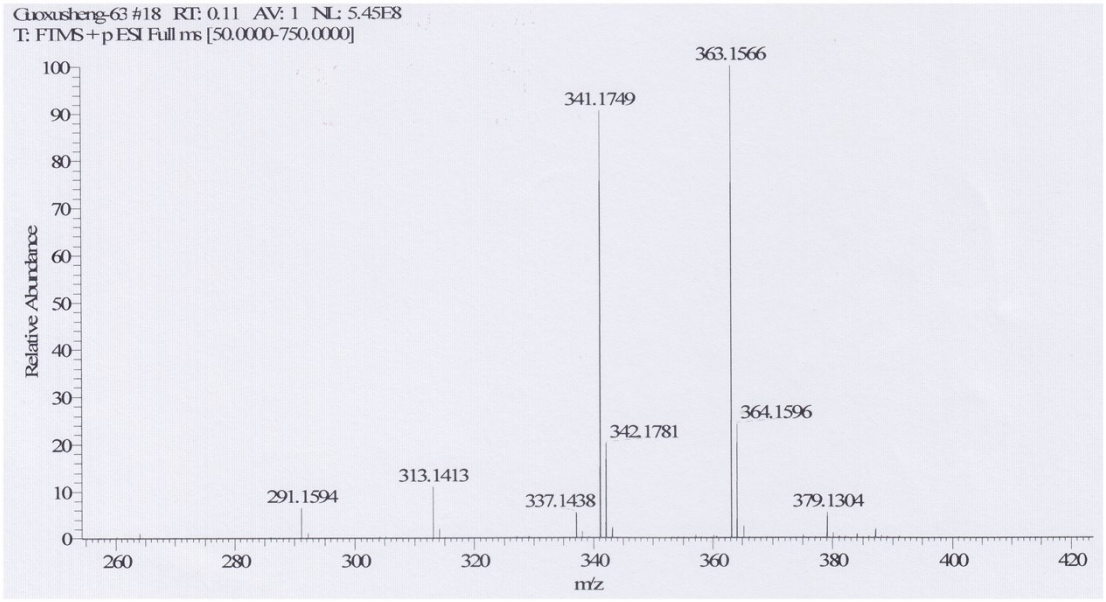


**Figure S8**. ESI-MS spectrum of **L4**.

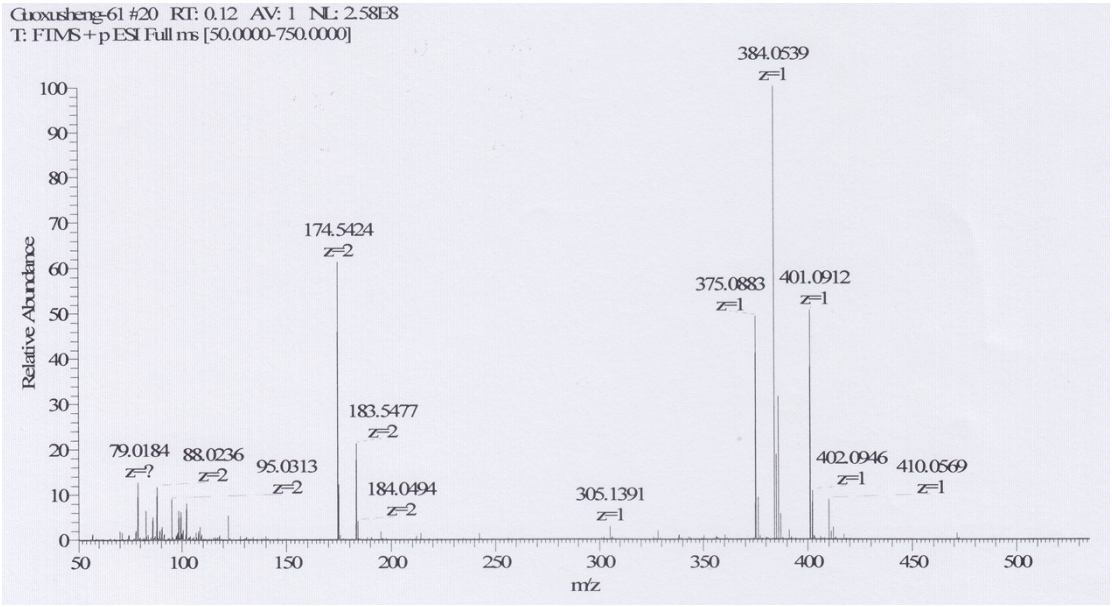


**Figure S9**. ESI-MS spectrum of **C1**.

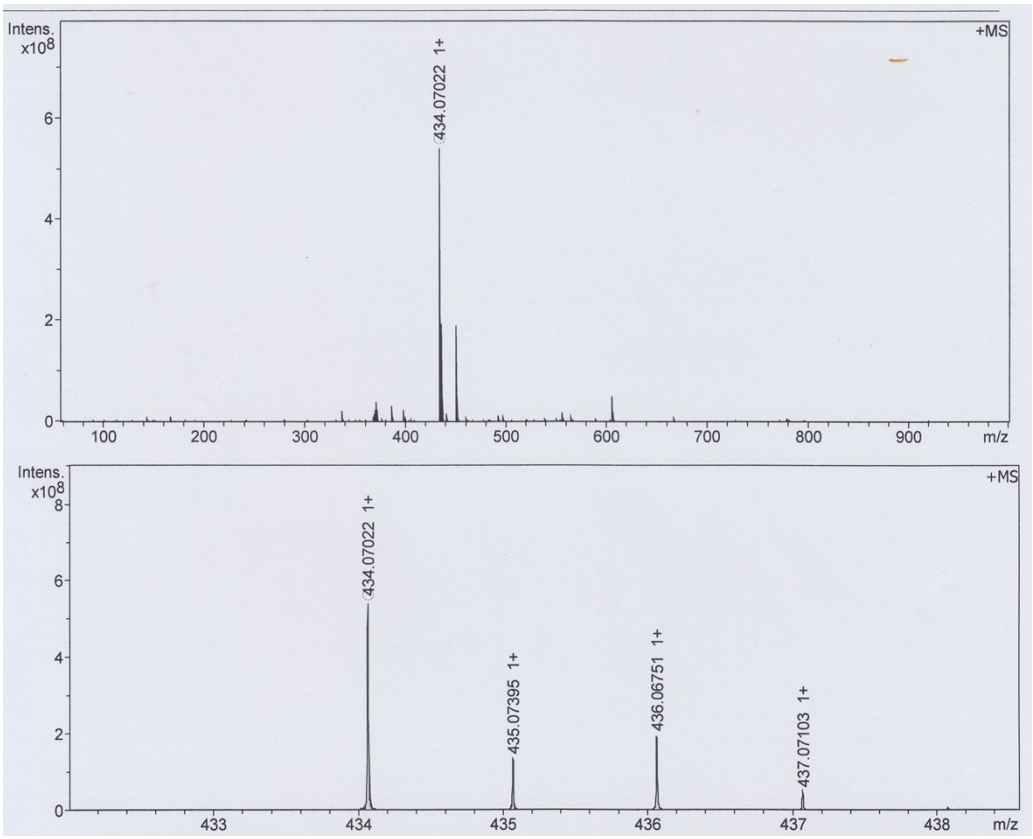


**Figure S10**. ESI-MS spectra of **C2**.

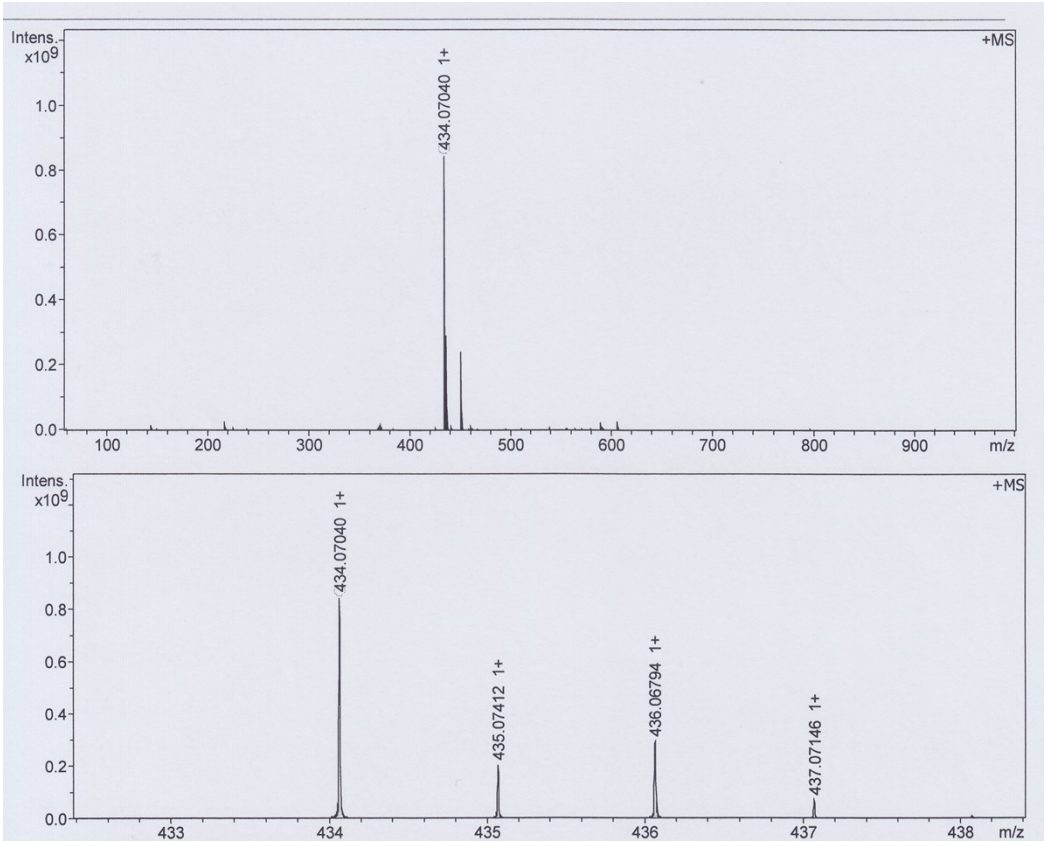


**Figure S11**. ESI-MS spectra of **C3**.

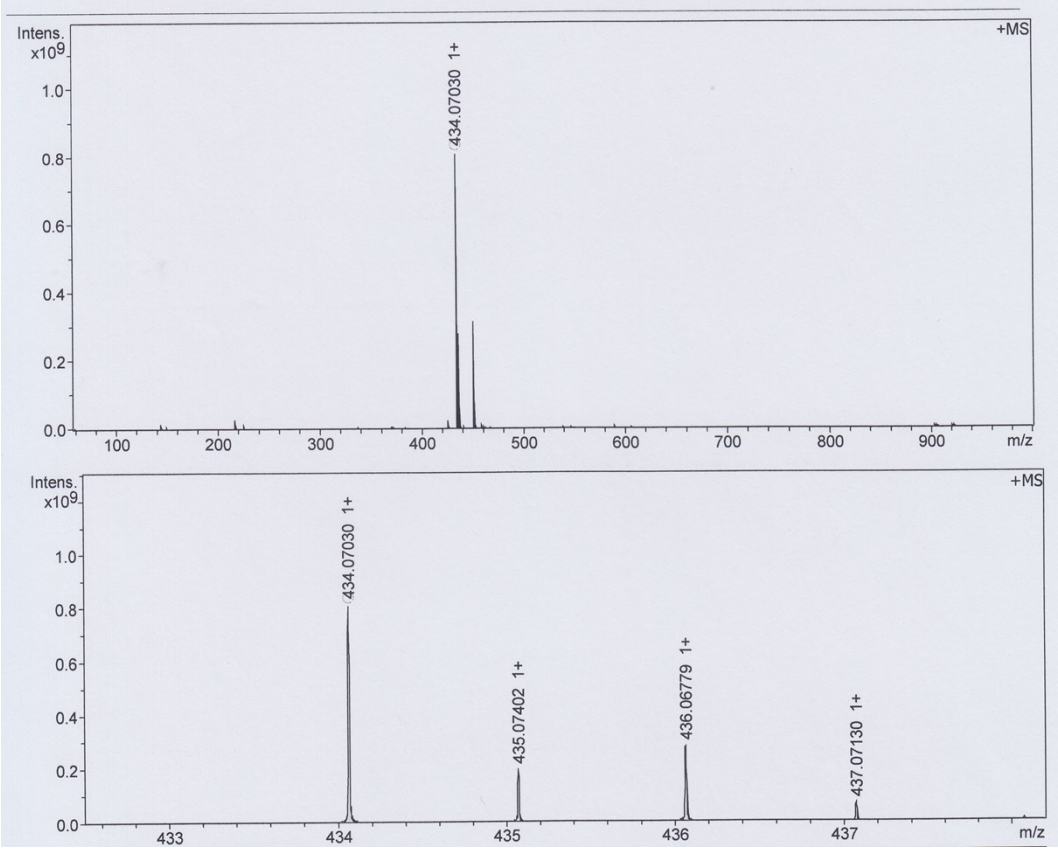


**Figure S12**. ESI-MS spectra of **C4**.

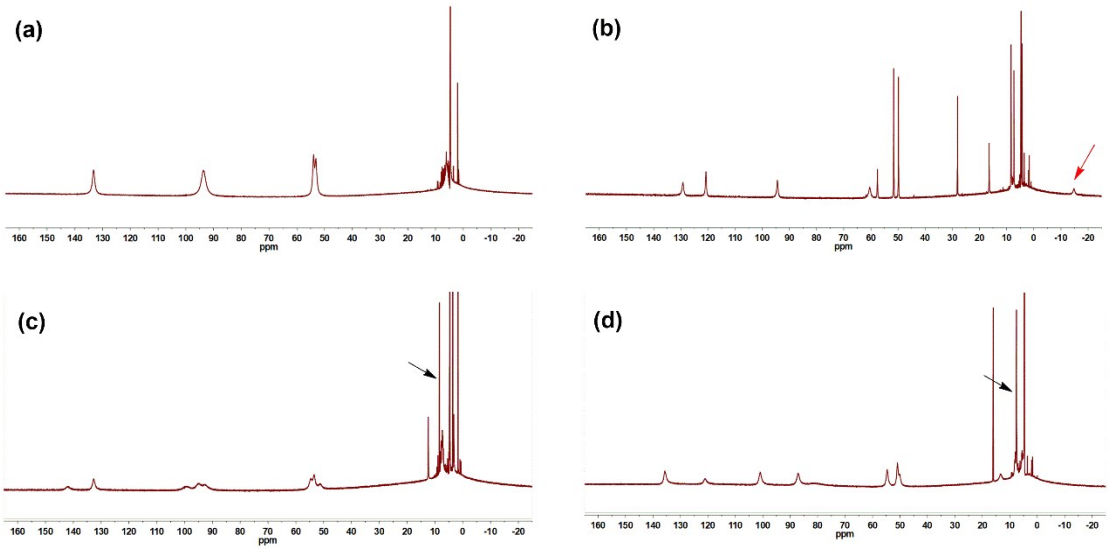


**Figure S13.** $^{1}$H NMR spectra of **C1**-**C4** (**a**-**d**) in D2O. The red arrow in **b** pointing at peaks was assigned to C8-H of quinoline part of complex **C2**, which is 2.885 Å away from cobalt atoms. The black arrows in **c** and **d** pointing at peaks assigned to C6-H of isoquinoline part of Complexes **C3** and **C4**, whose distance between cobalt atoms were 8.178 Å and 8.183 Å, respectively.

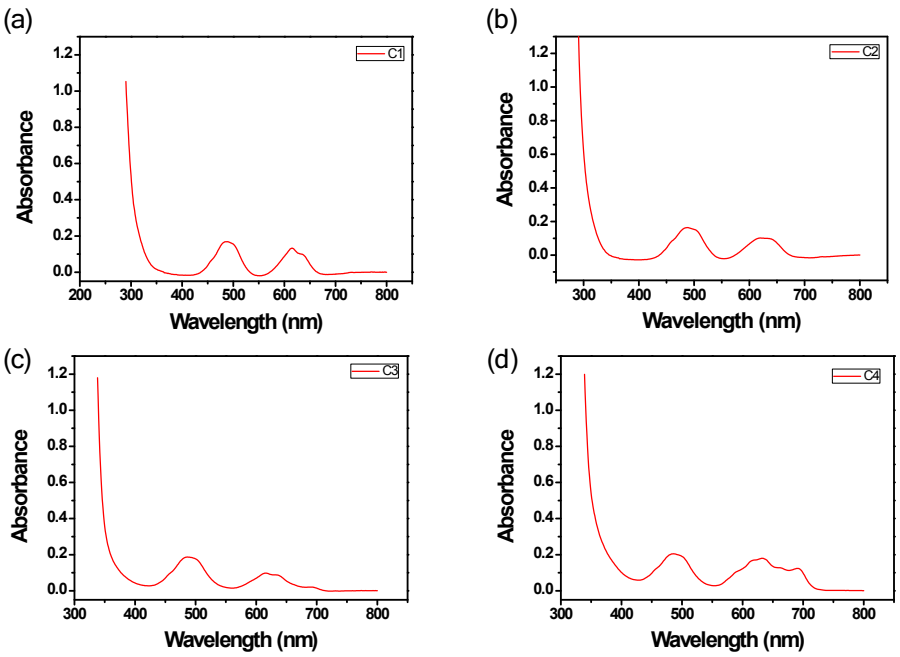


**Figure S14.** UV-vis absorption spectra of 1 mM **C1-C4 (a-d)** in $CH_3CN$.

**Table S1.** Absorption maxima and molar absorptivity (ε) of **C1-C4** in the region of 400-800 nm.

| List of molar absorptivity for all complexes | | | | |
|---|---|---|---|---|
| Complex | C1 | C2 | C3 | C4 |
| $\lambda_{max}$/nm | 487 | 485 | 491 | 484 |
| ε/moL$^{-1}$ ·cm$^{-1}$ | 174 | 167 | 188 | 208 |

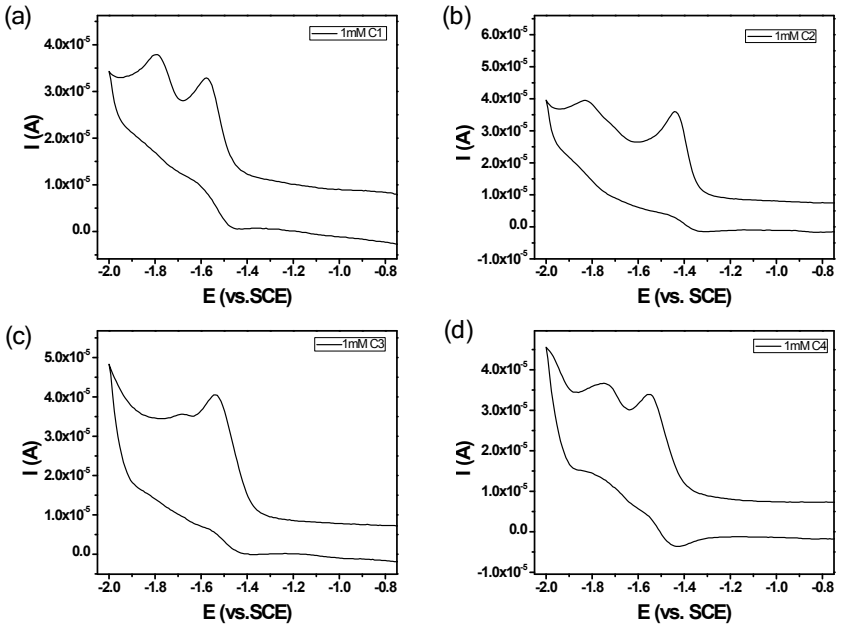


**Figure S15.** Cyclic Voltammograms of 1.0 mM **C1-C4 (a-d)** in 0.1 M $TBAPF_6$ anhydrous MeCN under an argon atmosphere and at a scan rate of 100 mV/s.

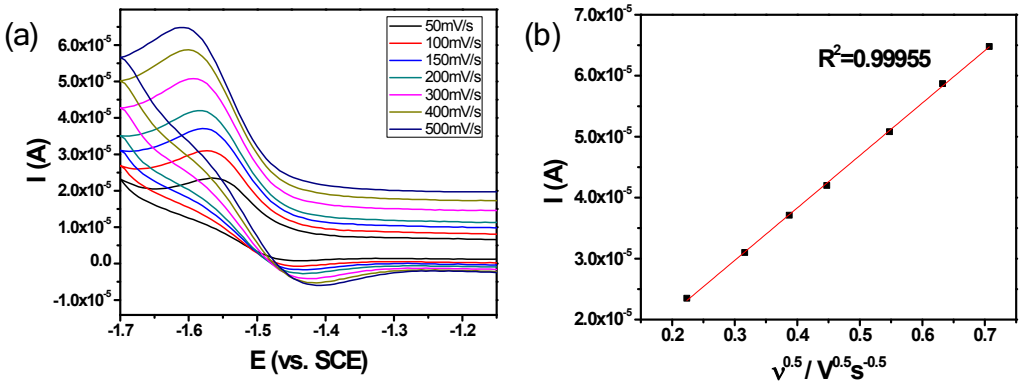


**Figure S16.** (a) CVs of 1.0 mM **C1** in 0.1 M $TBAPF_6$ anhydrous MeCN under an argon atmosphere and at various scan rates. (b) plots of the peak current *vs.* the square root of the scan rate.

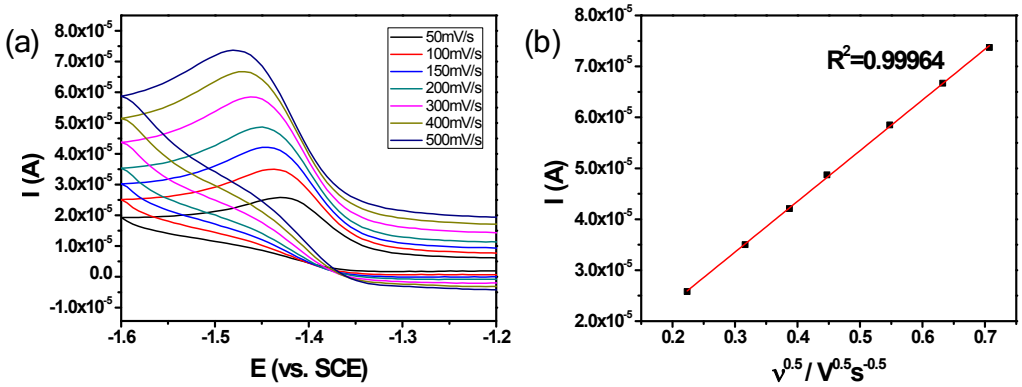


**Figure S17.** (a) CVs of 1.0 mM **C2** in 0.1 M $TBAPF_6$ anhydrous MeCN under an argon atmosphere and at various scan rates. (b) plots of the peak current *vs.* the square root of the scan rate.

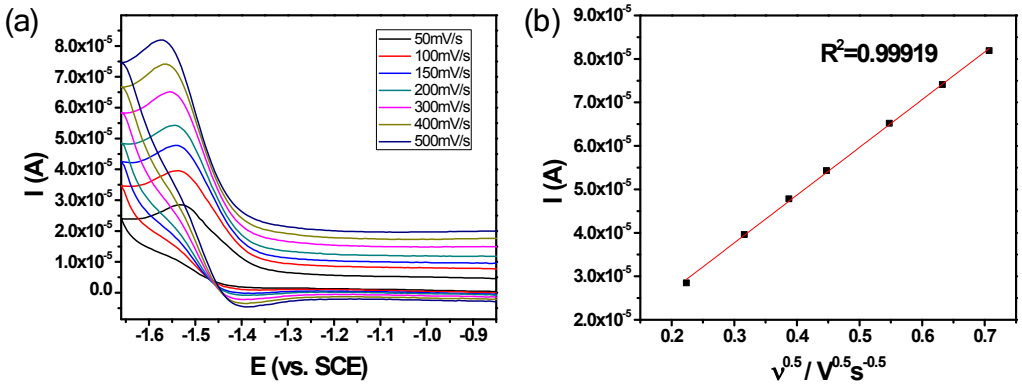


**Figure S18.** (a) CVs of 1.0 mM **C3** in 0.1 M $TBAPF_6$ anhydrous MeCN under an argon atmosphere and at various scan rates. (b) plots of the peak current *vs.* the square root of the scan rate.

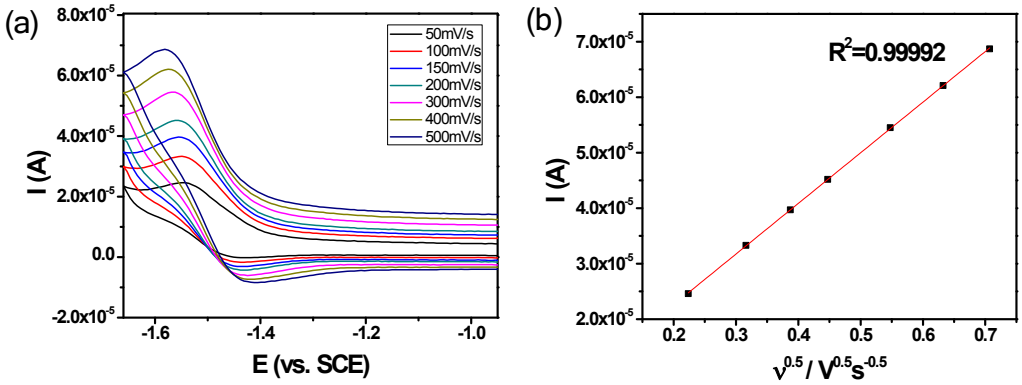


**Figure S19.** (a) CVs of 1.0 mM **C4** in 0.1 M $TBAPF_6$ anhydrous MeCN under an argon atmosphere and at various scan rates. (b) plots of the peak current *vs.* the square root of the scan rate.

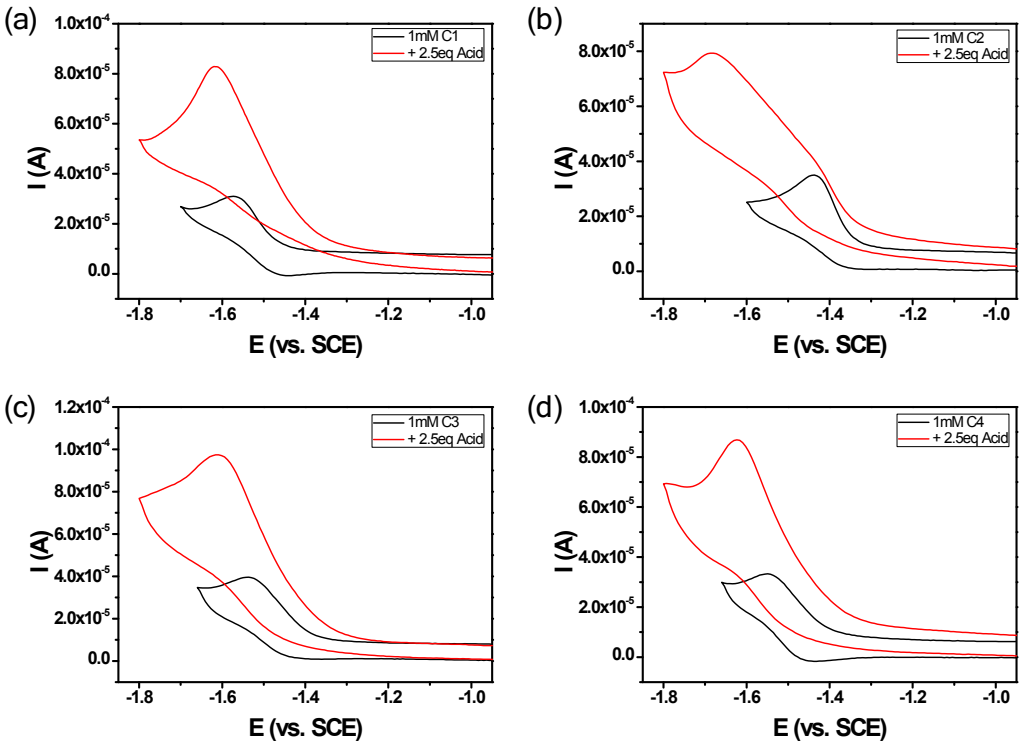


**Figure S20.** CVs of 1.0 mM **C1-C4 (a-d)** in 0.1 M $TBAPF_6$ anhydrous MeCN under an argon atmosphere without (black) or with (red) 2.5 mM acetic acid at a sacn rate of 100 mV/s.

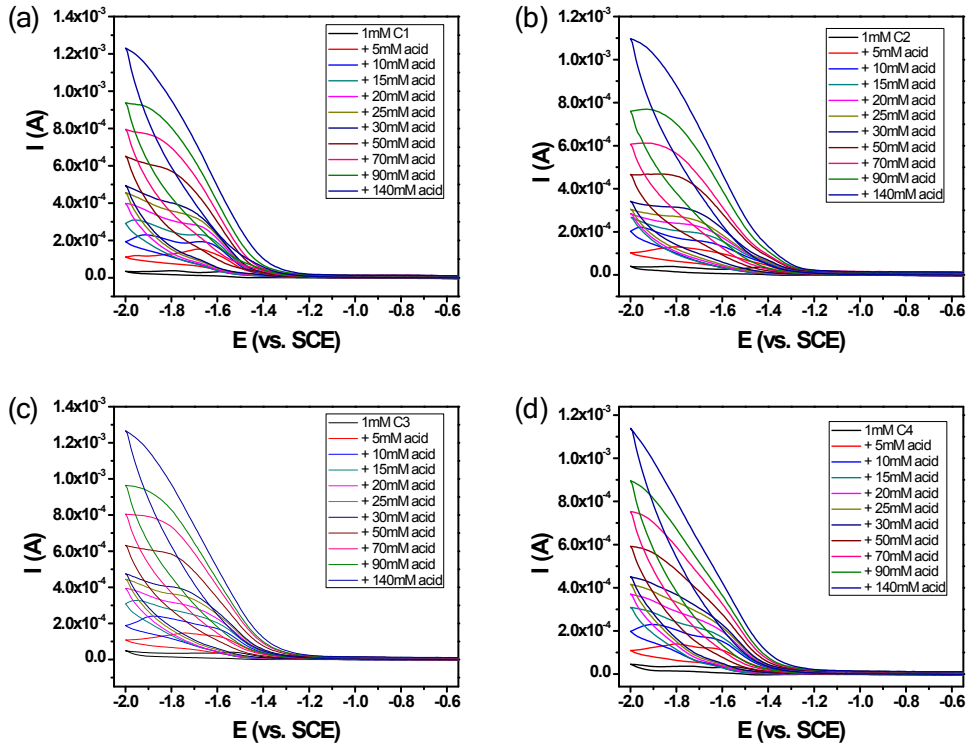


**Figure S21.** CVs of 1.0 mM **C1-C4 (a-d)** in 0.1 M $TBAPF_6$ anhydrous MeCN under an argon atmosphere with increasing concentrations of acetic acid at a sacn rate of 100 mV/s.

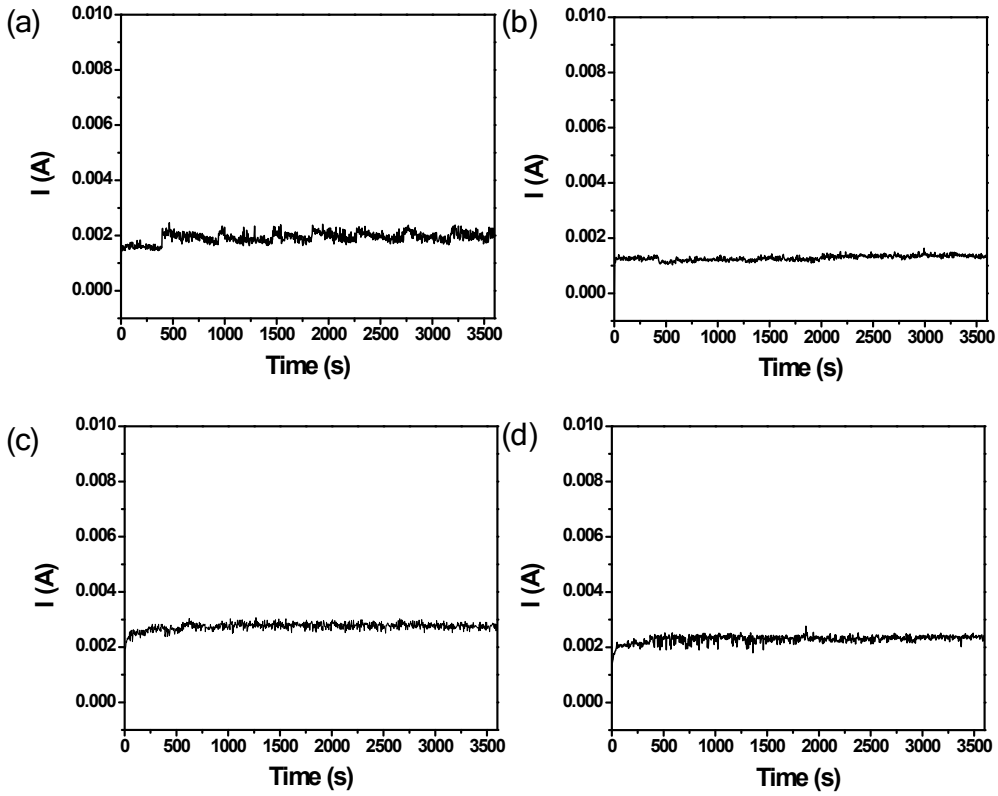


**Figure S22.** Current *vs.* time plots of 1 mM **C1-C4 (a-d)** at an applied potential of -2.0 V *vs.* SCE in 0.1M $TBAPF_6$ MeCN with 140 mM acetic acid under an argon atmosphere.

**Table S2**. Current efficiencies of 1 mM **C1-C4** in controlled-potential electrolysis.

| | Charge /C | $n(H_2)_{tested}$ /mmol | $n(H_2)_{calcd}$ /mmol | Current Efficiency |
|---|---|---|---|---|
| C1 | 6.92 | 0.0322 | 0.0359 | 89.50% |
| C2 | 4.62 | 0.0211 | 0.0239 | 88.10% |
| C3 | 9.89 | 0.0446 | 0.0513 | 86.90% |
| C4 | 8.26 | 0.0381 | 0.0429 | 88.80% |

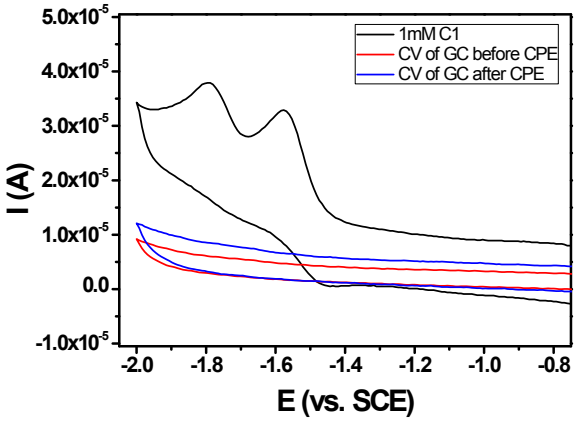


**Figure S23.** CVs of 1.0 mM **C1** (black) and glassy carbon working electrode in the absence of catalyst before (red) and after (blue) electrolysis in 0.1 M $TBAPF_6$ anhydrous MeCN under an argon atmosphere at a scan rate of 100 mV/s. The GC electrode after CPE experiment was rinsed by water and used without further polishing.

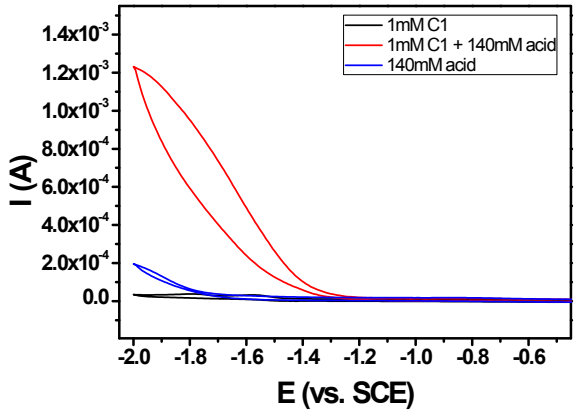


**Figure S24.** CVs of 1.0 mM **C1** (black) and 140 mM acetic acid with (red) or without (blue) 1.0 mM **C1** in 0.1 M $TBAPF_6$ anhydrous MeCN under an argon atmosphere at a scan rate of 100 mV/s.

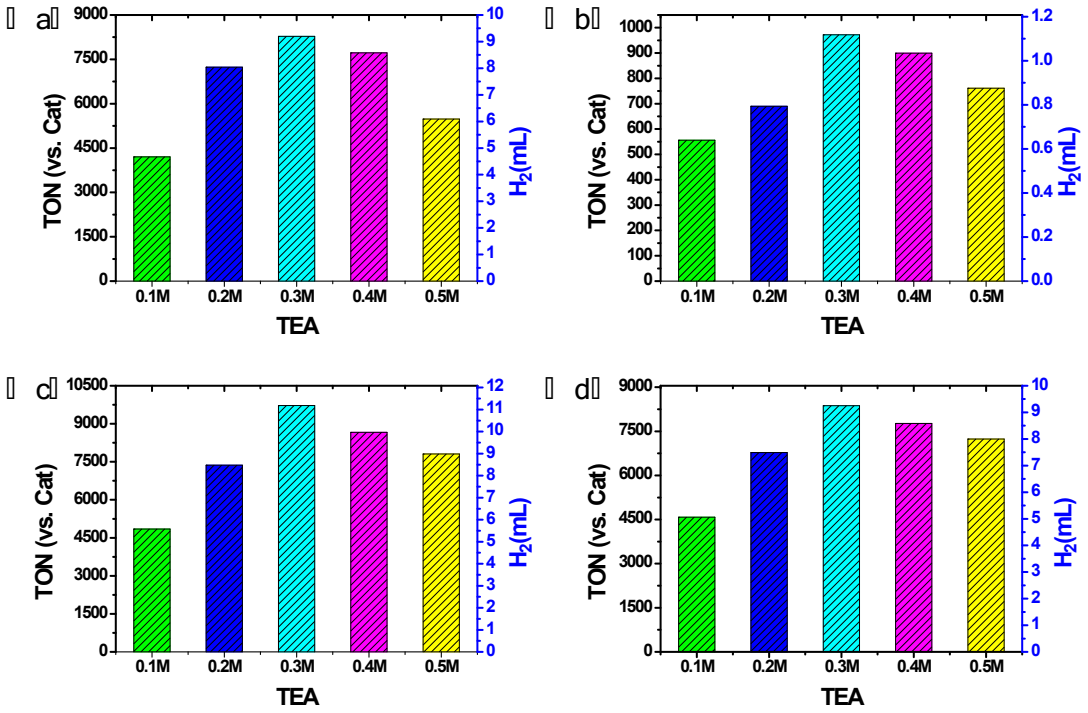


**Figure S25.** Photocatalytic $H_2$ production of **C1-C4 (a-d)** after 6 h irradiation at varied concentrations of TEA. Conditions: 0.2 mM $[Ir(ppy)_2(dtbpy)]Cl$, MeCN/$H_2O$ (8:2), 10 μM catalyst.

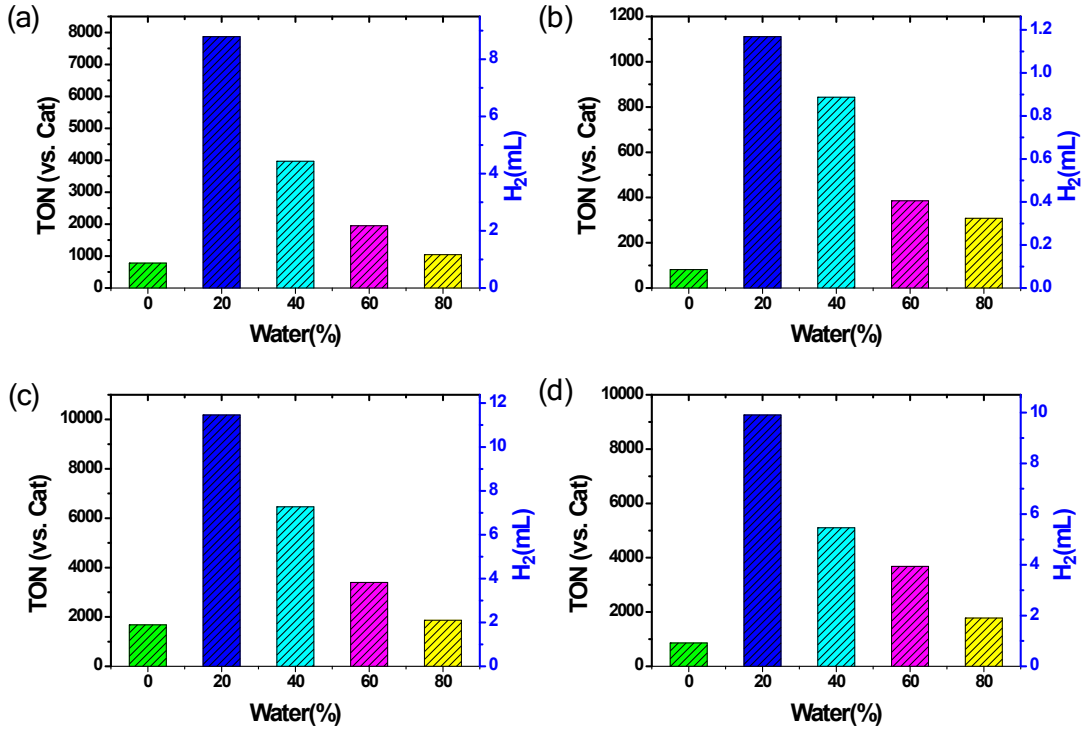


**Figure S26.** Photocatalytic $H_2$ production of **C1-C4 (a-d)** after 6 h irradiation at varied compositions of water in MeCN. Conditions: 0.2 mM $[Ir(ppy)_2(dtbpy)]Cl$, 0.3 M TEA, 10 μM catalyst.

**Table S3.** Photocatalytic $H_2$ production in varied conditions. PS: 0.2 mM of $[Ir(ppy)_2(dtbpy)]Cl$, SD: 0.3 M of TEA, Cat: 10 μM **C1**, Solvent: MeCN/$H_2O$ (8:2), Light: 6 h irradiation by a blue LED (455 nm, 40 W).

| Light | PS | Cat | SD | n($H_2$) / mmol |
|---|---|---|---|---|
| -- | 0.2mM | 10μM | 0.3M | 8.26E-5 |
| 6h | -- | 10μM | 0.3M | 0 |
| 6h | 0.2mM | -- | 0.3M | 5.96E-4 |
| 6h | 0.2mM | 10μM | -- | 0 |
| 6h | 0.2mM | 10μM | 0.3M | 0.403 |

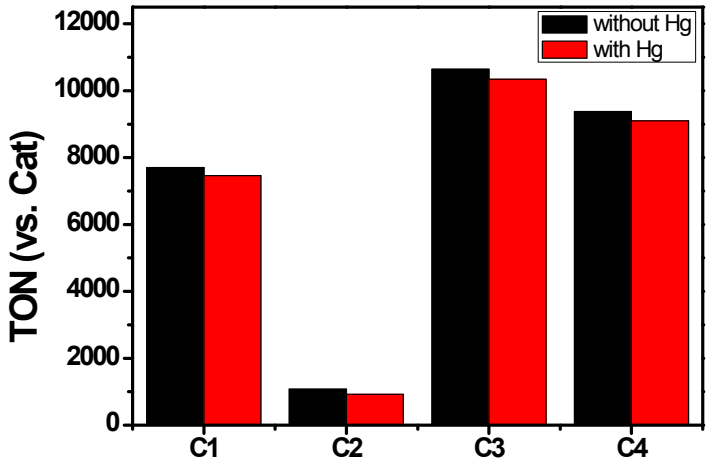


**Figure S27.** Photocatalytic $H_2$ production of **C1-C4** with or without Hg (10% volume) after 6 h irradiation. Conditions: 0.2 mM $[Ir(ppy)_2(dtbpy)]Cl$, 0.3 M TEA, MeCN/$H_2O$ (8:2), 10 μM catalyst.

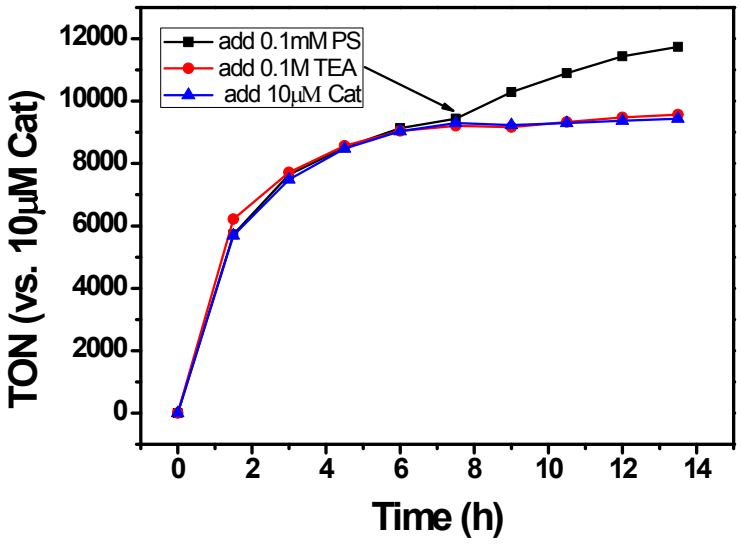


**Figure S28.** Consecutive photocatalytic $H_2$ production by adding 0.1 mM $[Ir(ppy)_2(dtbpy)]Cl$ (black), 0.1 M TEA (red), 10 μM **C3** (blue). Initial condition: 0.2 mM $[Ir(ppy)_2(dtbpy)]Cl$, 0.3 M TEA, MeCN/$H_2O$ (8:2), 10 μM catalyst.

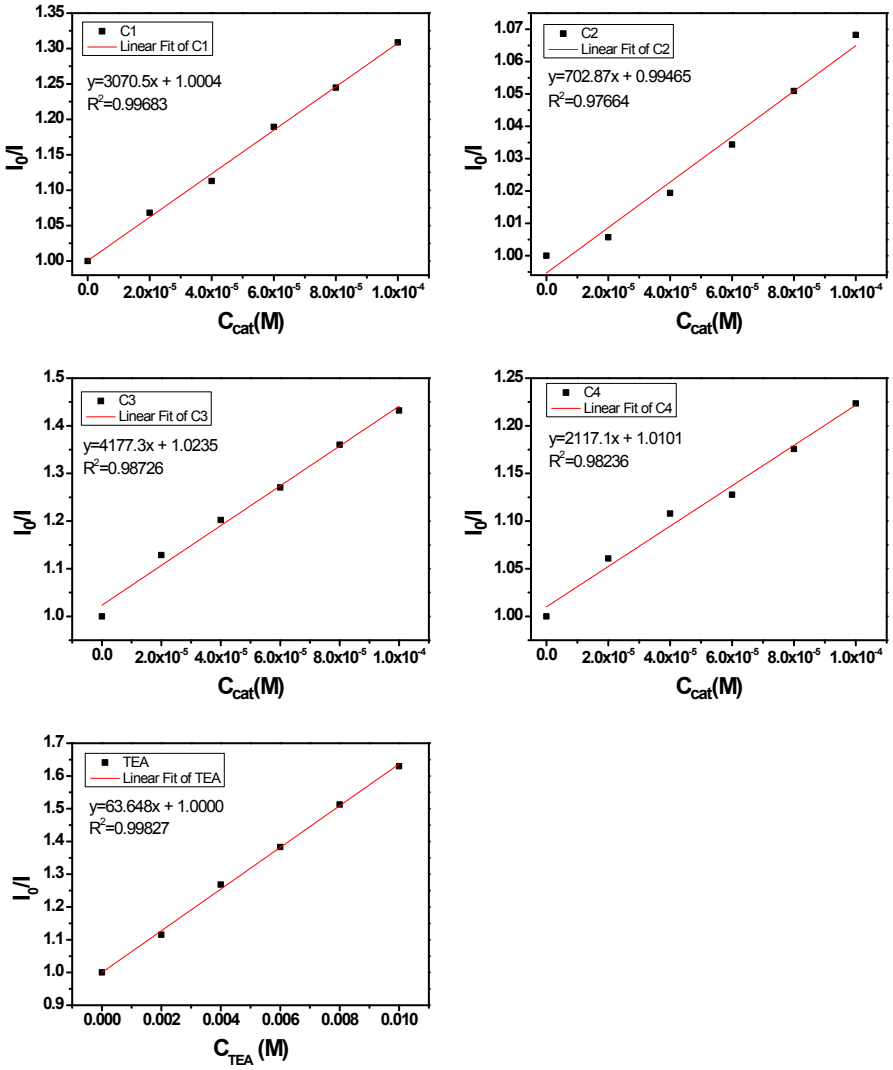


**Figure S29.** Stern-Volmer plots of the luminescence quenching of $[Ir(ppy)_2(dtbpy)]Cl$ by **C1-C4** and **TEA (a-e)** in MeCN/$H_2O$ (8:2).

**Table S4.** Slopes of the Stern-Volmer plots for TEA and **C1-C4** and quenching rate constants.

| | TEA | C1 | C2 | C3 | C4 |
|---|---|---|---|---|---|
| slope | 63.648 | 3070.5 | 702.87 | 4177.3 | 2117.1 |
| $k_q$ ($10^9$ $M^{-1}s^{-1}$) | 0.114 | 5.513 | 1.262 | 7.499 | 3.801 |

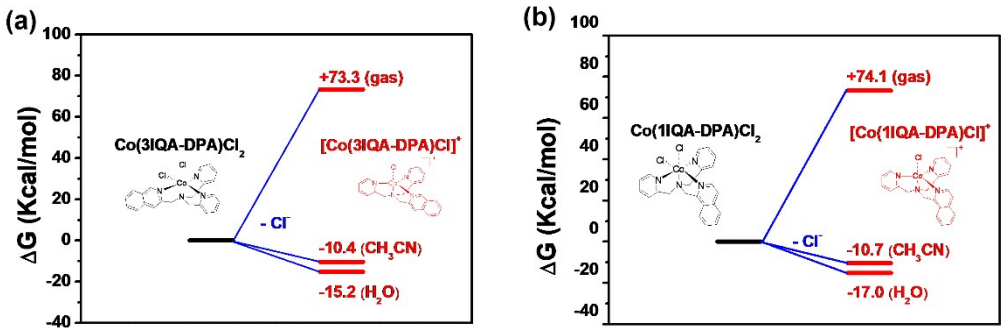


**Figure S30**. Calculated Gibbs free energy changes (in Kcal/mol) during configuration transformation of complexes **C3** and **C4** in different spheres (gas, acetonitrile and water).

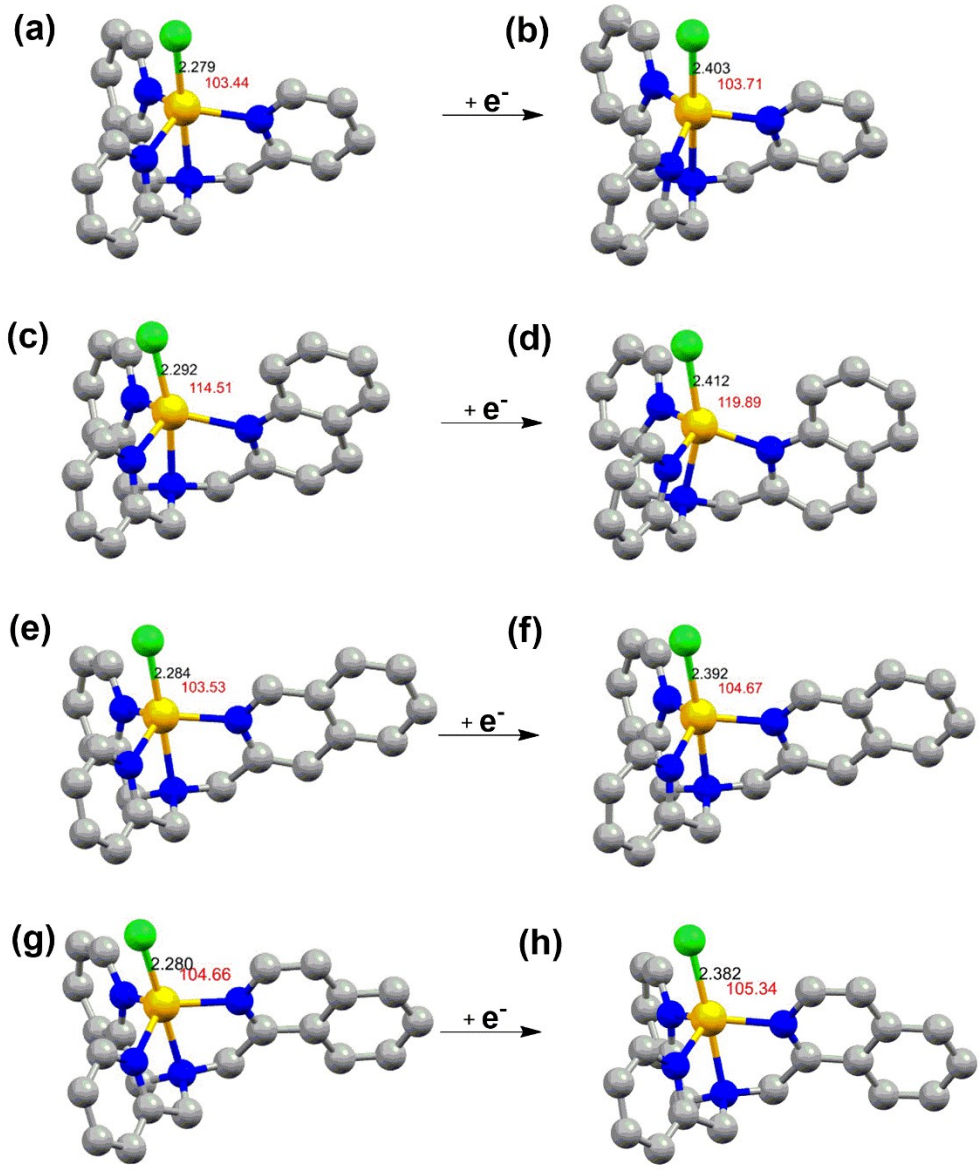


**Figure S31.** Optimized structures of Co(II)-Cl complexes **C1-C4** (**a, c, e, g**) and Co(I)-Cl intermediates of **C1-C4** (**b, d, f, h**), key parameters were shown in corresponding positions and the bond lengths were given in Å, bond angles were given in °.

**Table S5.** Selected bond lengths (Å) and angles (°) from the optimized structures of Co(II)-Cl complexes and Co(I)-Cl intermediates of **C1**-**C4**, N1 atoms refers the N atoms at quinoline (**C2**), isoquinoline (**C3**, **C4**) or pyridine (**C1**).

| | **C1** | **C2** | **C3** | **C4** |
|---|---|---|---|---|
| **Co(II)-Cl /Å** | 2.279 | 2.292 | 2.284 | 2.28 |
| **N1-Co-Cl /°** | 103.44 | 114.51 | 103.53 | 104.66 |
| **Co(I)-Cl /Å** | 2.403 | 2.412 | 2.392 | 2.382 |
| **N1-Co-Cl /°** | 103.71 | 119.89 | 104.67 | 105.34 |

**Table S6.** Calculated Gibbs free energy changes (in Kcal/mol) during corresponding bond dissociation from Co(I)-Cl intermediates of **C1**-**C4**. Nqa in the table refers the N atoms at quinoline (**C2**), isoquinoline (**C3**, **C4**) and Npy refers the N atoms at pyridine.

| **Bonds dissociation** | **C1** | **C2** | **C3** | **C4** |
|---|---|---|---|---|
| **Co-Npy** | 3.97 | 0.11 | 4.65 | 4.07 |
| **Co-Nqa** | \ | 0.38 | 2.78 | 6.18 |
| **Co-Cl** | -4.93 | -6.34 | -4.53 | -4.35 |

**Table S7.** Selected bond lengths (Å) and angles (°) of the DFT optimized Co(I) intermediates of **C1**-**C4**. N1 is the N atom in pyridine, quinoline or isoquinoline group; N2 and N3 is the N atoms in the rest of pyridine groups.

| | **Co-N1** | **Co-N2** | **Co-N3** | **N1-Co-N4** | **N2-Co-N4** | **N3-Co-N4** |
|---|---|---|---|---|---|---|
| **$[Co^{I}(TPA)]^{+}$** | 2.05382 | 2.06454 | 2.08442 | 84.23667 | 82.02955 | 82.23636 |
| **$[Co^{I}(BPQA)]^{+}$** | 2.05272 | 2.06301 | 2.08825 | 84.07570 | 82.19080 | 82.21601 |
| **$[Co^{I}(DPA\text{-}3\text{-}IQA)]^{+}$** | 2.05159 | 2.06985 | 2.08127 | 84.71457 | 81.84394 | 82.27962 |
| **$[Co^{I}(DPA\text{-}1\text{-}IQA)]^{+}$** | 2.04032 | 2.05386 | 2.08977 | 83.48750 | 82.88224 | 82.46036 |

**Table S8.** Calculated Gibbs free energies (in Hartrees) in aqueous sphere ($G_{sol}$) and gas sphere ($G_{gas}$) of the Co(III)-H species of **C1**-**C4**.

| **Species** | **Spin State** | **Ggas** | **Gsol** | **Spin State** | **Ggas** | **Gsol** |
|---|---|---|---|---|---|---|
| **$[Co^{III}\text{-}H(TPA)]^{2+}$** | **singlet** | -1060.880506 | -1061.130235 | **triplet** | -1060.895631 | -1061.134257 |
| **$[Co^{III}\text{-}H(BPQA)]^{2+}$** | | -1214.519600 | -1214.761775 | | -1214.538023 | -1214.768029 |
| **$[Co^{III}\text{-}H(DPA\text{-}3\text{-}IQA)]^{2+}$** | | -1214.525607 | -1214.765150 | | -1214.541488 | -1214.771644 |
| **$[Co^{III}\text{-}H(DPA\text{-}1\text{-}IQA)]^{2+}$** | | -1214.522212 | -1214.764665 | | -1214.536807 | -1214.768243 |

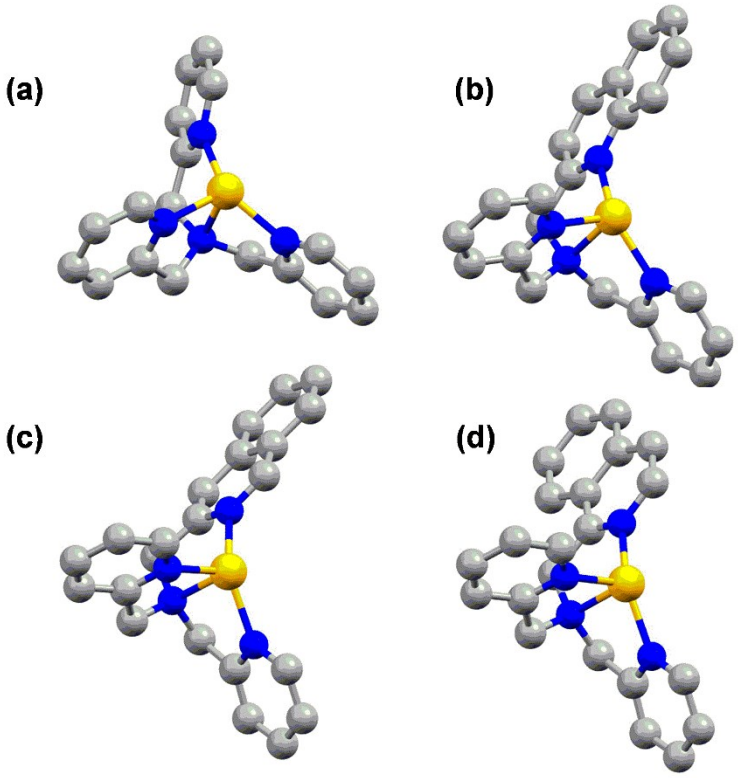


**Figure S32.** Optimized structures for the Co(I) intermediates of **C1-C4 (a-d)**

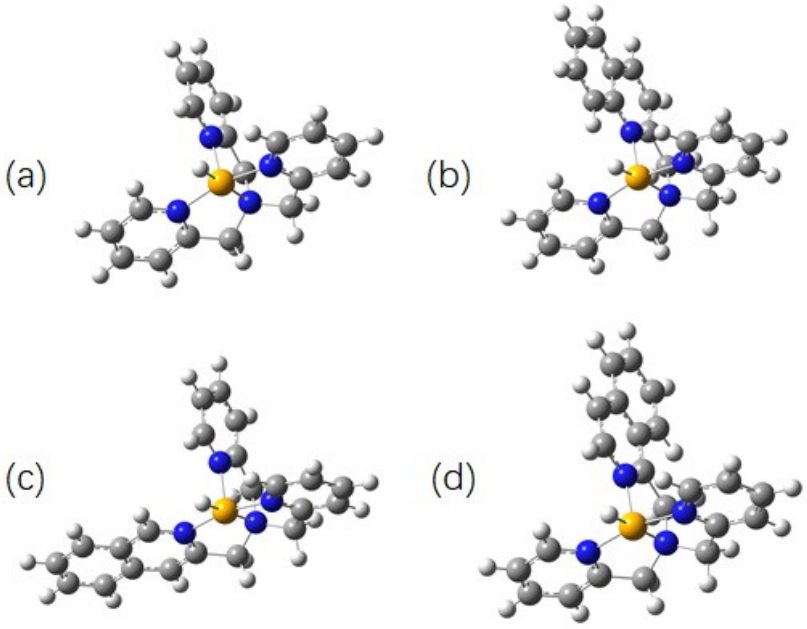


**Figure S33.** Optimized structures of singlet Co(III)-H intermediates of **C1-C4 (a-d)**, all structures were optimized from original distorted trigonal-bipyramidal geometry.

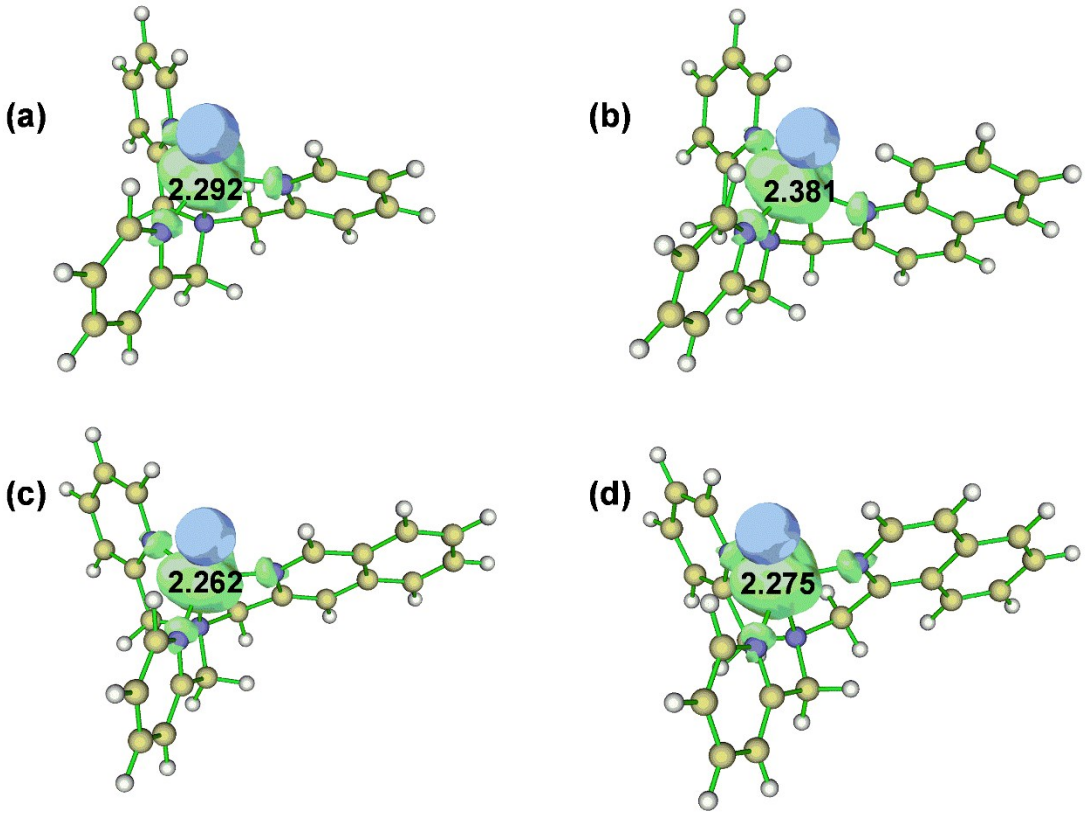


**Figure S34.** Optimized structures and spin density distributions for the Co(III)-H intermediates of **C1**-**C4** (**a**-**d**). The spin population values on cobalt atoms are given.

**Table S9.** Crystallographic data of **C3** and **C4**.

| **Complexes** | | **C3** | **C4** |
|---|---|---|---|
| **CCDC number** | | 2053060 | 2053061 |
| **Bond precision** | | C-C=0.0190A | C-C = 0.0076A |
| **Wavelength** | | 0.71073 | 0.71073 |
| **Unit cell dimensions** | **a** | 17.0415(16) | 10.8073(13) |
| | **b** | 8.9433(9) | 12.9956(15) |
| | **c** | 16.9729(19) | 15.6175(19) |
| | **alpha** | 90 | 90 |
| | **beta** | 90.314(4) | 93.956(5) |
| | **gamma** | 90 | 90 |
| **Temperature** | | 273K | 273K |
| **Volume** | | 2586.8(5) | 2188.2(5) |

| | | |
|---|---|---|
| **Space group** | C 1 2 1 | p 1 21/c 1 |
| **Hall group** | C 2y | -p 2ybc |
| **Moiety formula** | C22 H22 Cl2 Co N4 | C22 H22 Cl2 Co N4 |
| **Sum formula** | C22 H22 Cl2 Co N4 | C22 H20 Cl2 Co N4 |
| **Mr** | 470.25 | 470.25 |
| **Dx, g cm-3** | 1.207 | 1.427 |
| **z** | 4 | 4 |
| **Mu (mm-1)** | 0.883 | 1.044 |
| **F000** | 964.0 | 964.0 |
| **h, k, lmax** | 22, 11, 22 | 14, 16, 20 |
| **Nref** | 5946 | 4943 |
| **Tmin, Tmax** | 0.668, 0.746 | 0.520, 0.746 |
| **Data completeness** | 1.87/1.00 | 0.979 |
| **Theta(max)** | 27.509 | 27.556 |
| **R(reflections)** | 0.0682( 5234) | 0.0716( 3186) |
| **wR2(reflections)** | 0.1968( 5946) | 0.1349( 4943) |
| **S** | 1.052 | 1.080 |
| **Npar** | 263 | 262 |

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## Appendix: DFT calculated coordinates (xyz) of all the complexes and selected intermediates.

**Complex C1**

**$[Co^{II}(TPA)Cl]^+$**

| | | | |
|---|---|---|---|
| Co | -0.00079064 | 0.00161878 | 0.75365687 |
| N | 0.00011226 | 0.00111510 | -1.55413219 |
| C | -4.09111548 | 1.50940597 | 0.82236121 |
| H | -4.73901835 | 1.92655259 | 1.58234802 |
| C | -2.37138340 | 0.43018612 | -1.02310844 |
| N | 1.47464770 | 1.41665846 | 0.25437239 |
| N | 0.48948543 | -1.98349696 | 0.25330045 |
| C | 1.55931289 | 1.83723952 | -1.02226820 |
| C | -2.80906966 | 1.09520652 | 1.15965379 |
| H | -2.42619034 | 1.16977658 | 2.17074645 |
| C | -1.39460650 | -0.25159707 | -1.95872918 |
| H | -1.56792689 | -1.33080627 | -1.90124168 |
| H | -1.58146829 | 0.04645440 | -2.99822209 |
| N | -1.96575908 | 0.56882400 | 0.25355268 |
| C | 3.45495333 | 3.20836232 | -0.49564926 |
| H | 4.23009067 | 3.90456230 | -0.79355792 |
| C | -3.63644295 | 0.83346058 | -1.43567924 |
| H | -3.93290645 | 0.71806677 | -2.47139995 |
| C | 0.45301296 | -2.97632768 | 1.16019670 |
| C | 2.54421679 | 2.72793290 | -1.43397798 |
| H | 2.59409449 | 3.04261498 | -2.46955859 |
| C | 0.81365908 | -2.26678947 | -1.02275851 |
| C | 3.35647460 | 2.78016921 | 0.82445518 |
| H | 4.04273684 | 3.12990123 | 1.58476845 |
| C | 0.73523941 | -4.29415875 | 0.82462655 |
| C | 1.09668601 | -3.56456454 | -1.43360869 |
| H | 1.34564996 | -3.76501191 | -2.46887922 |
| C | 2.35302932 | 1.88071868 | 1.16107467 |
| H | 2.22333214 | 1.51197209 | 2.17188378 |
| C | 0.91512292 | -1.08107940 | -1.96002798 |
| H | 1.93714416 | -0.69276675 | -1.90736876 |
| H | 0.74695151 | -1.39329098 | -2.99861254 |
| C | -4.51023314 | 1.37888697 | -0.49790121 |
| H | -5.50182532 | 1.69840423 | -0.79610879 |
| C | 1.05993664 | -4.59305483 | -0.49480029 |
| H | 1.27932641 | -5.61183313 | -0.79184714 |
| C | 0.47974878 | 1.33480186 | -1.95878090 |
| H | -0.36709987 | 2.02596746 | -1.90233967 |
| H | 0.83200566 | 1.34738683 | -2.99811179 |
| Cl | -0.00149438 | 0.00105782 | 3.00840603 |
| H | 0.19489012 | -2.68104144 | 2.17047720 |
| H | 0.69631147 | -5.06309647 | 1.58526373 |

**$[Co^{I}(TPA)Cl]$**

| | | | |
|---|---|---|---|
| Co | 0.057000 | -0.026000 | 1.009000 |
| N | 0.044000 | 0.081000 | -1.304000 |
| C | -3.635000 | 2.264000 | 1.263000 |
| H | -4.184000 | 2.748000 | 2.065000 |
| C | -2.178000 | 0.985000 | -0.682000 |
| N | 1.648000 | 1.255000 | 0.667000 |
| N | -0.084000 | -2.060000 | 0.425000 |
| C | 1.956000 | 1.499000 | -0.636000 |
| C | -2.482000 | 1.540000 | 1.555000 |
| H | -2.098000 | 1.434000 | 2.566000 |
| C | -1.373000 | 0.170000 | -1.681000 |
| H | -1.776000 | -0.850000 | -1.687000 |

| | | | |
|---|---|---|---|
| H | -1.505000 | 0.570000 | -2.701000 |
| N | -1.757000 | 0.915000 | 0.603000 |
| C | 4.197000 | 2.179000 | -0.063000 |
| H | 5.183000 | 2.533000 | -0.349000 |
| C | -3.311000 | 1.706000 | -1.053000 |
| H | -3.606000 | 1.751000 | -2.099000 |
| C | -0.377000 | -3.071000 | 1.265000 |
| C | 3.210000 | 1.951000 | -1.032000 |
| H | 3.412000 | 2.128000 | -2.086000 |
| C | 0.251000 | -2.344000 | -0.853000 |
| C | 3.880000 | 1.930000 | 1.277000 |
| H | 4.608000 | 2.090000 | 2.067000 |
| C | -0.374000 | -4.404000 | 0.859000 |
| C | 0.260000 | -3.653000 | -1.337000 |
| H | 0.518000 | -3.847000 | -2.375000 |
| C | 2.607000 | 1.468000 | 1.600000 |
| H | 2.309000 | 1.247000 | 2.621000 |
| C | 0.687000 | -1.169000 | -1.716000 |
| H | 1.770000 | -1.031000 | -1.591000 |
| H | 0.518000 | -1.399000 | -2.783000 |
| C | -4.059000 | 2.359000 | -0.067000 |
| H | -4.946000 | 2.927000 | -0.332000 |
| C | -0.057000 | -4.702000 | -0.470000 |
| H | -0.054000 | -5.729000 | -0.825000 |
| C | 0.815000 | 1.294000 | -1.615000 |
| H | 0.126000 | 2.142000 | -1.522000 |
| H | 1.190000 | 1.294000 | -2.653000 |
| H | -0.607000 | -2.769000 | 2.283000 |
| H | -0.621000 | -5.187000 | 1.570000 |
| Cl | 0.054000 | -0.261000 | 3.400000 |

**[Co$^{I}$(TPA)Cl] with dissociated Co-Npy bond**

| | | | |
|---|---|---|---|
| Co | 0.488000 | 0.586000 | 0.775000 |
| N | -0.034000 | -0.209000 | -1.285000 |
| C | -3.417000 | -3.383000 | 0.766000 |
| H | -4.267000 | -3.832000 | 1.271000 |
| C | -1.331000 | -2.266000 | -0.621000 |
| N | -1.018000 | 1.892000 | 0.307000 |
| N | 2.331000 | -0.017000 | 0.159000 |
| C | -1.405000 | 1.862000 | -0.991000 |
| C | -3.404000 | -3.268000 | -0.626000 |
| H | -4.242000 | -3.627000 | -1.221000 |
| C | -0.203000 | -1.679000 | -1.447000 |
| H | 0.747000 | -2.135000 | -1.150000 |
| H | -0.387000 | -1.931000 | -2.503000 |
| N | -2.391000 | -2.720000 | -1.313000 |
| C | -2.311000 | 4.091000 | -0.830000 |
| H | -2.807000 | 4.948000 | -1.278000 |
| C | -1.251000 | -2.340000 | 0.777000 |
| H | -0.391000 | -1.943000 | 1.310000 |
| C | 3.403000 | -0.295000 | 0.931000 |
| C | -2.038000 | 2.953000 | -1.592000 |
| H | -2.323000 | 2.903000 | -2.640000 |
| C | 2.441000 | -0.148000 | -1.186000 |
| C | -1.936000 | 4.105000 | 0.518000 |
| H | -2.137000 | 4.964000 | 1.151000 |
| C | 4.620000 | -0.704000 | 0.393000 |
| C | 3.624000 | -0.564000 | -1.793000 |
| H | 3.672000 | -0.669000 | -2.874000 |
| C | -1.286000 | 2.993000 | 1.042000 |
| H | -0.960000 | 2.946000 | 2.078000 |
| C | 1.202000 | 0.241000 | -1.968000 |
| H | 1.154000 | 1.337000 | -2.035000 |
| H | 1.257000 | -0.144000 | -3.000000 |
| C | -2.317000 | -2.902000 | 1.479000 |
| H | -2.283000 | -2.957000 | 2.564000 |
| C | 4.738000 | -0.843000 | -0.994000 |
| H | 5.672000 | -1.167000 | -1.444000 |
| C | -1.220000 | 0.546000 | -1.730000 |
| H | -2.099000 | -0.075000 | -1.527000 |
| H | -1.208000 | 0.720000 | -2.819000 |
| H | 3.239000 | -0.187000 | 2.000000 |
| H | 5.456000 | -0.918000 | 1.053000 |
| Cl | 0.534000 | 0.172000 | 3.076000 |

**[Co$^{II}$(TPA)]$^{2+}$**

| | | | |
|---|---|---|---|
| Co | 0.00011223 | -0.00121446 | -0.82231553 |
| N | 0.00067074 | -0.00141292 | 1.31070768 |
| C | 4.00622138 | -1.61800325 | -1.19685933 |
| H | 4.74697955 | -1.74318693 | -1.97958085 |
| C | 2.05519694 | -1.26173667 | 0.72566227 |
| N | -0.13925811 | 2.00922871 | -0.55546238 |
| N | -1.67217540 | -1.12555572 | -0.55510370 |
| C | 0.06651242 | 2.40820526 | 0.72623758 |
| C | 2.77001319 | -1.05799066 | -1.49535130 |
| H | 2.52904983 | -0.73545429 | -2.50424194 |
| C | 0.90599640 | -1.11015844 | 1.70659685 |
| H | 0.31988154 | -2.03706084 | 1.70276615 |

| | | | |
|---|---|---|---|
| H | 1.28102291 | -0.97510469 | 2.72932694 |
| N | 1.81167186 | -0.88474649 | -0.55604953 |
| C | -0.39649324 | 4.69229999 | 0.12290707 |
| H | -0.49893798 | 5.73930536 | 0.39306977 |
| C | 3.27810713 | -1.81617033 | 1.09423671 |
| H | 3.45647287 | -2.10345590 | 2.12624308 |
| C | -2.30357770 | -1.86670700 | -1.49467901 |
| C | -0.06355461 | 3.74450790 | 1.09503896 |
| H | 0.09636966 | 4.04222018 | 2.12711725 |
| C | -2.12026610 | -1.14736958 | 0.72666101 |
| C | -0.59990482 | 4.27701168 | -1.19554522 |
| H | -0.86164089 | 4.98155120 | -1.97794164 |
| C | -3.40898632 | -2.65413237 | -1.19652009 |
| C | -3.21410255 | -1.92633986 | 1.09487952 |
| H | -3.55168893 | -1.93705346 | 2.12702485 |
| C | -0.46823608 | 2.92631596 | -1.49437864 |
| H | -0.62852422 | 2.55663326 | -2.50313002 |
| C | -1.41136531 | -0.23164221 | 1.70913778 |
| H | -1.92045905 | 0.73970569 | 1.71120662 |
| H | -1.47892267 | -0.62828661 | 2.73041471 |
| C | 4.26519895 | -2.00096809 | 0.12166669 |
| H | 5.22376120 | -2.43462381 | 0.39148967 |
| C | -3.87026598 | -2.68608969 | 0.12199474 |
| H | -4.72704921 | -3.29680461 | 0.39155954 |
| C | 0.50816662 | 1.33667946 | 1.70760577 |
| H | 1.60394620 | 1.29253650 | 1.70608631 |
| H | 0.20179909 | 1.59392405 | 2.72979664 |
| H | -1.90376801 | -1.81977105 | -2.50357957 |
| H | -3.88964779 | -3.23115128 | -1.97950068 |

**$[Co^{I}(TPA)]^{+}$**

| | | | |
|---|---|---|---|
| Co | −0.02500009 | 0.12136403 | 0.84374363 |
| N | 0.02967171 | 0.15358745 | −1.30640055 |
| C | −3.89471469 | 2.07593224 | 1.21229177 |
| H | −4.51568388 | 2.48203033 | 2.00425114 |
| C | −2.25126071 | 0.98715076 | −0.71083991 |
| N | 1.86961265 | 0.90973387 | 0.61733386 |
| N | 0.04524051 | −1.93855962 | 0.53295793 |
| C | 2.04700057 | 1.42429650 | −0.62575333 |
| C | −2.76767617 | 1.32202426 | 1.53069073 |
| H | −2.49839313 | 1.13088256 | 2.56534379 |
| C | −1.37494551 | 0.27994054 | −1.73643529 |
| H | −1.76854673 | −0.73587430 | −1.86092279 |
| H | −1.46197383 | 0.77425239 | −2.71544051 |
| N | −1.95328973 | 0.78441702 | 0.59831656 |
| C | 4.26793734 | 2.17535869 | −0.06555666 |
| H | 5.19890347 | 2.66465936 | −0.33631310 |
| C | −3.35662912 | 1.74048462 | −1.10491634 |
| H | −3.56100151 | 1.88847629 | −2.16162466 |
| C | −0.22548583 | −2.93123140 | 1.40257753 |
| C | 3.23628015 | 2.05104489 | −0.99852048 |
| H | 3.35067658 | 2.43866317 | −2.00694969 |
| C | 0.26753122 | −2.26462155 | −0.76287337 |
| C | 4.08155604 | 1.65325353 | 1.21542240 |
| H | 4.85580218 | 1.72042371 | 1.97286275 |
| C | −0.28569019 | −4.27292585 | 1.03171490 |
| C | 0.20652884 | −3.58270158 | −1.21486861 |
| H | 0.37764926 | −3.80378610 | −2.26480513 |
| C | 2.87390794 | 1.02628472 | 1.51009238 |
| H | 2.69236997 | 0.59315773 | 2.48921084 |
| C | 0.65505831 | −1.12635084 | −1.69531710 |
| H | 1.74153092 | −0.99376035 | −1.63518210 |
| H | 0.42737398 | −1.39248240 | −2.73867615 |
| C | −4.19406927 | 2.29374736 | −0.13286383 |
| H | −5.05953071 | 2.88257316 | −0.42210328 |
| C | −0.06990101 | −4.60684911 | −0.30563853 |
| H | −0.11857171 | −5.63998181 | −0.63674882 |
| C | 0.86974936 | 1.32962085 | −1.58988051 |
| H | 0.23919651 | 2.21933441 | −1.46281731 |
| H | 1.23325201 | 1.34365288 | −2.63001881 |
| H | −0.40255424 | −2.62884585 | 2.43094885 |
| H | −0.50520637 | −5.03120696 | 1.77649791 |

**$[Co^{III}\text{-}H(TPA)]^{2+}$**

| | | | |
|---|---|---|---|
| Co | 0.04110519 | -0.36512373 | -0.76307291 |
| N | 0.07401329 | -0.99060706 | 1.18855875 |
| C | 4.22876021 | -0.19305888 | -1.40246400 |
| H | 4.91404165 | 0.20743394 | -2.14212434 |
| C | 2.40859811 | -1.19173441 | 0.41559557 |
| N | -0.07335638 | 1.39677639 | -0.13604937 |
| N | -1.86496025 | -0.66444969 | -0.68664768 |
| C | -0.07832825 | 1.51661742 | 1.20996489 |
| C | 2.85942404 | -0.01247797 | -1.56307305 |
| H | 2.44985811 | 0.52288549 | -2.41142755 |
| C | 1.33263769 | -1.78917396 | 1.30353593 |
| H | 1.11922674 | -2.80860278 | 0.96157430 |
| H | 1.67099383 | -1.86713460 | 2.34382261 |
| N | 1.96748559 | -0.50346246 | -0.67407928 |

| | | | |
|---|---|---|---|
| C | -0.34456006 | 3.90188193 | 1.00733860 |
| H | -0.45328201 | 4.88329172 | 1.45942283 |
| C | 3.76690726 | -1.39212442 | 0.63870955 |
| H | 4.09924414 | -1.93995892 | 1.51551259 |
| C | -2.77611163 | -0.29576749 | -1.61550654 |
| C | -0.21797843 | 2.76822795 | 1.80960994 |
| H | -0.22382113 | 2.84784626 | 2.89298857 |
| C | -2.27330454 | -1.30361982 | 0.44320470 |
| C | -0.32931652 | 3.75767471 | -0.38289748 |
| H | -0.42248482 | 4.61319864 | -1.04375111 |
| C | -4.13178364 | -0.55686910 | -1.45573939 |
| C | -3.61924475 | -1.57741157 | 0.66809579 |
| H | -3.92596032 | -2.08548348 | 1.57761459 |
| C | -0.19627668 | 2.48684153 | -0.92732976 |
| H | -0.18584208 | 2.31763739 | -1.99620944 |
| C | -1.17935823 | -1.78246824 | 1.37978055 |
| H | -1.51420710 | -1.76014356 | 2.42321073 |
| H | -0.95769707 | -2.82977198 | 1.14472075 |
| C | 4.69051053 | -0.88846445 | -0.28290713 |
| H | 5.75479946 | -1.03777784 | -0.12613146 |
| C | -4.56162753 | -1.20131152 | -0.29318707 |
| H | -5.61575734 | -1.40966654 | -0.13517152 |
| C | 0.11952404 | 0.25406562 | 2.02191721 |
| H | 1.09438407 | 0.30989251 | 2.51927116 |
| H | -0.63167980 | 0.19941213 | 2.81735265 |
| H | -2.39094693 | 0.20744822 | -2.49429166 |
| H | -4.83194415 | -0.25574352 | -2.22785700 |
| H | 0.02812932 | 0.07081346 | -2.16592513 |

**$[Co^{II}$-H(TPA)$]^{+}$**

| | | | |
|---|---|---|---|
| Co | 0.04636811 | −0.31523390 | −0.77833602 |
| N | 0.11802380 | −0.26742122 | 1.28958068 |
| C | −0.89372613 | 4.02552035 | −1.00694212 |
| H | −1.23905770 | 4.73088280 | −1.75598366 |
| C | −0.00874319 | 2.16334822 | 0.80421258 |
| N | −1.81556734 | −0.95572379 | −0.54426428 |
| N | 1.99720292 | −0.66886087 | −0.59879298 |
| C | −2.14733458 | −1.19182761 | 0.74761464 |
| C | −0.62410841 | 2.70379605 | −1.36364123 |
| H | −0.75360104 | 2.34859881 | −2.38156790 |
| C | 0.56736985 | 1.09022148 | 1.70691715 |
| H | 1.65906892 | 1.11774137 | 1.62062186 |
| H | 0.32552964 | 1.27929893 | 2.76194846 |
| N | −0.19129844 | 1.79239593 | −0.47883858 |
| C | −4.15713060 | −2.33504056 | 0.08048723 |
| H | −5.06507513 | −2.87767167 | 0.32691287 |
| C | −0.26979081 | 3.46013763 | 1.24744642 |
| H | −0.12264784 | 3.72523738 | 2.29077457 |
| C | 2.96302976 | −0.56406068 | −1.53024086 |
| C | −3.30591434 | −1.88823361 | 1.09252985 |
| H | −3.53916633 | −2.07311610 | 2.13733459 |
| C | 2.31002354 | −1.16103941 | 0.62590745 |
| C | −3.81961088 | −2.07649695 | −1.24919549 |
| H | −4.45179403 | −2.40606242 | −2.06716093 |
| C | 4.28141843 | −0.93803524 | −1.27807180 |
| C | 3.60876863 | −1.54574356 | 0.95182110 |
| H | 3.83221414 | −1.92712315 | 1.94394281 |
| C | −2.63766753 | −1.39310836 | −1.52037556 |
| H | −2.31209716 | −1.18479498 | −2.53267864 |
| C | 1.14279137 | −1.29955484 | 1.59033861 |
| H | 0.67231768 | −2.28079192 | 1.45022800 |
| H | 1.48553537 | −1.24750941 | 2.63454177 |
| C | −0.71753898 | 4.40814827 | 0.32340426 |
| H | −0.92654545 | 5.42603963 | 0.63963338 |
| C | 4.61034868 | −1.43643397 | −0.01678895 |
| H | 5.62926825 | −1.73419786 | 0.21263305 |
| C | −1.24011056 | −0.58896625 | 1.80635736 |
| H | −1.69913651 | 0.34722406 | 2.14460241 |
| H | −1.18106455 | −1.24156920 | 2.68742145 |
| H | 2.64770835 | −0.17442928 | −2.49147297 |
| H | 5.02903076 | −0.83795438 | −2.05808070 |
| H | 0.02660152 | −0.37700344 | −2.29305550 |

## Complex C2

**$[Co^{II}$(BPQA)Cl$]^{+}$**

| | | | |
|---|---|---|---|
| Co | -0.36599559 | 0.02332800 | -0.60978178 |
| N | -1.00637691 | -0.05687034 | 1.57301252 |
| C | -1.81500155 | 4.15192524 | -0.89848645 |
| H | -2.02383432 | 4.83631112 | -1.71053039 |
| C | -1.25168956 | 2.34062258 | 1.08122687 |
| N | -1.83149206 | -1.52514645 | -0.54029204 |
| N | 1.47769019 | -0.42546961 | 0.45132950 |
| C | -2.62248229 | -1.59574364 | 0.54638309 |
| C | 5.37664845 | -0.04536883 | -1.07240984 |
| H | 6.37308952 | 0.05812806 | -1.48471764 |
| C | -1.33203646 | 2.87933398 | -1.17571622 |
| H | -1.15526548 | 2.53531045 | -2.18769529 |

| | | | |
|---|---|---|---|
| C | -0.85384325 | 1.30669257 | 2.11211130 |
| H | 0.20236789 | 1.45495379 | 2.35621796 |
| H | -1.42035047 | 1.44586881 | 3.04154407 |
| N | -1.05465952 | 1.99148565 | -0.20420849 |
| C | -3.78310406 | -3.48987011 | -0.35336222 |
| H | -4.54348307 | -4.25804534 | -0.27567256 |
| C | -1.74397021 | 3.59204196 | 1.43393065 |
| H | -1.90135681 | 3.84020963 | 2.47682657 |
| C | 2.97346743 | 0.18279263 | -1.36194287 |
| H | 2.12452761 | 0.43345191 | -1.98334904 |
| C | 2.76001648 | -0.30083064 | -0.05089827 |
| C | -3.60603742 | -2.56961466 | 0.67694461 |
| H | -4.22277985 | -2.60436227 | 1.56711983 |
| C | 1.31817215 | -0.86707923 | 1.69584126 |
| C | 4.25303037 | 0.30361438 | -1.85441823 |
| H | 4.39889037 | 0.66981405 | -2.86393828 |
| C | -2.96963055 | -3.40918304 | -1.47908274 |
| H | -3.07505522 | -4.10280678 | -2.30319956 |
| C | 3.89720132 | -0.65413901 | 0.74497136 |
| C | 2.39262763 | -1.22628887 | 2.53329008 |
| H | 2.19269695 | -1.57450472 | 3.53918829 |
| C | -2.00490307 | -2.41146261 | -1.53644197 |
| H | -1.35408153 | -2.28923024 | -2.39337618 |
| C | -0.09756982 | -1.02366763 | 2.20634577 |
| H | -0.44568823 | -2.03042012 | 1.95459924 |
| H | -0.11708226 | -0.94574965 | 3.30122716 |
| C | 5.20072819 | -0.51606564 | 0.20473039 |
| H | 6.05147787 | -0.78883089 | 0.81905071 |
| C | -2.02563200 | 4.51361944 | 0.42824777 |
| H | -2.40819095 | 5.49607909 | 0.67877815 |
| C | 3.67469616 | -1.12652853 | 2.05563134 |
| H | 4.52114659 | -1.39756144 | 2.67737851 |
| C | -2.40674627 | -0.51576445 | 1.58368001 |
| H | -3.04236150 | 0.33887493 | 1.33148233 |
| H | -2.71557879 | -0.86338113 | 2.57798267 |
| Cl | -0.15629347 | 0.12434263 | -2.86689137 |

**[Co$^{I}$(BPQA)Cl]**

| | | | |
|---|---|---|---|
| Co | -0.319000 | 0.057000 | -0.592000 |
| N | -1.016000 | -0.055000 | 1.572000 |
| C | -1.509000 | 4.318000 | -0.774000 |
| H | -1.640000 | 5.053000 | -1.563000 |
| C | -1.134000 | 2.380000 | 1.135000 |
| N | -1.782000 | -1.549000 | -0.576000 |
| N | 1.429000 | -0.514000 | 0.348000 |
| C | -2.630000 | -1.565000 | 0.471000 |
| C | 5.305000 | -0.220000 | -1.291000 |
| H | 6.296000 | -0.140000 | -1.730000 |
| C | -0.961000 | 3.070000 | -1.075000 |
| H | -0.672000 | 2.787000 | -2.083000 |
| C | -0.832000 | 1.284000 | 2.144000 |
| H | 0.223000 | 1.370000 | 2.431000 |
| H | -1.428000 | 1.431000 | 3.061000 |
| N | -0.768000 | 2.121000 | -0.139000 |
| C | -3.871000 | -3.391000 | -0.485000 |
| H | -4.683000 | -4.112000 | -0.443000 |
| C | -1.702000 | 3.596000 | 1.511000 |
| H | -1.992000 | 3.764000 | 2.545000 |
| C | 2.890000 | -0.064000 | -1.531000 |
| H | 2.011000 | 0.124000 | -2.141000 |
| C | 2.716000 | -0.421000 | -0.175000 |
| C | -3.681000 | -2.480000 | 0.556000 |
| H | -4.339000 | -2.474000 | 1.422000 |
| C | 1.307000 | -0.882000 | 1.645000 |
| C | 4.160000 | 0.033000 | -2.077000 |
| H | 4.270000 | 0.306000 | -3.123000 |
| C | -3.004000 | -3.355000 | -1.579000 |
| H | -3.118000 | -4.040000 | -2.414000 |
| C | 3.875000 | -0.686000 | 0.628000 |
| C | 2.400000 | -1.140000 | 2.485000 |
| H | 2.219000 | -1.417000 | 3.521000 |
| C | -1.973000 | -2.417000 | -1.585000 |
| H | -1.280000 | -2.321000 | -2.415000 |
| C | -0.108000 | -1.064000 | 2.148000 |
| H | -0.476000 | -2.046000 | 1.827000 |
| H | -0.133000 | -1.054000 | 3.251000 |
| C | 5.161000 | -0.574000 | 0.038000 |
| H | 6.035000 | -0.775000 | 0.655000 |
| C | -1.889000 | 4.588000 | 0.541000 |
| H | -2.329000 | 5.544000 | 0.810000 |
| C | 3.690000 | -1.046000 | 1.991000 |
| H | 4.553000 | -1.241000 | 2.622000 |
| C | -2.412000 | -0.494000 | 1.530000 |
| H | -3.028000 | 0.378000 | 1.272000 |
| H | -2.766000 | -0.855000 | 2.512000 |
| Cl | -0.373000 | 0.287000 | -2.992000 |

**[Co$^{I}$(BPQA)Cl] with dissociated Co-Nqa bond**

| | | | |
|---|---|---|---|
| Co | 1.396000 | 0.039000 | 0.964000 |
| N | 0.520000 | 0.012000 | -1.157000 |
| C | 2.465000 | 4.238000 | 0.665000 |
| H | 3.083000 | 4.922000 | 1.239000 |
| C | 0.924000 | 2.415000 | -0.700000 |
| N | 2.756000 | -1.205000 | 0.014000 |
| N | -2.865000 | -0.506000 | -1.373000 |
| C | 2.614000 | -1.313000 | -1.326000 |
| C | -6.560000 | -0.168000 | 0.565000 |
| H | -7.530000 | -0.071000 | 1.045000 |
| C | 2.382000 | 2.901000 | 1.040000 |
| H | 2.921000 | 2.519000 | 1.902000 |
| C | 0.033000 | 1.386000 | -1.372000 |
| H | -0.970000 | 1.453000 | -0.936000 |
| H | -0.075000 | 1.621000 | -2.444000 |
| N | 1.631000 | 1.995000 | 0.376000 |
| C | 4.105000 | -3.202000 | -1.402000 |
| H | 4.624000 | -3.980000 | -1.955000 |
| C | 0.968000 | 3.737000 | -1.144000 |
| H | 0.395000 | 4.029000 | -2.021000 |
| C | -5.207000 | 0.001000 | -1.448000 |
| H | -5.087000 | 0.213000 | -2.506000 |
| C | -4.059000 | -0.407000 | -0.716000 |
| C | 3.265000 | -2.300000 | -2.065000 |
| H | 3.116000 | -2.361000 | -3.140000 |
| C | -1.796000 | -0.893000 | -0.694000 |
| C | -6.430000 | 0.117000 | -0.819000 |
| H | -7.304000 | 0.429000 | -1.385000 |
| C | 4.252000 | -3.087000 | -0.019000 |
| H | 4.885000 | -3.769000 | 0.540000 |
| C | -4.190000 | -0.697000 | 0.681000 |
| C | -1.827000 | -1.209000 | 0.693000 |
| H | -0.920000 | -1.500000 | 1.214000 |
| C | 3.555000 | -2.082000 | 0.652000 |
| H | 3.606000 | -1.967000 | 1.731000 |
| C | -0.511000 | -1.008000 | -1.490000 |
| H | -0.050000 | -1.985000 | -1.308000 |
| H | -0.777000 | -0.947000 | -2.557000 |
| C | -5.463000 | -0.567000 | 1.299000 |
| H | -5.556000 | -0.787000 | 2.361000 |
| C | 1.746000 | 4.669000 | -0.453000 |
| H | 1.789000 | 5.704000 | -0.780000 |
| C | -3.019000 | -1.103000 | 1.372000 |
| H | -3.067000 | -1.326000 | 2.435000 |
| C | 1.744000 | -0.246000 | -1.953000 |
| H | 2.323000 | 0.683000 | -1.981000 |
| H | 1.494000 | -0.507000 | -2.995000 |
| Cl | 1.011000 | -0.741000 | 3.076000 |

**[Co$^{I}$(BPQA)Cl] with dissociated Co-Npy bond**

| | | | |
|---|---|---|---|
| Co | -0.865000 | -0.617000 | 0.896000 |
| N | -0.492000 | 0.137000 | -1.191000 |
| C | -4.646000 | -2.442000 | -0.256000 |
| H | -5.555000 | -2.783000 | 0.230000 |
| C | -2.309000 | -1.542000 | -1.389000 |
| N | -0.299000 | 3.548000 | -1.465000 |
| N | 1.156000 | -0.539000 | 0.936000 |
| C | -0.989000 | 2.580000 | -0.835000 |
| C | -3.618000 | -1.895000 | 0.506000 |
| H | -3.683000 | -1.779000 | 1.585000 |
| C | -0.961000 | -1.077000 | -1.904000 |
| H | -0.220000 | -1.867000 | -1.717000 |
| H | -0.996000 | -0.916000 | -2.994000 |
| N | -2.468000 | -1.455000 | -0.046000 |
| C | -1.166000 | 3.891000 | 1.174000 |
| H | -1.496000 | 4.009000 | 2.203000 |
| C | -3.301000 | -2.067000 | -2.217000 |
| H | -3.144000 | -2.109000 | -3.291000 |
| C | 1.858000 | -1.009000 | 2.015000 |
| C | -1.443000 | 2.711000 | 0.484000 |
| H | -1.976000 | 1.901000 | 0.975000 |
| C | 1.783000 | -0.334000 | -0.228000 |
| C | -0.457000 | 4.903000 | 0.523000 |
| H | -0.222000 | 5.840000 | 1.020000 |
| C | 3.194000 | -1.297000 | 1.956000 |
| C | 3.177000 | -0.607000 | -0.396000 |
| C | -0.048000 | 4.681000 | -0.793000 |
| H | 0.510000 | 5.442000 | -1.337000 |
| C | 0.968000 | 0.323000 | -1.337000 |
| H | 1.175000 | 1.399000 | -1.311000 |
| H | 1.296000 | -0.027000 | -2.326000 |
| C | -4.489000 | -2.531000 | -1.644000 |
| H | -5.276000 | -2.946000 | -2.267000 |
| C | 3.903000 | -1.112000 | 0.734000 |
| C | -1.263000 | 1.328000 | -1.644000 |
| H | -2.321000 | 1.062000 | -1.565000 |
| H | -1.044000 | 1.548000 | -2.701000 |
| H | 1.271000 | -1.125000 | 2.921000 |

| | | | |
|---|---|---|---|
| H | 3.715000 | -1.661000 | 2.837000 |
| C | 5.286000 | -1.392000 | 0.608000 |
| C | 5.944000 | -1.175000 | -0.587000 |
| C | 5.237000 | -0.666000 | -1.702000 |
| C | 3.888000 | -0.387000 | -1.611000 |
| H | 5.823000 | -1.777000 | 1.472000 |
| H | 7.006000 | -1.391000 | -0.674000 |
| H | 5.761000 | -0.491000 | -2.638000 |
| H | 3.373000 | 0.009000 | -2.480000 |
| Cl | -1.718000 | -0.166000 | 3.028000 |

**$[Co^{II}(BPQA)]^{2+}$**

| | | | |
|---|---|---|---|
| Co | −0.33597939 | 0.01918802 | −0.55749764 |
| N | −1.06957530 | −0.04973032 | 1.44335571 |
| C | −2.15581550 | 3.85807452 | −1.46748506 |
| H | −2.35698048 | 4.45837927 | −2.34847530 |
| C | −1.59593280 | 2.26070368 | 0.71427472 |
| N | −1.55152997 | −1.60391143 | −0.76835443 |
| N | 1.44061941 | −0.25481002 | 0.35995088 |
| C | −2.37444288 | −1.81435411 | 0.29060296 |
| C | 5.19905680 | 0.19034646 | −1.45097720 |
| H | 6.16182307 | 0.30630568 | −1.93865643 |
| C | −1.49519336 | 2.64179017 | −1.59275164 |
| H | −1.17732743 | 2.27385633 | −2.56403424 |
| C | −1.18376601 | 1.36738520 | 1.87078646 |
| H | −0.19697045 | 1.68896599 | 2.22521294 |
| H | −1.87712806 | 1.47702976 | 2.71483081 |
| N | −1.21774709 | 1.85835152 | −0.52596994 |
| C | −3.21002641 | −3.83426010 | −0.71789678 |
| H | −3.85708994 | −4.70621291 | −0.69250114 |
| C | −2.27285572 | 3.46213602 | 0.90833811 |
| H | −2.57416852 | 3.76135506 | 1.90785773 |
| C | 2.77108834 | 0.23967444 | −1.60304807 |
| H | 1.86506127 | 0.39024443 | −2.19077216 |
| C | 2.67619178 | −0.10210012 | −0.23519790 |
| C | −3.20997480 | −2.92717169 | 0.34556312 |
| H | −3.85391461 | −3.08191658 | 1.20622482 |
| C | 1.34829813 | −0.57875009 | 1.65734960 |
| C | 4.00953924 | 0.38113745 | −2.19834412 |
| H | 4.07618755 | 0.64102791 | −3.25067608 |
| C | −2.36637661 | −3.60855484 | −1.80837881 |
| H | −2.33566962 | −4.29022713 | −2.65180809 |
| C | 3.87351021 | −0.29462860 | 0.52606589 |
| C | 2.48667667 | −0.76697657 | 2.46146960 |
| H | 2.37279853 | −1.02124581 | 3.51058754 |
| C | −1.54749239 | −2.48586394 | −1.79365885 |
| H | −0.86665136 | −2.27738103 | −2.61388316 |
| C | −0.04825282 | −0.80666628 | 2.21036850 |
| H | −0.28327399 | −1.87449662 | 2.12341936 |
| H | −0.08883088 | −0.56027502 | 3.27938183 |
| C | 5.13283924 | −0.13988131 | −0.11461721 |
| H | 6.03956911 | −0.28721652 | 0.46543242 |
| C | −2.55378610 | 4.27381028 | −0.19438666 |
| H | −3.07899353 | 5.21502879 | −0.06063256 |
| C | 3.74011359 | −0.62917665 | 1.89449310 |
| H | 4.63245607 | −0.77392826 | 2.49818135 |
| C | −2.37953060 | −0.74376831 | 1.36747434 |
| H | −3.13932228 | 0.00442960 | 1.11119506 |
| H | −2.66570793 | −1.16853134 | 2.33841127 |

**$[Co^{I}(BPQA)]^{+}$**

| | | | |
|---|---|---|---|
| Co | −0.30698706 | 0.12731638 | −0.54351499 |
| N | −1.03003065 | −0.02503342 | 1.47466443 |
| C | −3.36301065 | 3.07867538 | −1.68198352 |
| H | −3.79131552 | 3.49422076 | −2.58839884 |
| C | −2.20646166 | 1.95263383 | 0.54836294 |
| N | −1.13961281 | −1.78254125 | −0.68479791 |
| N | 1.54302031 | −0.01865587 | 0.33388412 |
| C | −1.84872225 | −2.12475161 | 0.41730680 |
| C | 5.23705640 | 0.70799155 | −1.51768689 |
| H | 6.18230599 | 0.89379663 | −2.01894847 |
| C | −2.39065762 | 2.08572215 | −1.76488997 |
| H | −2.05486815 | 1.71012770 | −2.72684909 |
| C | −1.49057921 | 1.34708864 | 1.75006579 |
| H | −0.60045269 | 1.95132920 | 1.96897289 |
| H | −2.13837757 | 1.40034197 | 2.63994569 |
| N | −1.81597879 | 1.52744204 | −0.67993206 |
| C | −2.15188195 | −4.37604936 | −0.38825814 |
| H | −2.54179598 | −5.38224465 | −0.26614282 |
| C | −3.18760078 | 2.93112605 | 0.70983745 |
| H | −3.48717967 | 3.23718234 | 1.70827183 |
| C | 2.84744855 | 0.29129257 | −1.67065525 |
| H | 1.94146330 | 0.14683877 | −2.25508255 |
| C | 2.76309066 | 0.22324923 | −0.25793877 |
| C | −2.35704695 | −3.41057584 | 0.60062163 |
| H | −2.90969823 | −3.65102046 | 1.50469511 |
| C | 1.46926331 | −0.08828213 | 1.66622698 |
| C | 4.05925251 | 0.52729445 | −2.28641338 |

| | | | |
|---|---|---|---|
| H | 4.11514790 | 0.57414558 | -3.37026037 |
| C | -1.43300384 | -4.02049811 | -1.52981697 |
| H | -1.24482401 | -4.73395513 | -2.32573352 |
| C | 3.94984068 | 0.40111537 | 0.52101961 |
| C | 2.59803468 | 0.08620208 | 2.49932463 |
| H | 2.48317481 | 0.02626888 | 3.57771381 |
| C | -0.94237165 | -2.72034454 | -1.63137742 |
| H | -0.36490574 | -2.40884494 | -2.49727275 |
| C | 0.12429928 | -0.45723503 | 2.28322880 |
| H | 0.08724542 | -1.55113716 | 2.35480902 |
| H | 0.06485642 | -0.07594865 | 3.31355445 |
| C | 5.18284879 | 0.64623659 | -0.14202208 |
| H | 6.08112185 | 0.78382907 | 0.45443158 |
| C | -3.77199211 | 3.50962042 | -0.41888053 |
| H | -4.53377836 | 4.27631985 | -0.31270536 |
| C | 3.82995696 | 0.32799684 | 1.93100961 |
| H | 4.71070871 | 0.46674464 | 2.55284721 |
| C | -2.12451482 | -1.01486319 | 1.42018292 |
| H | -3.02951533 | -0.48648028 | 1.09879944 |
| H | -2.34193890 | -1.43940882 | 2.41194206 |

**$[Co^{III}\text{-}H(BPQA)]^{2+}$**

| | | | |
|---|---|---|---|
| Co | -0.47325287 | 0.50696752 | -0.74936576 |
| N | -1.21544612 | 1.50075666 | 0.87393186 |
| C | 2.99624796 | 2.84440754 | -1.41979755 |
| H | 3.88577416 | 2.79938566 | -2.03921008 |
| C | 0.69577528 | 2.87854666 | 0.10256327 |
| N | -2.14731836 | -0.46620446 | -0.70429184 |
| N | 0.48767098 | -0.65868865 | 0.40904206 |
| C | -3.09247052 | 0.12050572 | 0.08124382 |
| C | 3.19675689 | -3.75759773 | -0.56353676 |
| H | 3.88964990 | -4.55270631 | -0.82007882 |
| C | 2.04829772 | 1.83141028 | -1.50142486 |
| H | 2.17428425 | 0.98458020 | -2.16475226 |
| C | -0.63883559 | 2.87705637 | 0.82365809 |
| H | -0.54687385 | 3.31071045 | 1.82639599 |
| H | -1.33397105 | 3.51696676 | 0.26806328 |
| N | 0.91839576 | 1.84953080 | -0.75905356 |
| C | -4.66206451 | -1.62120505 | -0.45515899 |
| H | -5.64501968 | -2.07202815 | -0.35467772 |
| C | 1.61545861 | 3.91421038 | 0.24111924 |
| H | 1.42033751 | 4.72476145 | 0.93706326 |
| C | 1.62422172 | -2.04054408 | -1.28949494 |
| H | 1.11357297 | -1.52769748 | -2.09484096 |
| C | 1.37352518 | -1.66992624 | 0.05311641 |
| C | -4.35707135 | -0.44013237 | 0.22854814 |
| H | -5.09485854 | 0.04109486 | 0.86362398 |
| C | 0.27665001 | -0.35756602 | 1.69605599 |
| C | 2.51194966 | -3.05895914 | -1.58440801 |
| H | 2.68305863 | -3.32625192 | -2.62324157 |
| C | -3.68890877 | -2.21314860 | -1.26290468 |
| H | -3.88833007 | -3.12839028 | -1.81025435 |
| C | 2.06568277 | -2.38328242 | 1.09411180 |
| C | 0.93619911 | -1.02707522 | 2.74402365 |
| H | 0.73205488 | -0.74136535 | 3.77141299 |
| C | -2.43891369 | -1.61161698 | -1.35878418 |
| H | -1.64650523 | -2.03534341 | -1.96383412 |
| C | -0.74792343 | 0.70288377 | 2.04473542 |
| H | -1.61158157 | 0.20031562 | 2.49598381 |
| H | -0.34131089 | 1.36673160 | 2.81600855 |
| C | 2.97316584 | -3.42176669 | 0.75259925 |
| H | 3.48489202 | -3.94628717 | 1.55479477 |
| C | 2.78007340 | 3.89972407 | -0.53074440 |
| H | 3.50709423 | 4.70115087 | -0.43759729 |
| C | 1.82571312 | -2.03206418 | 2.44203730 |
| H | 2.34874260 | -2.56258175 | 3.23330967 |
| C | -2.69895424 | 1.44344513 | 0.71132725 |
| H | -2.99862442 | 2.25893826 | 0.04280698 |
| H | -3.21941600 | 1.60346979 | 1.66362940 |
| H | -0.00570887 | -0.14516837 | -1.96244132 |

**$[Co^{II}\text{-}H(BPQA)]^{+}$**

| | | | |
|---|---|---|---|
| Co | -0.37278349 | -0.25657961 | -0.54405435 |
| N | -0.88342581 | -0.25687924 | 1.45367720 |
| C | -1.37125222 | 4.06035772 | -0.94344867 |
| H | -1.53558329 | 4.77391016 | -1.74451942 |
| C | -0.92647313 | 2.17692184 | 1.00285017 |
| N | -2.17876701 | -1.02885056 | -0.83451046 |
| N | 1.49516588 | -0.56619045 | 0.21337272 |
| C | -2.85713287 | -1.27270673 | 0.31268328 |
| C | 5.25916341 | -0.03687887 | -1.56813883 |
| H | 6.22316710 | 0.10890677 | -2.04660015 |
| C | -0.99664118 | 2.75006258 | -1.24130091 |
| H | -0.86613858 | 2.41287440 | -2.26523644 |
| C | -0.58574941 | 1.09230737 | 2.00664416 |
| H | 0.48853993 | 1.13625756 | 2.21471024 |
| H | -1.10520903 | 1.25298307 | 2.96182417 |
| N | -0.77753038 | 1.82777710 | -0.29108472 |

| | | | |
|---|---|---|---|
| C | −4.51337114 | −2.55524729 | −0.87098559 |
| H | −5.41902083 | −3.15460262 | −0.88115868 |
| C | −1.31129946 | 3.46203190 | 1.38619991 |
| H | −1.43158851 | 3.70880039 | 2.43742069 |
| C | 2.83838152 | 0.22182502 | −1.63960557 |
| H | 1.92596747 | 0.53145353 | −2.13596271 |
| C | 2.73236002 | −0.39668779 | −0.36920286 |
| C | −4.02109351 | −2.04066512 | 0.32995839 |
| H | −4.53529219 | −2.22847338 | 1.26825860 |
| C | 1.41470415 | −1.12977449 | 1.42290490 |
| C | 4.07723186 | 0.39535055 | −2.22142475 |
| H | 4.14963616 | 0.86701173 | −3.19742536 |
| C | −3.82033303 | −2.28998865 | −2.05316091 |
| H | −4.16642122 | −2.67030367 | −3.00866375 |
| C | 3.92410144 | −0.83330686 | 0.29760896 |
| C | 2.54620225 | −1.57647578 | 2.13909772 |
| H | 2.42185781 | −2.02271244 | 3.12110086 |
| C | −2.65413431 | −1.53172510 | −1.99237703 |
| H | −2.06330280 | −1.31280230 | −2.87384081 |
| C | 0.01867848 | −1.29701770 | 2.00101193 |
| H | −0.38454424 | −2.27387334 | 1.70476716 |
| H | 0.04724468 | −1.27808732 | 3.10118372 |
| C | 5.18348744 | −0.64037702 | −0.33240331 |
| H | 6.08226561 | −0.97589778 | 0.17858601 |
| C | −1.53383246 | 4.42124924 | 0.39480405 |
| H | −1.83231992 | 5.43017409 | 0.66452543 |
| C | 3.79403805 | −1.43489601 | 1.57229762 |
| H | 4.68349417 | −1.77154373 | 2.09897004 |
| C | −2.32717403 | −0.59356476 | 1.56119817 |
| H | −2.87714370 | 0.34601496 | 1.68883825 |
| H | −2.52774429 | −1.19849225 | 2.45581660 |
| H | −0.01997987 | −0.28971061 | −2.01457499 |

## Complex C3

### $Co^{II}$(DPA-3-IQA)$Cl_2$

| | | | |
|---|---|---|---|
| Co | 6.001000 | 6.381000 | 13.460000 |
| C | 5.253000 | 5.454000 | 18.245000 |
| H | 5.039000 | 5.244000 | 19.290000 |
| N | 7.230000 | 4.404000 | 13.098000 |
| Cl | 7.992000 | 7.606000 | 13.404000 |
| Cl | 4.228000 | 7.970000 | 13.544000 |
| N | 5.793000 | 5.978000 | 15.575000 |
| C | 8.285000 | 2.136000 | 11.867000 |
| H | 8.689000 | 1.255000 | 11.376000 |
| C | 8.491000 | 4.401000 | 12.639000 |
| H | 9.026000 | 5.337000 | 12.765000 |
| N | 5.631000 | 6.029000 | 11.348000 |
| C | 3.704000 | 4.873000 | 12.296000 |
| H | 3.045000 | 4.017000 | 12.082000 |
| H | 3.087000 | 5.763000 | 12.456000 |
| N | 4.478000 | 4.666000 | 13.538000 |
| C | 9.063000 | 3.285000 | 12.025000 |
| H | 10.087000 | 3.328000 | 11.669000 |
| C | 6.636000 | 6.397000 | 16.527000 |
| H | 7.505000 | 6.938000 | 16.163000 |
| C | 6.402000 | 6.159000 | 17.882000 |
| H | 7.104000 | 6.519000 | 18.628000 |
| C | 4.378000 | 5.018000 | 17.247000 |
| H | 3.474000 | 4.468000 | 17.495000 |
| C | 6.407000 | 6.425000 | 10.363000 |
| H | 7.210000 | 7.105000 | 10.642000 |
| C | 4.675000 | 5.311000 | 15.914000 |
| C | 3.728000 | 4.960000 | 14.778000 |
| H | 3.071000 | 4.124000 | 15.066000 |
| H | 3.113000 | 5.843000 | 14.586000 |
| C | 5.119000 | 3.339000 | 13.607000 |
| H | 4.465000 | 2.564000 | 13.180000 |
| H | 5.255000 | 3.091000 | 14.667000 |
| C | 6.489000 | 3.294000 | 12.966000 |
| C | 6.974000 | 2.140000 | 12.346000 |
| H | 6.335000 | 1.267000 | 12.240000 |
| C | 4.598000 | 5.165000 | 11.105000 |
| C | 4.357000 | 4.665000 | 9.846000 |
| H | 3.538000 | 3.968000 | 9.679000 |
| C | 5.174000 | 5.067000 | 8.755000 |
| C | 4.992000 | 4.602000 | 7.426000 |
| H | 4.188000 | 3.901000 | 7.215000 |
| C | 5.825000 | 5.040000 | 6.416000 |
| H | 5.677000 | 4.682000 | 5.400000 |
| C | 6.873000 | 5.958000 | 6.684000 |
| H | 7.515000 | 6.292000 | 5.873000 |
| C | 7.075000 | 6.426000 | 7.966000 |
| H | 7.873000 | 7.130000 | 8.185000 |
| C | 6.233000 | 5.990000 | 9.022000 |

### $[Co^{II}$(DPA-3-IQA)Cl$]^{+}$

| | | | |
|---|---|---|---|
| Co | 0.69999342 | 0.00012237 | 0.72532138 |

| | | | |
|---|---|---|---|
| N | 0.85266587 | -0.02516571 | -1.57688718 |
| C | 2.68715379 | 3.87788105 | 0.88318443 |
| H | 3.12656067 | 4.47904854 | 1.66860404 |
| C | 1.53616197 | 2.28040213 | -1.02859268 |
| N | 1.96033439 | -1.63811504 | 0.32863615 |
| N | -1.29056453 | -0.23420824 | 0.11382050 |
| C | 2.44381815 | -1.79359501 | -0.91849334 |
| C | 2.09409532 | 2.66128336 | 1.19462264 |
| H | 2.04844223 | 2.28454149 | 2.20981040 |
| C | 0.80889680 | 1.38266529 | -2.00854438 |
| H | -0.24104201 | 1.69117846 | -2.03923304 |
| H | 1.20726767 | 1.51539284 | -3.02264031 |
| N | 1.53307449 | 1.87739740 | 0.25648473 |
| C | 3.53978706 | -3.83237313 | -0.29133881 |
| H | 4.15319025 | -4.69083989 | -0.53855214 |
| C | 2.12204266 | 3.48072634 | -1.41563878 |
| H | 2.11865820 | 3.77547433 | -2.45828401 |
| C | -2.30806104 | -0.04514213 | 0.93967469 |
| C | 3.23141276 | -2.88589071 | -1.26540274 |
| H | 3.59898627 | -2.99059806 | -2.27924278 |
| C | -1.53811565 | -0.49659512 | -1.21025212 |
| C | 3.04808878 | -3.66118615 | 0.99889435 |
| H | 3.26562949 | -4.37206147 | 1.78546795 |
| C | -3.65726868 | -0.08915126 | 0.52346613 |
| C | -2.81113573 | -0.53979229 | -1.71360463 |
| H | -2.97305760 | -0.73904235 | -2.76730983 |
| C | 2.25863061 | -2.55135257 | 1.27010906 |
| H | 1.84863750 | -2.36135306 | 2.25511171 |
| C | -0.31201885 | -0.80517798 | -2.03976574 |
| H | -0.06531044 | -1.86440756 | -1.91508002 |
| H | -0.51412957 | -0.64686983 | -3.10675634 |
| C | 2.70377158 | 4.29256461 | -0.44483748 |
| H | 3.16452641 | 5.23294471 | -0.72361368 |
| C | -3.92425479 | -0.33845916 | -0.85643367 |
| C | 2.13507010 | -0.67734549 | -1.89467769 |
| H | 2.92516094 | 0.07549750 | -1.80887267 |
| H | 2.16169937 | -1.04918848 | -2.92705529 |
| Cl | 0.56839881 | 0.03184131 | 2.98187798 |
| C | -4.73108492 | 0.11359866 | 1.42667551 |
| C | -6.02566937 | 0.06973651 | 0.96862487 |
| C | -6.29209466 | -0.17536520 | -0.39991685 |
| C | -5.26825197 | -0.37517277 | -1.29776475 |
| H | -4.51705958 | 0.30115568 | 2.47280830 |
| H | -6.85105437 | 0.22280414 | 1.65331894 |
| H | -7.32026441 | -0.20608182 | -0.74140780 |
| H | -5.48147396 | -0.56443988 | -2.34368789 |
| H | -2.05238404 | 0.14932556 | 1.97670422 |

**[Co$^{I}$(DPA-3-IQA)Cl]**

| | | | |
|---|---|---|---|
| Co | -0.753000 | -0.060000 | -0.952000 |
| N | -0.944000 | 0.042000 | 1.341000 |
| C | -3.018000 | 3.604000 | -1.447000 |
| H | -3.481000 | 4.117000 | -2.285000 |
| C | -1.782000 | 2.242000 | 0.596000 |
| N | -1.757000 | -1.857000 | -0.469000 |
| N | 1.223000 | -0.174000 | -0.360000 |
| C | -2.317000 | -1.929000 | 0.760000 |
| C | -2.160000 | 2.533000 | -1.684000 |
| H | -1.936000 | 2.174000 | -2.685000 |
| C | -1.008000 | 1.478000 | 1.652000 |
| H | 0.025000 | 1.848000 | 1.657000 |
| H | -1.426000 | 1.669000 | 2.655000 |
| N | -1.543000 | 1.861000 | -0.685000 |
| C | -3.216000 | -4.113000 | 0.286000 |
| H | -3.776000 | -4.994000 | 0.586000 |
| C | -2.638000 | 3.295000 | 0.908000 |
| H | -2.806000 | 3.565000 | 1.948000 |
| C | 2.255000 | 0.003000 | -1.191000 |
| C | -3.040000 | -3.049000 | 1.174000 |
| H | -3.462000 | -3.083000 | 2.175000 |
| C | 1.482000 | -0.293000 | 0.986000 |
| C | -2.660000 | -4.019000 | -0.995000 |
| H | -2.776000 | -4.818000 | -1.721000 |
| C | 3.600000 | 0.093000 | -0.757000 |
| C | 2.747000 | -0.198000 | 1.509000 |
| H | 2.901000 | -0.284000 | 2.582000 |
| C | -1.938000 | -2.877000 | -1.331000 |
| H | -1.486000 | -2.736000 | -2.309000 |
| C | 0.271000 | -0.618000 | 1.841000 |
| H | 0.095000 | -1.699000 | 1.788000 |
| H | 0.468000 | -0.377000 | 2.899000 |
| C | -3.267000 | 3.998000 | -0.128000 |
| H | -3.937000 | 4.824000 | 0.092000 |
| C | 3.868000 | 0.001000 | 0.647000 |
| C | -2.175000 | -0.696000 | 1.638000 |
| H | -3.016000 | -0.021000 | 1.426000 |
| H | -2.252000 | -0.975000 | 2.704000 |
| C | 4.683000 | 0.282000 | -1.659000 |

| | | | |
|---|---|---|---|
| C | 5.977000 | 0.378000 | -1.184000 |
| C | 6.241000 | 0.289000 | 0.204000 |
| C | 5.202000 | 0.105000 | 1.104000 |
| H | 4.475000 | 0.351000 | -2.725000 |
| H | 6.800000 | 0.522000 | -1.879000 |
| H | 7.264000 | 0.366000 | 0.563000 |
| H | 5.404000 | 0.035000 | 2.170000 |
| H | 1.998000 | 0.082000 | -2.245000 |
| Cl | -0.682000 | -0.233000 | -3.337000 |

**[Co$^{I}$(DPA-3-IQA)Cl] with dissociated Co-Nqa bond**

| | | | |
|---|---|---|---|
| Co | 1.035000 | -0.066000 | 0.941000 |
| N | 0.746000 | 0.161000 | -1.319000 |
| C | 2.519000 | 3.998000 | 1.343000 |
| H | 3.026000 | 4.565000 | 2.118000 |
| C | 1.226000 | 2.463000 | -0.538000 |
| N | 2.492000 | -1.371000 | 0.247000 |
| N | -2.520000 | 0.073000 | -2.437000 |
| C | 2.691000 | -1.366000 | -1.091000 |
| C | 2.217000 | 2.656000 | 1.553000 |
| H | 2.475000 | 2.156000 | 2.482000 |
| C | 0.451000 | 1.592000 | -1.509000 |
| H | -0.620000 | 1.733000 | -1.328000 |
| H | 0.640000 | 1.918000 | -2.546000 |
| N | 1.586000 | 1.890000 | 0.635000 |
| C | 3.990000 | -3.390000 | -0.975000 |
| H | 4.567000 | -4.176000 | -1.454000 |
| C | 1.508000 | 3.799000 | -0.826000 |
| H | 1.214000 | 4.216000 | -1.786000 |
| C | -3.779000 | 0.267000 | -2.095000 |
| C | 3.426000 | -2.360000 | -1.735000 |
| H | 3.554000 | -2.327000 | -2.814000 |
| C | -1.683000 | -0.507000 | -1.522000 |
| C | 3.785000 | -3.392000 | 0.406000 |
| H | 4.195000 | -4.175000 | 1.037000 |
| C | -4.327000 | -0.069000 | -0.827000 |
| C | -2.102000 | -0.886000 | -0.262000 |
| H | -1.409000 | -1.330000 | 0.448000 |
| C | 3.025000 | -2.371000 | 0.975000 |
| H | 2.807000 | -2.340000 | 2.039000 |
| C | -0.257000 | -0.716000 | -1.987000 |
| H | 0.057000 | -1.746000 | -1.788000 |
| H | -0.234000 | -0.556000 | -3.076000 |
| C | 2.161000 | 4.586000 | 0.126000 |
| H | 2.384000 | 5.629000 | -0.077000 |
| C | -3.446000 | -0.659000 | 0.131000 |
| C | 2.103000 | -0.172000 | -1.811000 |
| H | 2.751000 | 0.689000 | -1.610000 |
| H | 2.105000 | -0.333000 | -2.901000 |
| C | -5.686000 | 0.162000 | -0.482000 |
| C | -6.146000 | -0.176000 | 0.773000 |
| C | -5.267000 | -0.752000 | 1.730000 |
| C | -3.944000 | -0.990000 | 1.422000 |
| H | -6.352000 | 0.609000 | -1.217000 |
| H | -7.185000 | 0.001000 | 1.039000 |
| H | -5.645000 | -1.001000 | 2.718000 |
| H | -3.261000 | -1.412000 | 2.154000 |
| H | -4.423000 | 0.722000 | -2.849000 |
| Cl | 0.029000 | -0.979000 | 2.785000 |

**[Co$^{I}$(DPA-3-IQA)Cl] with dissociated Co-Npy bond**

| | | | |
|---|---|---|---|
| Co | 0.466000 | 0.637000 | 0.779000 |
| N | 0.921000 | -0.205000 | -1.269000 |
| C | 3.499000 | 3.740000 | 0.317000 |
| H | 4.079000 | 4.395000 | 0.959000 |
| C | 1.991000 | 2.030000 | -1.220000 |
| N | 2.107000 | -3.450000 | -1.290000 |
| N | -1.388000 | -0.059000 | 0.342000 |
| C | 2.169000 | -2.295000 | -0.604000 |
| C | 2.599000 | 2.838000 | 0.879000 |
| H | 2.450000 | 2.747000 | 1.951000 |
| C | 1.076000 | 1.086000 | -1.979000 |
| H | 0.078000 | 1.541000 | -2.052000 |
| H | 1.439000 | 0.941000 | -3.010000 |
| N | 1.856000 | 2.001000 | 0.127000 |
| C | 2.295000 | -3.449000 | 1.503000 |
| H | 2.357000 | -3.437000 | 2.587000 |
| C | 2.879000 | 2.899000 | -1.852000 |
| H | 2.972000 | 2.888000 | -2.935000 |
| C | -2.469000 | 0.255000 | 1.056000 |
| C | 2.267000 | -2.248000 | 0.793000 |
| H | 2.293000 | -1.299000 | 1.322000 |
| C | -1.550000 | -0.426000 | -0.977000 |
| C | 2.238000 | -4.652000 | 0.796000 |
| H | 2.261000 | -5.611000 | 1.306000 |
| C | -3.783000 | 0.252000 | 0.528000 |
| C | -2.782000 | -0.432000 | -1.588000 |
| H | -2.870000 | -0.716000 | -2.634000 |

| | | | |
|---|---|---|---|
| C | 2.146000 | -4.597000 | -0.596000 |
| H | 2.098000 | -5.510000 | -1.187000 |
| C | -0.301000 | -0.925000 | -1.683000 |
| H | -0.160000 | -1.977000 | -1.414000 |
| H | -0.442000 | -0.895000 | -2.775000 |
| C | 3.643000 | 3.774000 | -1.074000 |
| H | 4.342000 | 4.460000 | -1.544000 |
| C | -3.952000 | -0.091000 | -0.852000 |
| C | 2.149000 | -1.029000 | -1.439000 |
| H | 2.993000 | -0.390000 | -1.158000 |
| H | 2.274000 | -1.317000 | -2.494000 |
| C | -4.922000 | 0.586000 | 1.311000 |
| C | -6.179000 | 0.581000 | 0.743000 |
| C | -6.347000 | 0.244000 | -0.625000 |
| C | -5.256000 | -0.085000 | -1.407000 |
| H | -4.786000 | 0.845000 | 2.358000 |
| H | -7.048000 | 0.837000 | 1.343000 |
| H | -7.344000 | 0.245000 | -1.058000 |
| H | -5.387000 | -0.346000 | -2.455000 |
| H | -2.287000 | 0.519000 | 2.095000 |
| Cl | 0.893000 | 0.503000 | 3.072000 |

**$[Co^{II}(DPA\text{-}3\text{-}IQA)]^{2+}$**

| | | | |
|---|---|---|---|
| Co | −0.72390898 | −0.00361963 | −0.76803267 |
| N | −0.94589662 | −0.01811435 | 1.35260783 |
| C | −2.68091772 | 3.83619497 | −1.30209901 |
| H | −3.08507952 | 4.41933827 | −2.12281928 |
| C | −1.62125716 | 2.27993754 | 0.71913431 |
| N | −1.95681767 | −1.62045830 | −0.63489028 |
| N | 1.23159236 | −0.21973336 | −0.31843429 |
| C | −2.49949180 | −1.79628082 | 0.59707599 |
| C | −2.08030704 | 2.60853874 | −1.55315067 |
| H | −2.01028216 | 2.21453926 | −2.56302071 |
| C | −0.94865721 | 1.40672485 | 1.76392237 |
| H | 0.09577193 | 1.72563501 | 1.86608514 |
| H | −1.42045000 | 1.54126317 | 2.74602465 |
| N | −1.55855912 | 1.84552401 | −0.56558080 |
| C | −3.57534202 | −3.82336745 | −0.13051854 |
| H | −4.20570347 | −4.68440716 | 0.07174300 |
| C | −2.22181367 | 3.49479881 | 1.04066201 |
| H | −2.26993866 | 3.82013914 | 2.07562615 |
| C | 2.26765232 | −0.03321332 | −1.14055188 |
| C | −3.30799104 | −2.89475751 | 0.87901631 |
| H | −3.72488366 | −3.02189893 | 1.87369998 |
| C | 1.46471121 | −0.47044157 | 1.02152814 |
| C | −3.01899170 | −3.63398515 | −1.39814432 |
| H | −3.20048524 | −4.33363638 | −2.20715862 |
| C | 3.60997923 | −0.08128321 | −0.71600250 |
| C | 2.73956983 | −0.51128269 | 1.52818368 |
| H | 2.89622690 | −0.70011790 | 2.58705573 |
| C | −2.21070366 | −2.52373620 | −1.60939478 |
| H | −1.74860011 | −2.34187875 | −2.57542514 |
| C | 0.23389151 | −0.77431502 | 1.85167327 |
| H | −0.00654364 | −1.84069270 | 1.76279569 |
| H | 0.41709018 | −0.57310290 | 2.91507887 |
| C | −2.75477583 | 4.28543133 | 0.01885041 |
| H | −3.22429873 | 5.23663657 | 0.25208989 |
| C | 3.86333671 | −0.32231662 | 0.67675576 |
| C | −2.23265711 | −0.70571338 | 1.61969582 |
| H | −3.03360855 | 0.04032443 | 1.55023562 |
| H | −2.26483625 | −1.11282625 | 2.63881557 |
| C | 4.69574794 | 0.11138576 | −1.61380542 |
| C | 5.98819256 | 0.06385326 | −1.13814933 |
| C | 6.23912465 | −0.17363853 | 0.23892792 |
| C | 5.20171642 | −0.36230937 | 1.13335084 |
| H | 4.49815807 | 0.29364444 | −2.66679758 |
| H | 6.82397484 | 0.20820115 | −1.81581715 |
| H | 7.26580609 | −0.20714924 | 0.59174933 |
| H | 5.40579177 | −0.54474237 | 2.18461054 |
| H | 2.03910141 | 0.16658853 | −2.18536900 |

**$[Co^{I}(DPA\text{-}3\text{-}IQA)]^{+}$**

| | | | |
|---|---|---|---|
| Co | −0.65609045 | 0.02597447 | −0.77749589 |
| N | −0.89923580 | 0.06834925 | 1.35701804 |
| C | −3.31517368 | 3.48339240 | −1.38311067 |
| H | −3.86156273 | 3.95194136 | −2.19529735 |
| C | −1.89180333 | 2.20846994 | 0.59951812 |
| N | −1.81901127 | −1.69035680 | −0.59443462 |
| N | 1.31268346 | −0.36720897 | −0.35517844 |
| C | −2.29468253 | −1.87098058 | 0.66113031 |
| C | −2.58293994 | 2.32251602 | −1.61674711 |
| H | −2.55312964 | 1.86862343 | −2.60285754 |
| C | −1.03841989 | 1.50423111 | 1.64859160 |
| H | −0.03100999 | 1.94038045 | 1.63326668 |
| H | −1.44926258 | 1.69006311 | 2.65407602 |
| N | −1.88092495 | 1.68998341 | −0.65444670 |
| C | −3.17262100 | −4.04262543 | 0.09309626 |
| H | −3.69329694 | −4.95573360 | 0.36603470 |

| | | | |
|---|---|---|---|
| C | -2.61611076 | 3.35915752 | 0.91229436 |
| H | -2.61515115 | 3.73866363 | 1.93029948 |
| C | 2.35554399 | -0.25625974 | -1.17189562 |
| C | -2.96273043 | -3.03575524 | 1.03848493 |
| H | -3.31710819 | -3.15036428 | 2.05919349 |
| C | 1.53801275 | -0.31119638 | 1.00833838 |
| C | -2.69556267 | -3.85104678 | -1.20404665 |
| H | -2.83183821 | -4.60313074 | -1.97457156 |
| C | 3.69051967 | -0.07482834 | -0.73284036 |
| C | 2.79406285 | -0.12130517 | 1.53132144 |
| H | 2.93163480 | -0.07476503 | 2.60891073 |
| C | -2.02151256 | -2.66812787 | -1.49921855 |
| H | -1.62181197 | -2.48773647 | -2.49323125 |
| C | 0.32669181 | -0.55692346 | 1.89201718 |
| H | 0.14360803 | -1.63766570 | 1.92519950 |
| H | 0.53575836 | -0.23717199 | 2.92390858 |
| C | -3.33397144 | 4.01248068 | -0.09157880 |
| H | -3.90009924 | 4.91176499 | 0.13262558 |
| C | 3.92346914 | 0.00378158 | 0.67535546 |
| C | -2.12429086 | -0.70852009 | 1.62740674 |
| H | -2.97807797 | -0.03316669 | 1.49816559 |
| H | -2.15950879 | -1.06715447 | 2.66742683 |
| C | 4.78113744 | 0.03642851 | -1.63593135 |
| C | 6.05912125 | 0.21941606 | -1.15125709 |
| C | 6.29139207 | 0.29910228 | 0.24590612 |
| C | 5.24752817 | 0.19481142 | 1.14434824 |
| H | 4.59688542 | -0.02450578 | -2.70566513 |
| H | 6.89575440 | 0.30345367 | -1.83860358 |
| H | 7.30486964 | 0.44281466 | 0.60962364 |
| H | 5.43122070 | 0.25443226 | 2.21397370 |
| H | 2.14602788 | -0.31473237 | -2.23789489 |

**$[Co^{III}$-H(DPA-3-IQA)$]^{2+}$**

| | | | |
|---|---|---|---|
| Co | -0.64765084 | -0.30682347 | -0.69145155 |
| N | -0.82158494 | -1.05317828 | 1.20798266 |
| C | -1.35004013 | 3.74934664 | -0.09698080 |
| H | -1.44779091 | 4.63869284 | -0.71077159 |
| C | -1.07262824 | 1.43042031 | 1.37452654 |
| N | -2.53997351 | -0.67352008 | -0.86551166 |
| N | 1.25094508 | -0.40766285 | -0.39382286 |
| C | -3.06718545 | -1.36154045 | 0.18187470 |
| C | -1.06645829 | 2.53086718 | -0.70122732 |
| H | -0.94322235 | 2.43464634 | -1.77221202 |
| C | -0.82562827 | 0.13534636 | 2.11635690 |
| H | 0.15478771 | 0.20187013 | 2.60173265 |
| H | -1.56305934 | 0.00504839 | 2.91591577 |
| N | -0.93162016 | 1.40228755 | 0.03004749 |
| C | -5.24550349 | -1.30359665 | -0.83935749 |
| H | -6.30351727 | -1.54837565 | -0.82467595 |
| C | -1.37018013 | 2.62387877 | 2.03176737 |
| H | -1.48580848 | 2.62848748 | 3.11175184 |
| C | 2.21705083 | 0.16657184 | -1.11309695 |
| C | -4.42160330 | -1.68325026 | 0.22278523 |
| H | -4.82426421 | -2.23043589 | 1.07010339 |
| C | 1.58384912 | -1.17182275 | 0.71602428 |
| C | -4.69226266 | -0.60850509 | -1.91787915 |
| H | -5.29749856 | -0.30294851 | -2.76483552 |
| C | 3.58534598 | 0.02665904 | -0.80600947 |
| C | 2.88927885 | -1.33471114 | 1.10256962 |
| H | 3.12451916 | -1.93520094 | 1.97742976 |
| C | -3.33720793 | -0.30290568 | -1.89354429 |
| H | -2.85921902 | 0.23874089 | -2.70106108 |
| C | 0.41743129 | -1.87684763 | 1.37494931 |
| H | 0.24020406 | -2.83651836 | 0.87435697 |
| H | 0.62149141 | -2.09513726 | 2.43045333 |
| C | -1.50979502 | 3.79725466 | 1.29059898 |
| H | -1.74003571 | 4.73490390 | 1.78771072 |
| C | 3.94385368 | -0.74376422 | 0.35153123 |
| C | -2.09258439 | -1.83632341 | 1.24417040 |
| H | -2.55213714 | -1.80574265 | 2.23871903 |
| H | -1.85087497 | -2.88677784 | 1.04536341 |
| C | 4.59876706 | 0.62903429 | -1.60042527 |
| C | 5.92282038 | 0.46791345 | -1.25190781 |
| C | 6.27680821 | -0.29135807 | -0.10654595 |
| C | 5.31046499 | -0.88662730 | 0.68458187 |
| H | 4.32183742 | 1.20798831 | -2.47729681 |
| H | 6.70390092 | 0.92104512 | -1.85452768 |
| H | 7.32611118 | -0.40681275 | 0.14898822 |
| H | 5.59380739 | -1.46838039 | 1.55716097 |
| H | 1.90196408 | 0.75621426 | -1.96779490 |
| H | -0.50654653 | 0.22120259 | -2.05675945 |

**$[Co^{II}$-H(DPA-3-IQA)$]^{+}$**

| | | | |
|---|---|---|---|
| Co | 0.62503260 | -0.18116624 | -0.70523858 |
| N | 0.92157351 | -0.12761050 | 1.33996920 |
| C | 3.95854800 | -2.82115583 | -1.46170772 |
| H | 4.58363019 | -3.11888222 | -2.29718761 |
| C | 2.31765378 | -2.00199186 | 0.58723262 |

| | | | |
|---|---|---|---|
| N | 1.46424705 | 1.77084222 | −0.54017376 |
| N | −1.30171338 | 0.07329826 | −0.28045183 |
| C | 1.94374341 | 2.03637102 | 0.69166350 |
| C | 2.97836343 | −1.84723293 | −1.64190830 |
| H | 2.80906443 | −1.36997680 | −2.60055982 |
| C | 1.33111860 | −1.52144628 | 1.63959246 |
| H | 0.42786034 | −2.14349808 | 1.60252851 |
| H | 1.75373968 | −1.61893857 | 2.65094072 |
| N | 2.17625472 | −1.44237088 | −0.64008822 |
| C | 2.37881706 | 4.32400718 | 0.09864338 |
| H | 2.73149025 | 5.31976004 | 0.35139450 |
| C | 3.28191614 | −2.97464777 | 0.84316646 |
| H | 3.37871549 | −3.39802689 | 1.83871690 |
| C | −2.31641975 | −0.03468745 | −1.12952385 |
| C | 2.40205507 | 3.30355875 | 1.05308781 |
| H | 2.77264459 | 3.48817411 | 2.05758004 |
| C | −1.55582346 | 0.01317393 | 1.07513004 |
| C | 1.89341407 | 4.04368677 | −1.17939538 |
| H | 1.85850920 | 4.80731685 | −1.94980756 |
| C | −3.66053552 | −0.20723901 | −0.71848358 |
| C | −2.82218229 | −0.18422829 | 1.57001038 |
| H | −2.98613935 | −0.24121856 | 2.64333300 |
| C | 1.44010673 | 2.75332068 | −1.45429583 |
| H | 1.04286437 | 2.48174356 | −2.42760868 |
| C | −0.36291294 | 0.28170487 | 1.97034235 |
| H | −0.30448412 | 1.36297265 | 2.14134738 |
| H | −0.49089131 | −0.18796072 | 2.95501691 |
| C | 4.11338499 | −3.39434647 | −0.19871882 |
| H | 4.86972928 | −4.15394659 | −0.02405262 |
| C | −3.92780076 | −0.29872716 | 0.68373478 |
| C | 2.01195915 | 0.84415346 | 1.62626780 |
| H | 2.96570342 | 0.33099073 | 1.46165199 |
| H | 1.99511237 | 1.15816056 | 2.67928400 |
| C | −4.72857941 | −0.30970925 | −1.64962234 |
| C | −6.01806509 | −0.49374118 | −1.19830366 |
| C | −6.28453700 | −0.58469135 | 0.19253144 |
| C | −5.26417962 | −0.49127278 | 1.11790727 |
| H | −4.51607722 | −0.24219158 | −2.71348329 |
| H | −6.83775594 | −0.57152120 | −1.90639317 |
| H | −7.30698455 | −0.73003938 | 0.52963333 |
| H | −5.47497656 | −0.56108915 | 2.18184906 |
| H | −2.06117015 | 0.00453340 | −2.18376153 |
| H | 0.43073487 | −0.25314198 | −2.20787305 |

## Complex C4

**$Co^{II}$(DPA-1-IQA)$Cl_2$**

| | | | |
|---|---|---|---|
| Co | -1.447000 | -0.000000 | 0.522000 |
| C | -1.189000 | -3.092000 | 1.013000 |
| H | -1.371000 | -2.769000 | 2.035000 |
| N | -1.209000 | -2.115000 | 0.100000 |
| Cl | -3.656000 | -0.001000 | -0.388000 |
| Cl | -1.771000 | -0.000000 | 2.837000 |
| C | -0.953000 | -4.425000 | 0.668000 |
| H | -0.948000 | -5.193000 | 1.435000 |
| N | -0.573000 | -0.000000 | -1.614000 |
| N | -1.210000 | 2.114000 | 0.100000 |
| C | -0.744000 | -3.707000 | -1.622000 |
| H | -0.572000 | -3.915000 | -2.675000 |
| N | 0.839000 | 0.000000 | 0.770000 |
| C | -0.721000 | -4.733000 | -0.673000 |
| H | -0.525000 | -5.758000 | -0.979000 |
| C | -1.004000 | 2.403000 | -1.198000 |
| C | -1.152000 | 1.242000 | -2.167000 |
| H | -2.225000 | 1.069000 | -2.294000 |
| H | -0.714000 | 1.497000 | -3.145000 |
| C | -1.152000 | -1.243000 | -2.167000 |
| H | -0.714000 | -1.497000 | -3.145000 |
| H | -2.225000 | -1.070000 | -2.294000 |
| C | -1.003000 | -2.403000 | -1.198000 |
| C | -0.745000 | 3.707000 | -1.622000 |
| H | -0.573000 | 3.914000 | -2.675000 |
| C | -0.722000 | 4.733000 | -0.673000 |
| H | -0.527000 | 5.758000 | -0.979000 |
| C | -0.954000 | 4.424000 | 0.668000 |
| H | -0.950000 | 5.193000 | 1.434000 |
| C | -1.190000 | 3.092000 | 1.013000 |
| H | -1.373000 | 2.769000 | 2.035000 |
| C | 0.901000 | 0.000000 | -1.663000 |
| H | 1.244000 | 0.877000 | -2.228000 |
| H | 1.245000 | -0.877000 | -2.228000 |
| C | 1.594000 | 0.000000 | -0.312000 |
| C | 3.026000 | 0.000000 | -0.254000 |
| C | 3.858000 | 0.000000 | -1.408000 |
| H | 3.419000 | 0.000000 | -2.400000 |
| C | 5.233000 | 0.001000 | -1.282000 |
| H | 5.858000 | 0.001000 | -2.171000 |
| C | 5.837000 | 0.001000 | -0.000000 |

| | | | |
|---|---|---|---|
| H | 6.920000 | 0.001000 | 0.083000 |
| C | 5.056000 | 0.001000 | 1.136000 |
| H | 5.513000 | 0.001000 | 2.123000 |
| C | 3.638000 | 0.001000 | 1.038000 |
| C | 2.788000 | 0.001000 | 2.175000 |
| H | 3.213000 | 0.001000 | 3.175000 |
| C | 1.427000 | 0.001000 | 2.004000 |
| H | 0.728000 | 0.001000 | 2.832000 |

**[$Co^{II}$(DPA-1-IQA)Cl]$^{+}$**

| | | | |
|---|---|---|---|
| Co | -0.88615925 | -0.03240455 | 0.81272822 |
| N | -0.32626998 | 0.02944832 | -1.41018683 |
| C | -3.61652106 | -3.36238664 | 0.10776566 |
| H | -4.39757941 | -3.84726288 | 0.67891574 |
| C | -1.61182044 | -2.07075478 | -1.24873002 |
| N | -1.59029147 | 1.85022547 | 0.17312487 |
| N | 1.20568122 | -0.22405633 | 0.80057464 |
| C | -1.63832473 | 2.10041759 | -1.14896001 |
| C | -2.90837079 | -2.30539447 | 0.66499618 |
| H | -3.10542900 | -1.94212013 | 1.66686303 |
| C | -0.44020163 | -1.35951171 | -1.89280880 |
| H | 0.47546360 | -1.88737603 | -1.60852341 |
| H | -0.51049917 | -1.40926451 | -2.98705480 |
| N | -1.92783761 | -1.67151765 | -0.00231494 |
| C | -2.41442586 | 4.33332477 | -0.74775193 |
| H | -2.73164471 | 5.30372258 | -1.11108275 |
| C | -2.28549661 | -3.11355074 | -1.87477905 |
| H | -2.01648455 | -3.40816168 | -2.88214099 |
| C | 1.86550845 | -0.61279868 | 1.93339366 |
| C | -2.03827809 | 3.33622072 | -1.64494775 |
| H | -2.05895433 | 3.51262826 | -2.71379911 |
| C | 1.88661843 | 0.06310717 | -0.30190108 |
| C | -2.37486733 | 4.06633778 | 0.61708876 |
| H | -2.66058655 | 4.81245126 | 1.34718028 |
| C | 3.22674177 | -0.72608438 | 1.97672738 |
| C | 3.30688102 | -0.01082560 | -0.35773741 |
| C | -1.95442152 | 2.81171580 | 1.03986566 |
| H | -1.90491414 | 2.54177369 | 2.08826158 |
| C | 1.05673130 | 0.53682482 | -1.48247145 |
| H | 1.02045994 | 1.63077337 | -1.45580275 |
| H | 1.52409451 | 0.25926942 | -2.43337604 |
| C | -3.30130674 | -3.77086287 | -1.18435786 |
| H | -3.83878685 | -4.58803415 | -1.65088185 |
| C | 3.99554260 | -0.42477083 | 0.82700340 |
| C | -1.29550351 | 0.93527369 | -2.05369629 |
| H | -2.21246903 | 0.36681996 | -2.23857458 |
| H | -0.94059620 | 1.29217421 | -3.02901129 |
| Cl | -1.46222835 | -0.08063933 | 2.99368585 |
| H | 1.23548874 | -0.81238433 | 2.79051890 |
| H | 3.71411524 | -1.03774383 | 2.89256648 |
| C | 5.41021120 | -0.51179959 | 0.81360220 |
| C | 6.11521145 | -0.19456127 | -0.32136751 |
| C | 5.43932081 | 0.22342723 | -1.49186108 |
| C | 4.06892487 | 0.31396862 | -1.51384519 |
| H | 5.92489783 | -0.82889729 | 1.71327523 |
| H | 7.19676857 | -0.26106427 | -0.32517567 |
| H | 6.01109233 | 0.47380655 | -2.37744401 |
| H | 3.57784834 | 0.64208283 | -2.42078425 |

**[$Co^{I}$(DPA-1-IQA)Cl]**

| | | | |
|---|---|---|---|
| Co | 0.888000 | -0.036000 | -0.866000 |
| N | 0.385000 | 0.024000 | 1.376000 |
| C | 3.459000 | -3.527000 | -0.301000 |
| H | 4.189000 | -4.052000 | -0.909000 |
| C | 1.581000 | -2.134000 | 1.142000 |
| N | 1.841000 | 1.752000 | -0.245000 |
| N | -1.150000 | -0.278000 | -0.838000 |
| C | 1.744000 | 2.059000 | 1.071000 |
| C | 2.751000 | -2.456000 | -0.842000 |
| H | 2.896000 | -2.114000 | -1.863000 |
| C | 0.465000 | -1.371000 | 1.834000 |
| H | -0.485000 | -1.853000 | 1.575000 |
| H | 0.571000 | -1.440000 | 2.930000 |
| N | 1.830000 | -1.768000 | -0.135000 |
| C | 2.377000 | 4.344000 | 0.648000 |
| H | 2.583000 | 5.352000 | 0.999000 |
| C | 2.260000 | -3.184000 | 1.756000 |
| H | 2.040000 | -3.442000 | 2.789000 |
| C | -1.844000 | -0.643000 | -1.958000 |
| C | 2.000000 | 3.341000 | 1.549000 |
| H | 1.907000 | 3.552000 | 2.612000 |
| C | -1.830000 | 0.079000 | 0.270000 |
| C | 2.474000 | 4.023000 | -0.709000 |
| H | 2.761000 | 4.768000 | -1.446000 |
| C | -3.212000 | -0.707000 | -1.994000 |
| C | -3.255000 | 0.069000 | 0.331000 |
| C | 2.192000 | 2.721000 | -1.116000 |
| H | 2.245000 | 2.407000 | -2.156000 |

| | | | |
|---|---|---|---|
| C | -0.981000 | 0.569000 | 1.431000 |
| H | -0.896000 | 1.665000 | 1.378000 |
| H | -1.454000 | 0.340000 | 2.399000 |
| C | 3.217000 | -3.898000 | 1.025000 |
| H | 3.759000 | -4.721000 | 1.482000 |
| C | -3.974000 | -0.355000 | -0.839000 |
| C | 1.397000 | 0.898000 | 1.985000 |
| H | 2.301000 | 0.295000 | 2.133000 |
| H | 1.087000 | 1.267000 | 2.977000 |
| H | -1.225000 | -0.875000 | -2.820000 |
| H | -3.719000 | -1.020000 | -2.903000 |
| C | -5.386000 | -0.396000 | -0.813000 |
| C | -6.089000 | -0.020000 | 0.323000 |
| C | -5.391000 | 0.414000 | 1.469000 |
| C | -4.007000 | 0.462000 | 1.477000 |
| H | -5.916000 | -0.723000 | -1.704000 |
| H | -7.175000 | -0.054000 | 0.329000 |
| H | -5.944000 | 0.716000 | 2.355000 |
| H | -3.498000 | 0.809000 | 2.371000 |
| Cl | 1.495000 | -0.071000 | -3.169000 |

**[Co$^{I}$(DPA-1-IQA)Cl] with dissociated Co-Nqa bond**

| | | | |
|---|---|---|---|
| Co | 2.018000 | 0.536000 | -0.598000 |
| N | 0.274000 | -0.507000 | 0.427000 |
| C | 4.757000 | -2.722000 | -0.312000 |
| H | 5.793000 | -2.899000 | -0.583000 |
| C | 2.119000 | -2.195000 | 0.284000 |
| N | 1.329000 | 2.057000 | 0.565000 |
| N | -2.264000 | -1.479000 | 1.283000 |
| C | 0.537000 | 1.660000 | 1.592000 |
| C | 4.087000 | -1.612000 | -0.819000 |
| H | 4.566000 | -0.898000 | -1.484000 |
| C | 0.628000 | -1.940000 | 0.457000 |
| H | 0.114000 | -2.418000 | -0.386000 |
| H | 0.256000 | -2.433000 | 1.362000 |
| N | 2.796000 | -1.345000 | -0.526000 |
| C | 0.209000 | 3.940000 | 2.296000 |
| H | -0.231000 | 4.672000 | 2.969000 |
| C | 2.735000 | -3.312000 | 0.852000 |
| H | 2.168000 | -3.965000 | 1.511000 |
| C | -3.441000 | -1.973000 | 1.761000 |
| C | -0.045000 | 2.576000 | 2.468000 |
| H | -0.687000 | 2.223000 | 3.271000 |
| C | -2.278000 | -0.729000 | 0.196000 |
| C | 1.030000 | 4.343000 | 1.237000 |
| H | 1.246000 | 5.392000 | 1.060000 |
| C | -4.660000 | -1.738000 | 1.174000 |
| C | -3.490000 | -0.412000 | -0.512000 |
| C | 1.564000 | 3.374000 | 0.392000 |
| H | 2.189000 | 3.620000 | -0.462000 |
| C | -0.948000 | -0.191000 | -0.327000 |
| H | -0.791000 | -0.548000 | -1.352000 |
| H | -1.007000 | 0.901000 | -0.403000 |
| C | 4.072000 | -3.585000 | 0.551000 |
| H | 4.567000 | -4.452000 | 0.979000 |
| C | -4.717000 | -0.938000 | 0.001000 |
| C | 0.367000 | 0.161000 | 1.746000 |
| H | 1.264000 | -0.245000 | 2.237000 |
| H | -0.490000 | -0.074000 | 2.388000 |
| H | -3.360000 | -2.582000 | 2.659000 |
| H | -5.570000 | -2.156000 | 1.595000 |
| C | -5.934000 | -0.646000 | -0.672000 |
| C | -5.936000 | 0.133000 | -1.809000 |
| C | -4.722000 | 0.654000 | -2.321000 |
| C | -3.525000 | 0.388000 | -1.688000 |
| H | -6.864000 | -1.049000 | -0.276000 |
| H | -6.871000 | 0.351000 | -2.319000 |
| H | -4.735000 | 1.266000 | -3.219000 |
| H | -2.607000 | 0.796000 | -2.098000 |
| Cl | 2.775000 | 1.619000 | -2.519000 |

**[Co$^{I}$(DPA-1-IQA)Cl] with dissociated Co-Npy bond**

| | | | |
|---|---|---|---|
| Co | -0.865000 | -0.617000 | 0.896000 |
| N | -0.492000 | 0.137000 | -1.191000 |
| C | -4.646000 | -2.442000 | -0.256000 |
| H | -5.555000 | -2.783000 | 0.230000 |
| C | -2.309000 | -1.542000 | -1.389000 |
| N | -0.299000 | 3.548000 | -1.465000 |
| N | 1.156000 | -0.539000 | 0.936000 |
| C | -0.989000 | 2.580000 | -0.835000 |
| C | -3.618000 | -1.895000 | 0.506000 |
| H | -3.683000 | -1.779000 | 1.585000 |
| C | -0.961000 | -1.077000 | -1.904000 |
| H | -0.220000 | -1.867000 | -1.717000 |
| H | -0.996000 | -0.916000 | -2.994000 |
| N | -2.468000 | -1.455000 | -0.046000 |
| C | -1.166000 | 3.891000 | 1.174000 |
| H | -1.496000 | 4.009000 | 2.203000 |

| | | | |
|---|---|---|---|
| C | -3.301000 | -2.067000 | -2.217000 |
| H | -3.144000 | -2.109000 | -3.291000 |
| C | 1.858000 | -1.009000 | 2.015000 |
| C | -1.443000 | 2.711000 | 0.484000 |
| H | -1.976000 | 1.901000 | 0.975000 |
| C | 1.783000 | -0.334000 | -0.228000 |
| C | -0.457000 | 4.903000 | 0.523000 |
| H | -0.222000 | 5.840000 | 1.020000 |
| C | 3.194000 | -1.297000 | 1.956000 |
| C | 3.177000 | -0.607000 | -0.396000 |
| C | -0.048000 | 4.681000 | -0.793000 |
| H | 0.510000 | 5.442000 | -1.337000 |
| C | 0.968000 | 0.323000 | -1.337000 |
| H | 1.175000 | 1.399000 | -1.311000 |
| H | 1.296000 | -0.027000 | -2.326000 |
| C | -4.489000 | -2.531000 | -1.644000 |
| H | -5.276000 | -2.946000 | -2.267000 |
| C | 3.903000 | -1.112000 | 0.734000 |
| C | -1.263000 | 1.328000 | -1.644000 |
| H | -2.321000 | 1.062000 | -1.565000 |
| H | -1.044000 | 1.548000 | -2.701000 |
| H | 1.271000 | -1.125000 | 2.921000 |
| H | 3.715000 | -1.661000 | 2.837000 |
| C | 5.286000 | -1.392000 | 0.608000 |
| C | 5.944000 | -1.175000 | -0.587000 |
| C | 5.237000 | -0.666000 | -1.702000 |
| C | 3.888000 | -0.387000 | -1.611000 |
| H | 5.823000 | -1.777000 | 1.472000 |
| H | 7.006000 | -1.391000 | -0.674000 |
| H | 5.761000 | -0.491000 | -2.638000 |
| H | 3.373000 | 0.009000 | -2.480000 |
| Cl | -1.718000 | -0.166000 | 3.028000 |

**$[Co^{II}(DPA\text{-}1\text{-}IQA)]^{2+}$**

| | | | |
|---|---|---|---|
| Co | 0.88388393 | −0.02482686 | −0.90168064 |
| N | 0.52567857 | 0.00973311 | 1.19156205 |
| C | 3.66061990 | −3.36057646 | −0.75184932 |
| H | 4.37791490 | −3.83569151 | −1.41271191 |
| C | 1.80797357 | −2.08800377 | 0.85260233 |
| N | 1.67679204 | 1.81517724 | −0.52681773 |
| N | −1.11930773 | −0.19945850 | −0.96125333 |
| C | 1.82374352 | 2.08475621 | 0.79567513 |
| C | 2.91038560 | −2.28140316 | −1.20352948 |
| H | 3.03145712 | −1.89680515 | −2.21212006 |
| C | 0.71503141 | −1.39204828 | 1.64443830 |
| H | −0.22838682 | −1.92566634 | 1.47754516 |
| H | 0.92320462 | −1.44053717 | 2.72134360 |
| N | 2.00081206 | −1.65801681 | −0.42044786 |
| C | 2.60696467 | 4.30410257 | 0.28954695 |
| H | 2.96740000 | 5.27630028 | 0.61291465 |
| C | 2.53671958 | −3.15489852 | 1.37203375 |
| H | 2.37339430 | −3.47792313 | 2.39590012 |
| C | −1.83082161 | −0.55495265 | −2.08082712 |
| C | 2.28028843 | 3.32560013 | 1.23281793 |
| H | 2.38274266 | 3.52416852 | 2.29557266 |
| C | −1.75422995 | 0.05329559 | 0.19326237 |
| C | 2.46005508 | 4.01867291 | −1.07014931 |
| H | 2.70059705 | 4.75205579 | −1.83256558 |
| C | −3.19595675 | −0.65762338 | −2.06781486 |
| C | −3.16859332 | −0.01854127 | 0.30246186 |
| C | 1.98639615 | 2.76505510 | −1.43846568 |
| H | 1.84541682 | 2.50294248 | −2.48307941 |
| C | −0.87248079 | 0.48491052 | 1.35757753 |
| H | −0.85621777 | 1.58092918 | 1.39737507 |
| H | −1.27823142 | 0.13859999 | 2.31468893 |
| C | 3.47197208 | −3.80285014 | 0.55997634 |
| H | 4.04688886 | −4.63856945 | 0.94811737 |
| C | −3.91322993 | −0.38961394 | −0.87280308 |
| C | 1.53100894 | 0.94476071 | 1.75566548 |
| H | 2.45716118 | 0.38045754 | 1.91868423 |
| H | 1.21680277 | 1.32992982 | 2.73449722 |
| H | −1.25222404 | −0.75241528 | −2.97759136 |
| H | −3.72616868 | −0.94073800 | −2.97195577 |
| C | −5.32640725 | −0.47353138 | −0.80598649 |
| C | −5.98327804 | −0.19344961 | 0.37413380 |
| C | −5.25563245 | 0.18097475 | 1.53309744 |
| C | −3.88071664 | 0.26734685 | 1.50394100 |
| H | −5.88249157 | −0.75756076 | −1.69481540 |
| H | −7.06633495 | −0.25738622 | 0.42121209 |
| H | −5.79106646 | 0.40160055 | 2.45155303 |
| H | −3.35342230 | 0.56269213 | 2.40468138 |

**$[Co^{I}(DPA\text{-}1\text{-}IQA)]^{+}$**

| | | | |
|---|---|---|---|
| Co | 0.85804352 | −0.05987154 | −0.94314451 |
| N | 0.54095302 | −0.24748546 | 1.16962967 |
| C | 4.50442101 | −2.50755226 | −0.90319892 |
| H | 5.35497012 | −2.70971722 | −1.54615265 |
| C | 2.30465125 | −1.91758765 | 0.64346928 |

| | | | |
|---|---|---|---|
| N | 1.35588705 | 1.90009393 | -0.41604097 |
| N | -1.18081115 | -0.08428171 | -1.01640373 |
| C | 1.41549208 | 2.07133643 | 0.92611608 |
| C | 3.53184137 | -1.59543272 | -1.30447969 |
| H | 3.61153423 | -1.07248491 | -2.25273845 |
| C | 1.03620709 | -1.61171635 | 1.43129333 |
| H | 0.25113893 | -2.30834124 | 1.11062260 |
| H | 1.20772985 | -1.79528876 | 2.50446905 |
| N | 2.44974272 | -1.29878016 | -0.55594099 |
| C | 1.67517483 | 4.45490017 | 0.68319739 |
| H | 1.79377731 | 5.44458136 | 1.11430221 |
| C | 3.24679731 | -2.82953284 | 1.11871875 |
| H | 3.10689726 | -3.29512521 | 2.09026596 |
| C | -1.93524651 | -0.15703600 | -2.15886509 |
| C | 1.56279322 | 3.33116318 | 1.50603718 |
| H | 1.59376091 | 3.43020109 | 2.58763305 |
| C | -1.77757652 | -0.21509802 | 0.17096930 |
| C | 1.62791900 | 4.27761206 | -0.69974884 |
| H | 1.70991808 | 5.11805296 | -1.38157943 |
| C | -3.29028716 | -0.35518117 | -2.14515218 |
| C | -3.18830582 | -0.41066437 | 0.29738384 |
| C | 1.46124890 | 2.98847347 | -1.20221761 |
| H | 1.40578522 | 2.81082584 | -2.27266680 |
| C | -0.90323463 | -0.06965708 | 1.41837441 |
| H | -1.06069631 | 0.93974143 | 1.81821413 |
| H | -1.23964785 | -0.76309310 | 2.19958783 |
| C | 4.36108142 | -3.13643416 | 0.33451556 |
| H | 5.10273887 | -3.84762451 | 0.68603390 |
| C | -3.96655481 | -0.48754594 | -0.90440423 |
| C | 1.38361440 | 0.80705112 | 1.77057378 |
| H | 2.40584512 | 0.41577331 | 1.83110606 |
| H | 1.07403960 | 1.04136727 | 2.80040613 |
| H | -1.38868306 | -0.04313616 | -3.08927546 |
| H | -3.84446613 | -0.40432091 | -3.07794771 |
| C | -5.36997192 | -0.68229704 | -0.82021059 |
| C | -5.98822584 | -0.78782419 | 0.40727513 |
| C | -5.22640666 | -0.70219991 | 1.59892594 |
| C | -3.86040121 | -0.51843535 | 1.54918269 |
| H | -5.94822735 | -0.74280543 | -1.73840895 |
| H | -7.06275983 | -0.93501092 | 0.46609511 |
| H | -5.72506414 | -0.78168582 | 2.56061414 |
| H | -3.30635226 | -0.45025189 | 2.47908006 |

**[Co$^{III}$-H(DPA-1-IQA)]$^{2+}$**

| | | | |
|---|---|---|---|
| Co | -1.32925063 | 0.09216429 | -0.64955770 |
| N | -1.10636594 | 0.07267739 | 1.38393001 |
| C | -1.24769719 | 4.28706329 | -1.24829424 |
| H | -1.17442958 | 4.97863889 | -2.08098557 |
| C | -1.44935258 | 2.45194315 | 0.80168329 |
| N | -1.62428674 | -1.81094577 | -0.48731849 |
| N | 0.53161400 | -0.04097460 | -0.78540381 |
| C | -1.81176625 | -2.22264351 | 0.79635789 |
| C | -1.24056255 | 2.91740270 | -1.48785361 |
| H | -1.15904547 | 2.51294663 | -2.48962643 |
| C | -1.66654395 | 1.37689128 | 1.85012139 |
| H | -1.24800716 | 1.67464132 | 2.81882416 |
| H | -2.74501472 | 1.24884970 | 1.99829071 |
| N | -1.34222762 | 2.01856671 | -0.48397333 |
| C | -2.05106649 | -4.50011582 | 0.06021706 |
| H | -2.21539553 | -5.55150073 | 0.27725936 |
| C | -1.44516511 | 3.80930416 | 1.10690181 |
| H | -1.52714517 | 4.13498010 | 2.13964352 |
| C | 1.19234671 | -0.07712124 | -1.98859742 |
| C | -2.01992974 | -3.56454701 | 1.09898078 |
| H | -2.16339834 | -3.87389950 | 2.13001323 |
| C | 1.19245010 | -0.09215039 | 0.37260969 |
| C | -1.86818198 | -4.06699798 | -1.25473371 |
| H | -1.89063220 | -4.76175538 | -2.08769678 |
| C | 2.55843553 | -0.16687607 | -2.03691281 |
| C | 2.61109052 | -0.18671073 | 0.41088281 |
| C | -1.64826332 | -2.71461802 | -1.49155640 |
| H | -1.49258864 | -2.32719104 | -2.49127999 |
| C | 0.36547029 | -0.04866947 | 1.64157156 |
| H | 0.55259014 | -0.95595727 | 2.22665529 |
| H | 0.69210818 | 0.79339163 | 2.26173540 |
| C | -1.34508482 | 4.74060377 | 0.06884118 |
| H | -1.34347376 | 5.80432945 | 0.28794800 |
| C | 3.31698739 | -0.22414002 | -0.83914398 |
| C | -1.87026862 | -1.12619068 | 1.84375261 |
| H | -2.91668760 | -0.82934422 | 1.97977592 |
| H | -1.51492526 | -1.48582490 | 2.81683102 |
| H | 0.57946511 | -0.03111134 | -2.87777271 |
| H | 3.05469777 | -0.19375474 | -3.00237158 |
| C | 4.73165407 | -0.31637925 | -0.83689236 |
| C | 5.42041998 | -0.36944313 | 0.35726895 |
| C | 4.72539103 | -0.33353054 | 1.59322823 |
| C | 3.34955566 | -0.24425443 | 1.62616145 |
| H | 5.26287195 | -0.34443805 | -1.78399128 |

| | | | |
|---|---|---|---|
| H | 6.50406491 | −0.43949641 | 0.35632349 |
| H | 5.28562796 | −0.37701869 | 2.52234228 |
| H | 2.84341355 | −0.21880583 | 2.58586969 |
| H | −1.50497720 | 0.10569810 | −2.10793529 |

**$[Co^{II}$-H(DPA-1-IQA)$]^{+}$**

| | | | |
|---|---|---|---|
| Co | −0.73423057 | −0.11404213 | −0.89307409 |
| N | −0.39562955 | −0.21980492 | 1.13466917 |
| C | −3.01714546 | 3.68316547 | −0.47969237 |
| H | −3.67162138 | 4.29613504 | −1.09110797 |
| C | −1.33849571 | 2.07017021 | 0.97302329 |
| N | −2.23936654 | −1.36278240 | −0.55369349 |
| N | 1.23612313 | 0.16436930 | −0.93988119 |
| C | −2.29896264 | −1.79366969 | 0.72939917 |
| C | −2.41057588 | 2.55067028 | −1.02429344 |
| H | −2.57592779 | 2.25333560 | −2.05550012 |
| C | −0.32937114 | 1.17239760 | 1.66113420 |
| H | 0.67390976 | 1.55620661 | 1.44715075 |
| H | −0.45448874 | 1.18827382 | 2.75284359 |
| N | −1.58987069 | 1.76134836 | −0.31455198 |
| C | −3.89259281 | −3.49811231 | 0.14157223 |
| H | −4.53094911 | −4.33339680 | 0.41426624 |
| C | −1.91591126 | 3.17516025 | 1.59900606 |
| H | −1.70068745 | 3.39190764 | 2.64168223 |
| C | 1.96955893 | 0.66497222 | −1.97895532 |
| C | −3.10753809 | −2.86554638 | 1.10735455 |
| H | −3.12527945 | −3.19586122 | 2.14212904 |
| C | 1.85380806 | −0.33695518 | 0.13435627 |
| C | −3.83950141 | −3.04208523 | −1.17664482 |
| H | −4.43209313 | −3.50475512 | −1.95905514 |
| C | 3.34083757 | 0.67734782 | −1.96197494 |
| C | 3.27352272 | −0.38462605 | 0.24379723 |
| C | −2.99473783 | −1.97938006 | −1.48546587 |
| H | −2.89404025 | −1.59764499 | −2.49463685 |
| C | 0.92778763 | −0.89030100 | 1.21162776 |
| H | 0.77040898 | −1.96271187 | 1.03949149 |
| H | 1.36265455 | −0.78275948 | 2.21451381 |
| C | −2.76846939 | 3.99733152 | 0.85705916 |
| H | −3.23036776 | 4.86600571 | 1.31733407 |
| C | 4.03989477 | 0.14695282 | −0.84685207 |
| C | −1.50583864 | −0.99579282 | 1.74983590 |
| H | −2.19157223 | −0.28082669 | 2.21923638 |
| H | −1.13583042 | −1.64556485 | 2.55418466 |
| H | 1.39105678 | 1.04150121 | −2.81333246 |
| H | 3.89038809 | 1.08502925 | −2.80526893 |
| C | 5.45813291 | 0.11978120 | −0.77931415 |
| C | 6.09474536 | −0.41736129 | 0.31840039 |
| C | 5.34093512 | −0.95099740 | 1.39443591 |
| C | 3.96314490 | −0.93670435 | 1.36185027 |
| H | 6.03176110 | 0.52623131 | −1.60795590 |
| H | 7.17980849 | −0.43660234 | 0.36325916 |
| H | 5.85648447 | −1.37589663 | 2.25075759 |
| H | 3.41042638 | −1.35921753 | 2.19451464 |
| H | −0.93797597 | −0.05066933 | −2.39525997 |