Investigations into the Coordination Chemistry of 1,3-bis(2'-benzimidazolylimino)isoindoline

James T. Engle, Goran Martić, and Christopher J. Ziegler

Исследование координационной химии 1,3-бис(2’-бензимидазолилимино)изоиндолина

Д. Т. Энгл, Г. Мартич, К. Д. Циглер

Supporting Information

*Crystal Structure Report for Compound **1***

An orange needle-like specimen of **1**, approximate dimensions 0.07×0.07×0.36 mm, was used for the X-ray crystallographic analysis. The X-ray intensity data were measured.

The total exposure time was 15.70 hours. The frames were integrated with the Bruker SAINT software package using a narrow-frame algorithm. The integration of the data using a triclinic unit cell yielded a total of 22967 reflections to a maximum θ angle of 61.00° (0.88 Å resolution), of which 6517 were independent (average redundancy 3.524, completeness = 96.7%, R_{int} = 2.42%, R_{sig} = 2.08%) and 5694 (87.37%) were greater than $2\sigma(F^2)$. The final cell constants of a = 7.7287(2) Å, b = 17.0183(4) Å, c = 17.1125(4) Å, α = 84.6490(10)°, β = 81.6060(10)°, γ = 82.2480(10)°, volume = 2200.11(9) $Å^3$, are based upon the refinement of the XYZ-centroids of 118 reflections above 20 σ(I) with 12.19° < 2θ < 118.4°. Data were corrected for absorption effects using the multi-scan method (SADABS). The estimated minimum and maximum transmission as calculated from SADABS are 0.6759 and 0.7528.

The structure was solved and refined using the Bruker SHELXTL Software Package, using the space group P-1, with Z = 2 for the formula unit, $C_{50}H_{44}N_{16}O_2$. The final anisotropic full-matrix least-squares refinement on F^2 with 613 variables converged at R1 = 4.41%, for the observed data and wR2 = 12.20% for all data. The goodness-of-fit was 0.959. The largest peak in the final difference electron density synthesis was 0.572 $e^-/Å^3$ and the largest hole was -0.476 $e^-/Å^3$ with an RMS deviation of 0.046 $e^-/Å^3$. On the basis of the final model, the calculated density was 1.360 g/cm^3 and F(000), 944 e^-.

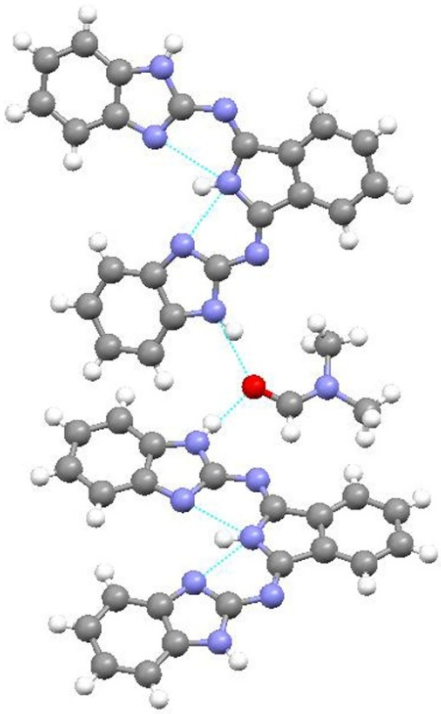

Figure S1. Hydrogen bonding contacts for the crystallographic structure of **1**.

Table S1. Crystal data and structure refinement for **1**.

Identification code	**1**
Empirical formula	$C_{50}H_{44}N_{16}O_2$
Formula weight	901.01
Temperature	100(2) K
Wavelength	1.54178 Å
Crystal system	Triclinic
Space group	P-1
Unit cell dimensions	a = 7.7287(2) Å α = 84.6490(10)°. b = 17.0183(4) Å β = 81.6060(10)°. c = 17.1125(4) Å γ = 82.2480(10)°.
Volume	2200.11(9) $Å^3$
Z	2
Density (calculated)	1.360 Mg/m^3
Absorption coefficient	0.718 mm^{-1}
F(000)	944
Crystal size	0.36 x 0.07 x 0.07 mm^3
Theta range for data collection	2.62 to 61.00°
Index ranges	-8<=h<=8, -19<=k<=19, -19<=l<=19
Reflections collected	22967
Independent reflections	6517 [R(int) = 0.0242]
Completeness to theta = 61.00°	96.7 %
Absorption correction	Multi-scan/SADABS
Max. and min. transmission	0.7528 and 0.6759
Refinement method	Full-matrix least-squares on F^2

Data / restraints / parameters	6517 / 0 / 613
Goodness-of-fit on F^2	0.959
Final R indices [I>2sigma(I)]	R1 = 0.0441, wR2 = 0.1159
R indices (all data)	R1 = 0.0503, wR2 = 0.1220
Largest diff. peak and hole	0.572 and -0.476 e.Å^{-3}

*Crystal Structure Report for Compound **2***

A red block-like specimen of **2**, approximate dimensions 0.15×0.20×0.37 mm, was used for the X-ray crystallographic analysis. The X-ray intensity data were measured.

The total exposure time was 1.01 hours. The frames were integrated with the Bruker SAINT software package using a narrow-frame algorithm. The integration of the data using a monoclinic unit cell yielded a total of 43296 reflections to a maximum θ angle of 25.14° (0.84 Å resolution), of which 11848 were independent (average redundancy 3.654, completeness = 99.1%, R_{int} = 7.84%, R_{sig} = 7.67%) and 8332 (70.32%) were greater than $2\sigma(F^2)$. The final cell constants of a = 18.608(12) Å, b = 18.168(12) Å, c = 19.784(13) Å, β = 91.977(9)°, volume = 6684.(8) Å^3, are based upon the refinement of the XYZ-centroids of 4604 reflections above 20 σ(I) with 4.379° < 2θ < 47.98°. Data were corrected for absorption effects using the multi-scan method (SADABS). The estimated minimum and maximum transmission as calculated from SADABS are 0.5957 and 0.7452.

The structure was solved and refined using the Bruker SHELXTL Software Package, using the space group P2(1)/n, with Z = 4 for the formula unit, $C_{62}H_{69}Cl_4Fe_2N_{20}O_6$. The final anisotropic full-matrix least-squares refinement on F^2 with 859 variables converged at R1 = 5.59%, for the observed data and wR2 = 12.60% for all data. The goodness-of-fit was 1.407. The largest peak in the final difference electron density synthesis was 1.118 e^-/Å^3 and the largest hole was -0.581 e^-/Å^3 with an RMS deviation of 0.091 e^-/Å^3. On the basis of the final model, the calculated density was 1.435 g/cm^3 and F(000), 2996 e^-.

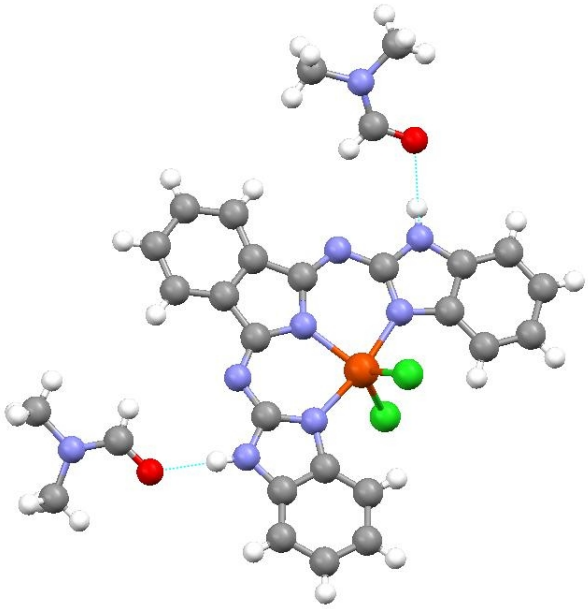

Figure S2. Hydrogen bonding contacts for the crystallographic structure of **2**.

Table S2. Crystal data and structure refinement for **2**.

Identification code	**2**
Empirical formula	$C_{62}H_{69}Cl_4Fe_2N_{20}O_6$
Formula weight	1443.87
Temperature	100(2) K
Wavelength	0.71073 Å
Crystal system	Monoclinic
Space group	P2(1)/n
Unit cell dimensions	a = 18.608(12) Å α = 90°.
	b = 18.168(12) Å β = 91.977(9)°.
	c = 19.784(13) Å γ = 90°.
Volume	6684(8) $Å^3$
Z	4
Density (calculated)	1.435 Mg/m^3
Absorption coefficient	0.660 mm^{-1}
F(000)	2996
Crystal size	0.37 x 0.20 x 0.15 mm^3
Theta range for data collection	1.48 to 25.14°
Index ranges	-22<=h<=22, -21<=k<=21, -23<=l<=23
Reflections collected	43296
Independent reflections	11848 [R(int) = 0.0784]
Completeness to theta = 25.14°	99.1 %
Absorption correction	Multi-scan/SADABS
Max. and min. transmission	0.7452 and 0.5957
Refinement method	Full-matrix least-squares on F^2
Data / restraints / parameters	11848 / 0 / 859
Goodness-of-fit on F^2	1.407
Final R indices [I>2sigma(I)]	R1 = 0.0559, wR2 = 0.1158
R indices (all data)	R1 = 0.0842, wR2 = 0.1260
Largest diff. peak and hole	1.118 and -0.581 e.$Å^{-3}$

*Crystal Structure Report for Compound **3***

A red rod-like specimen of $C_{27}H_{24}CoN_8O_3$, approximate dimensions 0.08×0.12×0.33 mm, was used for the X-ray crystallographic analysis. The X-ray intensity data were measured.

The total exposure time was 5.05 hours. The frames were integrated with the Bruker SAINT software package using a narrow-frame algorithm. The integration of the data using a monoclinic unit cell yielded a total of 21584 reflections to a maximum θ angle of 27.47° (0.77 Å resolution), of which 5762 were independent (average redundancy 3.746, completeness = 99.3%, R_{int} = 5.18%, R_{sig} = 4.85%) and 4765 (82.70%) were greater than $2\sigma(F^2)$. The final cell constants of a = 8.6790(11) Å, b = 22.814(3) Å, c = 12.9592(16) Å, β = 98.520(2)°, volume = 2537.6(6) $Å^3$, are based upon the refinement of the XYZ-centroids of 9995 reflections above 20 σ(I) with 4.780° < 2θ < 54.88°. Data were corrected for absorption effects using the multi-scan method (SADABS). The estimated minimum and maximum transmission as calculated from SADABS are 0.6258 and 0.7456.

The structure was solved and refined using the Bruker SHELXTL Software Package, using the space group P2(1)/n, with Z = 4 for the formula unit, $C_{27}H_{24}CoN_8O_3$. The final anisotropic full-matrix least-squares refinement on F^2 with 355 variables converged at R1 = 4.61%, for the observed data and wR2 = 12.34% for all data. The goodness-of-fit was 0.997. The largest peak in the final difference electron density synthesis was 0.935 $e^-/Å^3$ and the largest hole was -0.343 $e^-/Å^3$ with an RMS deviation of 0.092 $e^-/Å^3$. On the basis of the final model, the calculated density was 1.485 g/cm^3 and F(000), 1172 e^-.

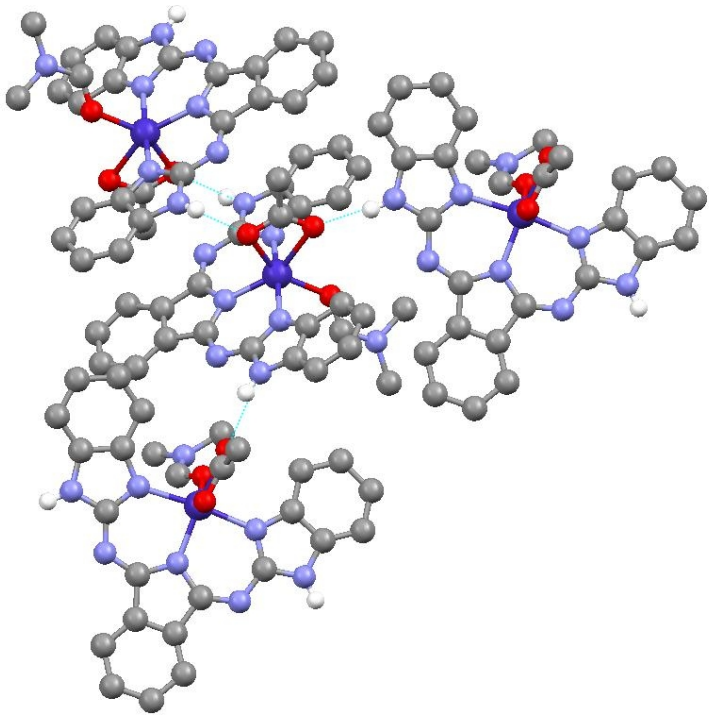

Figure S3. Hydrogen bonding contacts for the crystallographic structure of **3**.

Table S3. Crystal data and structure refinement for **3**.

Identification code	**3**
Empirical formula	$C_{27}H_{24}CoN_8O_3$
Formula weight	567.47
Temperature	100(2) K
Wavelength	0.71073 Å
Crystal system	Monoclinic
Space group	P2(1)/n
Unit cell dimensions	a = 8.6790(11) Å α = 90°.
	b = 22.814(3) Å β = 98.520(2)°.
	c = 12.9592(16) Å γ = 90°.
Volume	2537.7(6) $Å^3$
Z	4
Density (calculated)	1.485 Mg/m^3
Absorption coefficient	0.723 mm^{-1}
F(000)	1172
Crystal size	0.33 x 0.12 x 0.08 mm^3
Theta range for data collection	1.79 to 27.47°
Index ranges	-10<=h<=11, -29<=k<=29, -16<=l<=16

Reflections collected	21584
Independent reflections	5762 [R(int) = 0.0518]
Completeness to theta = 27.47°	99.3 %
Absorption correction	Multi-scan/SADABS
Max. and min. transmission	0.7456 and 0.6258
Refinement method	Full-matrix least-squares on F^2
Data / restraints / parameters	5762 / 0 / 355
Goodness-of-fit on F^2	0.997
Final R indices [I>2sigma(I)]	R1 = 0.0461, wR2 = 0.1144
R indices (all data)	R1 = 0.0580, wR2 = 0.1234
Largest diff. peak and hole	0.935 and -0.343 e.$Å^{-3}$

*Crystal Structure Report for Compound **4***

An orange plate-like specimen of **4**, approximate dimensions 0.08×0.15×0.15 mm, was used for the X-ray crystallographic analysis. The X-ray intensity data were measured.

The total exposure time was 10.10 hours. The frames were integrated with the Bruker SAINT software package using a narrow-frame algorithm. The integration of the data using a triclinic unit cell yielded a total of 22551 reflections to a maximum θ angle of 26.00° (0.81 Å resolution), of which 11176 were independent (average redundancy 2.018, completeness = 99.2%, R_{int} = 5.92%, R_{sig} = 9.17%) and 7798 (69.77%) were greater than $2\sigma(F^2)$. The final cell constants of a = 12.746(2) Å, b = 13.734(3) Å, c = 18.698(4) Å, α = 70.321(2)°, β = 72.422(2)°, γ = 72.583(2)°, volume = 2866.0(9) $Å^3$, are based upon the refinement of the XYZ-centroids of 8699 reflections above 20 σ(I) with 4.496° < 2θ < 55.86°. Data were corrected for absorption effects using the multi-scan method (SADABS). The estimated minimum and maximum transmission as calculated from SADABS are 0.6227 and 0.7457.

The structure was solved and refined using the Bruker SHELXTL Software Package, using the space group P-1, with Z = 2 for the formula unit, $C_{54}H_{48}N_{16}Ni_2O_6$. The final anisotropic full-matrix least-squares refinement on F^2 with 709 variables converged at R1 = 5.64%, for the observed data and wR2 = 8.24% for all data. The goodness-of-fit was 1.174. The largest peak in the final difference electron density synthesis was 0.948 $e^-/Å^3$ and the largest hole was -0.377 $e^-/Å^3$ with an RMS deviation of 0.095 $e^-/Å^3$. On the basis of the final model, the calculated density was 1.315 g/cm^3 and F(000), 1176 e^-.

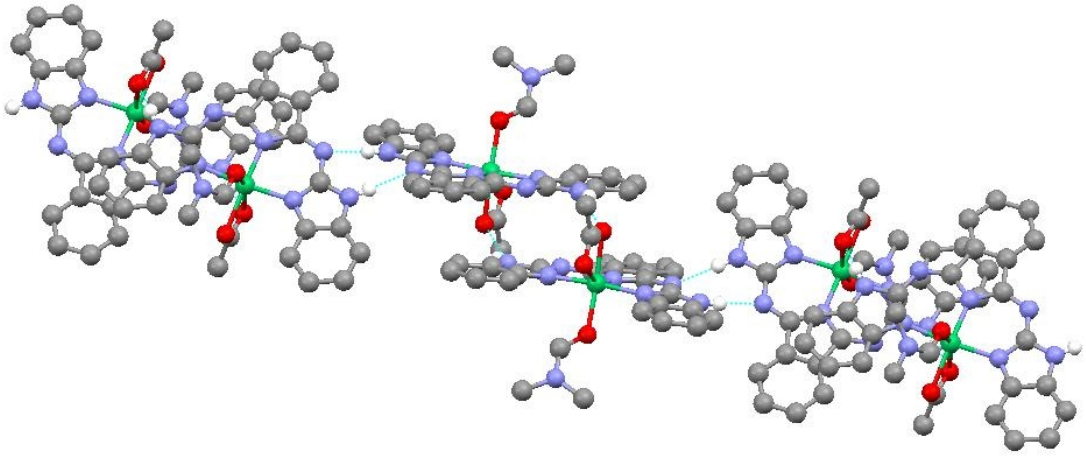

Figure S4. Linear hydrogen bonding network for the crystallographic structure of **4**.

Table S4. Crystal data and structure refinement for **4**.

Identification code	**4**
Empirical formula	$C_{54}H_{48}N_{16}Ni_2O_6$
Formula weight	1134.50
Temperature	100(2) K
Wavelength	0.71073 Å
Crystal system	Triclinic
Space group	P-1
Unit cell dimensions	a = 12.746(2) Å α = 70.321(2)°. b = 13.734(3) Å β = 72.422(2)°. c = 18.698(4) Å γ = 72.583(2)°.
Volume	2865.9(9) $Å^3$
Z	2
Density (calculated)	1.315 Mg/m^3
Absorption coefficient	0.719 mm^{-1}
F(000)	1176
Crystal size	0.15 x 0.15 x 0.08 mm^3
Theta range for data collection	1.61 to 26.00°
Index ranges	-15<=h<=15, -16<=k<=16, -23<=l<=23
Reflections collected	22551
Independent reflections	11176 [R(int) = 0.0592]
Completeness to theta = 26.00°	99.2 %
Absorption correction	Multi-scan/SADABS
Max. and min. transmission	0.7457 and 0.6227
Refinement method	Full-matrix least-squares on F^2
Data / restraints / parameters	11176 / 0 / 709
Goodness-of-fit on F^2	1.174
Final R indices [I>2sigma(I)]	R1 = 0.0564, wR2 = 0.0773

R indices (all data)	R1 = 0.0794, wR2 = 0.0824
Largest diff. peak and hole	0.948 and -0.377 e.Å^{-3}

*Crystal Structure Report for Compound **5***

An orange block-like specimen of **5**, approximate dimensions 0.05×0.07×0.10 mm, was used for the X-ray crystallographic analysis. The X-ray intensity data were measured.

The total exposure time was 8.65 hours. The frames were integrated with the Bruker SAINT software package using a narrow-frame algorithm. The integration of the data using a monoclinic unit cell yielded a total of 24299 reflections to a maximum θ angle of 28.22° (0.75 Å resolution), of which 7247 were independent (average redundancy 3.353, completeness = 99.0%, R_{int} = 6.24%, R_{sig} = 7.43%) and 4591 (63.35%) were greater than 2σ(F^2). The final cell constants of a = 11.896(18) Å, b = 12.751(18) Å, c = 19.61(3) Å, β = 92.960(16)°, volume = 2971.(8) Å^3, are based upon the refinement of the XYZ-centroids of 3287 reflections above 20 σ(I) with 5.091° < 2θ < 49.56°. Data were corrected for absorption effects using the multi-scan method (SADABS). The estimated minimum and maximum transmission as calculated from SADABS are 0.6219 and 0.7457.

The structure was solved and refined using the Bruker SHELXTL Software Package, using the space group P2(1)/c, with Z = 4 for the formula unit, $C_{30}H_{27}CuN_9O_4$. The final anisotropic full-matrix least-squares refinement on F^2 with 410 variables converged at R1 = 4.91%, for the observed data and wR2 = 15.80% for all data. The goodness-of-fit was 0.904. The largest peak in the final difference electron density synthesis was 0.886 e^-/Å^3 and the largest hole was -0.676 e^-/Å^3 with an RMS deviation of 0.084 e^-/Å^3. On the basis of the final model, the calculated density was 1.434 g/cm^3 and F(000), 1324 e^-.

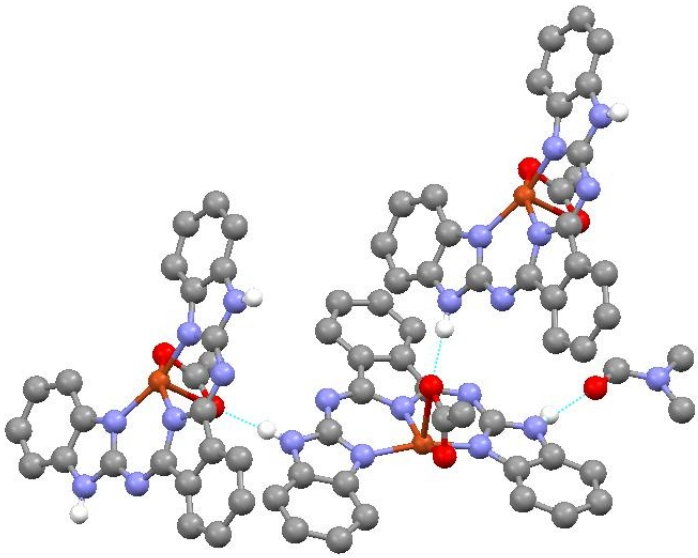

Figure S5. Hydrogen bonding contacts for the crystallographic structure of **5**.

Table S5. Crystal data and structure refinement for **5**.

Identification code	**5**
Empirical formula	$C_{30}H_{27}CuN_9O_4$
Formula weight	641.15
Temperature	100(2) K
Wavelength	0.71073 Å
Crystal system	Monoclinic
Space group	P2(1)/c
Unit cell dimensions	a = 11.896(18) Å α = 90°. b = 12.751(18) Å β = 92.960(16)°. c = 19.61(3) Å γ = 90°.
Volume	2970(8) $Å^3$
Z	4
Density (calculated)	1.434 Mg/m^3
Absorption coefficient	0.788 mm^{-1}
F(000)	1324
Crystal size	0.10 x 0.07 x 0.05 mm^3
Theta range for data collection	1.71 to 28.22°
Index ranges	-15<=h<=14, -16<=k<=14, -25<=l<=25
Reflections collected	24299
Independent reflections	7247 [R(int) = 0.0624]
Completeness to theta = 28.22°	99.0 %
Absorption correction	Multi-scan/SADABS
Max. and min. transmission	0.7457 and 0.6219
Refinement method	Full-matrix least-squares on F^2
Data / restraints / parameters	7247 / 0 / 410
Goodness-of-fit on F^2	0.904
Final R indices [I>2sigma(I)]	R1 = 0.0491, wR2 = 0.1295
R indices (all data)	R1 = 0.0941, wR2 = 0.1580
Largest diff. peak and hole	0.886 and -0.676 $e.Å^{-3}$

*Crystal Structure Report for Compound **6***

A light orange needle-like specimen of **6**, approximate dimensions 0.04×0.05×0.33 mm, was used for the X-ray crystallographic analysis. The X-ray intensity data were measured.

The total exposure time was 15.15 hours. The frames were integrated with the Bruker SAINT software package using a narrow-frame algorithm. The integration of the data using a monoclinic unit cell yielded a total of 22569 reflections to a maximum θ angle of 26.99° (0.78 Å resolution), of which 6098 were independent (average redundancy 3.701, completeness = 99.5%, R_{int} = 11.02%, R_{sig} = 10.72%) and 3733 (61.22%) were greater than $2\sigma(F^2)$. The final cell constants of a = 10.001(4) Å, b = 21.724(8) Å, c = 13.687(5) Å, β = 109.266(5)°, volume = 2807.1(18) $Å^3$, are based upon the refinement of the XYZ-centroids of 4954 reflections above 20 σ(I) with 4.701° < 2θ < 54.65°. Data were corrected for absorption effects using the multi-scan method (SADABS). The estimated minimum and maximum transmission as calculated from SADABS are 0.5750 and 0.7456.

The structure was solved and refined using the Bruker SHELXTL Software Package, using the space group P2(1)/c, with Z = 4 for the formula unit, $C_{28}H_{28}ClN_9O_2Zn$. The final anisotropic full-

matrix least-squares refinement on F^2 with 374 variables converged at R1 = 5.62%, for the observed data and wR2 = 16.40% for all data. The goodness-of-fit was 0.870. The largest peak in the final difference electron density synthesis was 0.462 $e^-/Å^3$ and the largest hole was -0.413 $e^-/Å^3$ with an RMS deviation of 0.099 $e^-/Å^3$. On the basis of the final model, the calculated density was 1.475 g/cm^3 and F(000), 1288 e^-.

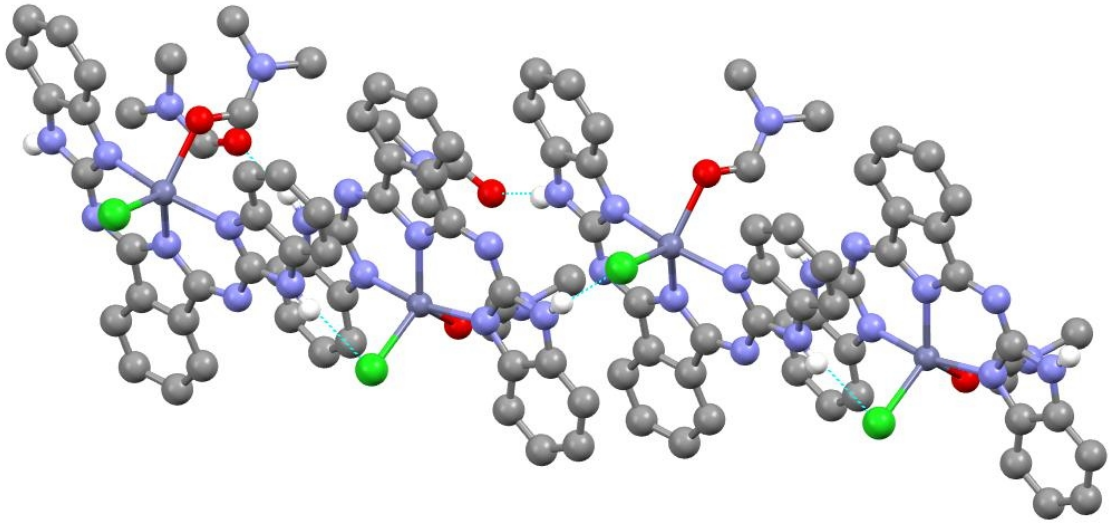

Figure S6. Hydrogen bonding contacts for the crystallographic structure of **6**.

Table S6. Crystal data and structure refinement for **6**.

Identification code	**6**
Empirical formula	$C_{28}H_{28}ClN_9O_2Zn$
Formula weight	623.41
Temperature	100(2) K
Wavelength	0.71073 Å
Crystal system	Monoclinic
Space group	P2(1)/c
Unit cell dimensions	a = 10.001(4) Å α = 90°.
	b = 21.724(8) Å β = 109.266(5)°.
	c = 13.687(5) Å γ = 90°.
Volume	2807.1(18) $Å^3$
Z	4
Density (calculated)	1.475 Mg/m^3
Absorption coefficient	1.014 mm^{-1}
F(000)	1288
Crystal size	0.33 x 0.05 x 0.04 mm^3
Theta range for data collection	1.83 to 26.99°
Index ranges	-12<=h<=12, -27<=k<=27, -17<=l<=17
Reflections collected	22569
Independent reflections	6098 [R(int) = 0.1102]

Completeness to theta = 26.99°	99.5 %
Absorption correction	Multi-scan/SADABS
Max. and min. transmission	0.7456 and 0.5750
Refinement method	Full-matrix least-squares on F^2
Data / restraints / parameters	6098 / 0 / 374
Goodness-of-fit on F^2	0.870
Final R indices [I>2sigma(I)]	R1 = 0.0562, wR2 = 0.1272
R indices (all data)	R1 = 0.1137, wR2 = 0.1640
Largest diff. peak and hole	0.462 and -0.413 e.$Å^{-3}$

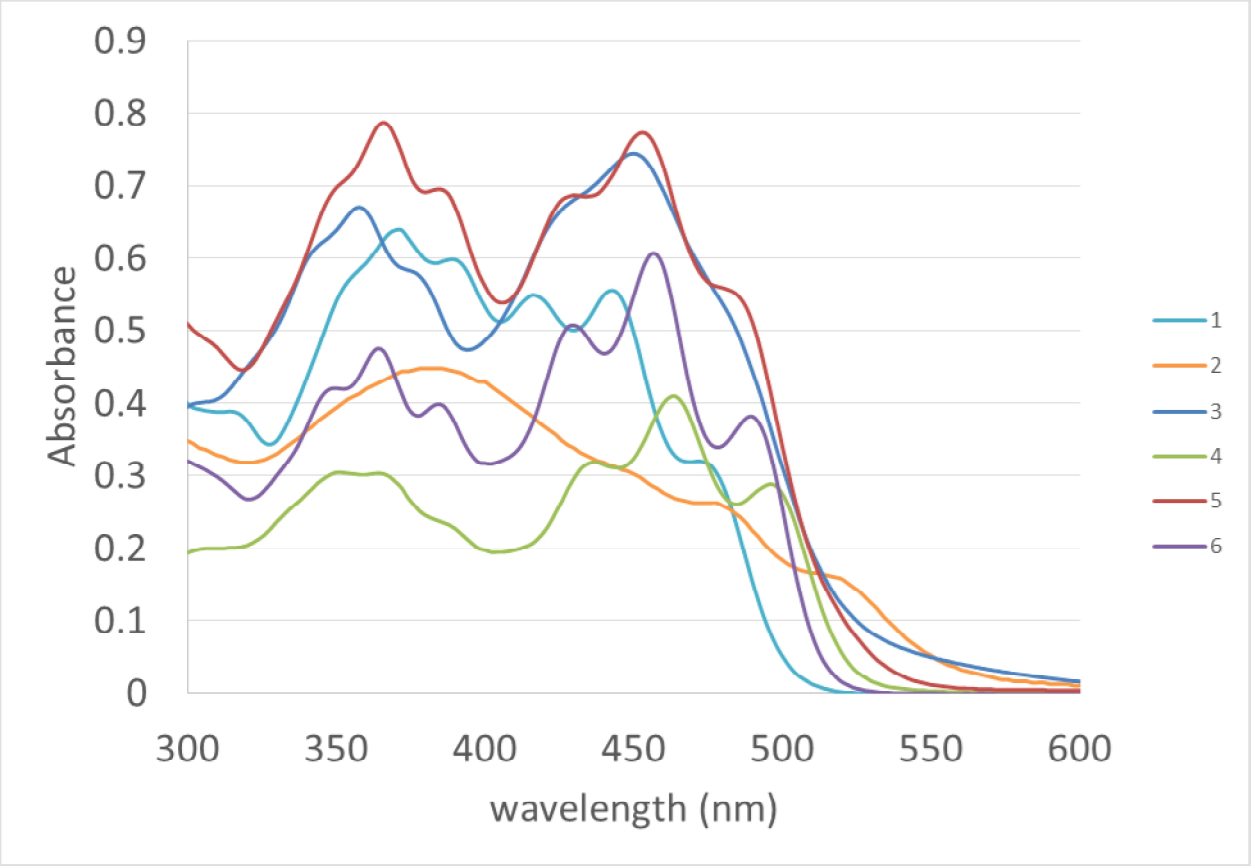

Figure S7. UV-visible spectra for compounds **1**-**6**.