Supporting Information

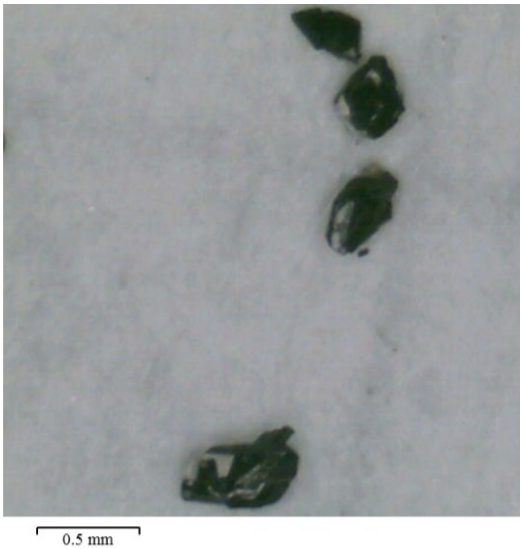

Figure S1. Photo of the crystalline sample of [Fe(Hsemsal)(semsal)] · $3H_2O$ **(1)**. The scale bar represents 0.5 mm.

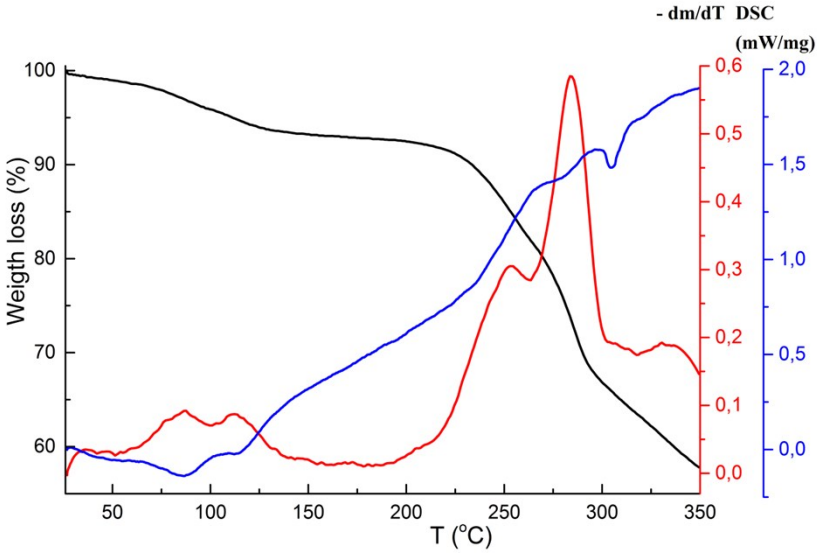

Figure S2. Thermogram of [Fe(Hsemsal)(semsal)]·$3H_2O$ (**1**) (normal curve (black), differential curve (red)) and DSC curve (blue).

Approximately 5,6 mg of sample were placed into an Al_2O_3 crucible with a pre-hole on the lid and then heated from 30 to 350 °C. The thermogravimetric analysis were performed in high-purity argon atmosphere with a gas flow rate of 40 mL/min, at the heating rate of 5 °C min^{-1}.

This figure demonstrates that temperature increasing leads to the gradual loss of water molecules. The weight loss of 9.11% (calc. 9.98%, 3 mol H_2O) is observed in the temperature range 75-200°C with *DSC* endothermic peaks at 86.0 and 113.8°C, assigned to the loss of lattice water molecules, and the next weight-loss step appears at a temperature above 200° C with an endothermic peak at 280 °C, which corresponds to the decomposition of the complex. Note that

the hydrogen bonding between the Fe complexes in crustal structure of **1** is observed with water mediation (see Figure 4, Table 2 in text). Before getting a full characterization the sample of complex [Fe(Hsemsal)(semsal)]·3H_2O was dried in vacuum for several hours to remove as much solvent as possible.

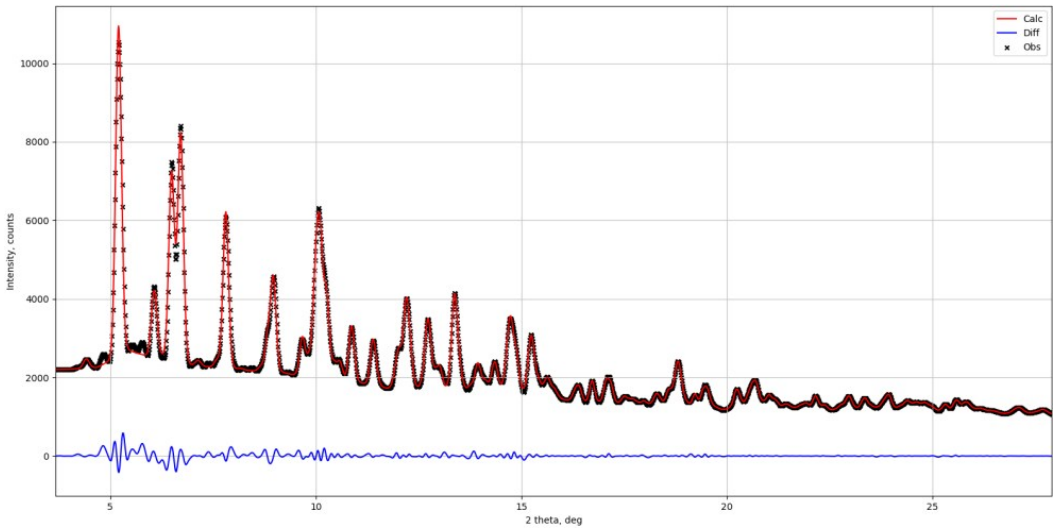

Figure S3. Powder diffractogram (XRPD) in the 2θ range from 1.95° to 30° of sample **1**. Rietveld fitting of the powder XRD data of Fe(Hsemsal)(semsal)]·3H_2O (black, experimental data; red line, calculated data; blue line, difference plot).

Powder patterns were used as a fingerprint for identification of the crystalline phases present in a material. The adequacy of unit cell and space group for **1** was confirmed by the Rietveld method: monoclinic, space group $P2_1/c$ (no. 14), a = 17.422(2) Å, b = 12.6736(15) Å, c = 9.2305(9) Å, β = 93.22(6)°, T = 293 K.

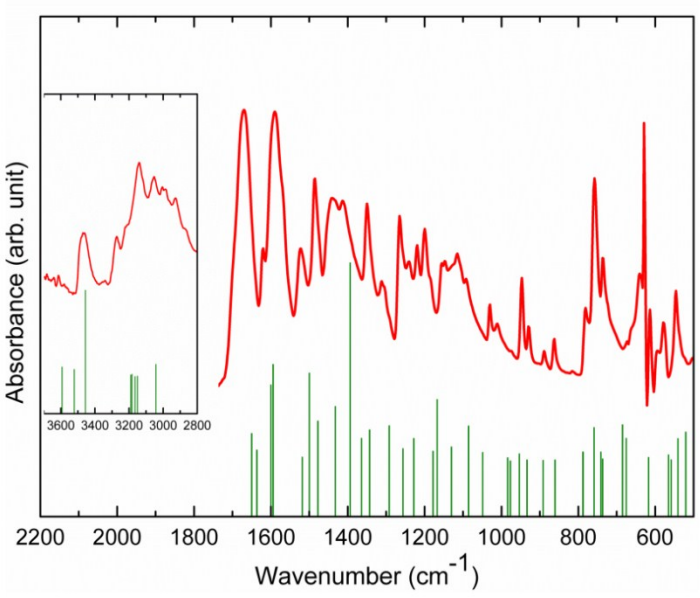

Figure S4. ATR FT-IR-spectrum for the H_2semsal (red line) over the ranges 3700–2800 cm^{-1} and 1750–500cm^{-1}. Calculated (B3LYP*/6-311++G(d,p)) IR vibrational frequencies of the H_2semsal (green bars).

The atomic coordinates for starting geometry of the H_2semsal were taken from the single crystal structural data.

1. Valdés-Martínez, J.; Toscano, R. A.; Salcedo, R.; Cea-Olivares, R.; Meléndez, A. Semicarbazones and thiosemicarbazones, XII: Crystal structure of salicylaldehyde semicarbazone. *Monatshefte für Chemie*. **1990**, 121, 8–9, 641–647.

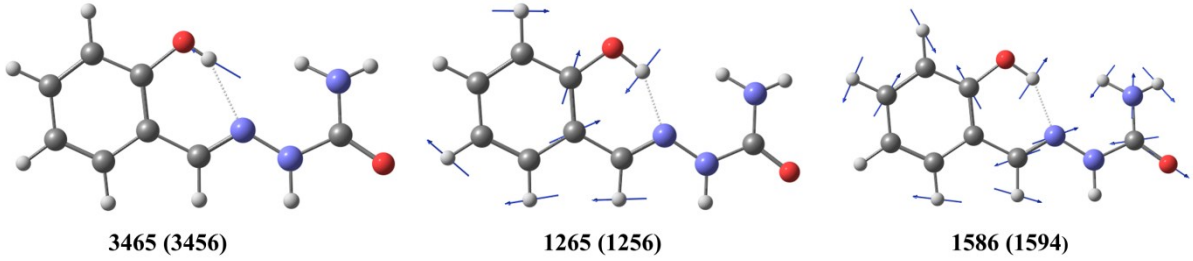

Figure S5. Selected experimental and calculated (in round brackets) IR vibrational modes of the ligand H_2semsal (B3LYP*/6-311++G(d,p)).

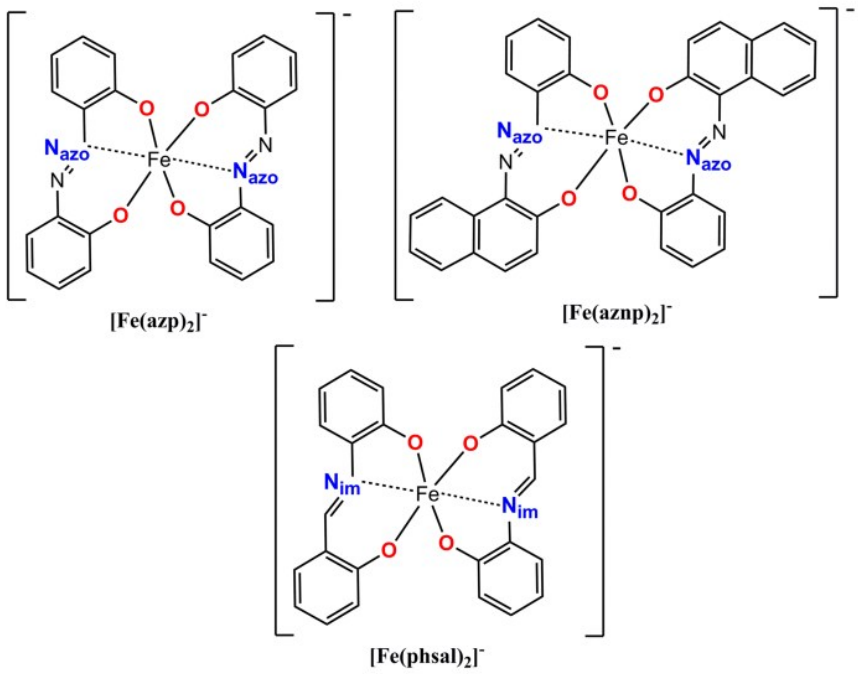

Figure S6. Molecular structures of $[Fe^{III}(azp)_2]^-$, $[Fe^{III}(aznp)_2]^-$ and $[Fe^{III}(phsal)_2]^-$ anionic complexes with N_2O_4 coordination environment.

Table S1. Bond Lengths for [Fe(Hsemsal)(semsal)]·3H_2O.

Atom	Atom	Length/Å	Atom	Atom	Length/Å
Fe1	O1	1.888(3)	N5	C16	1.328(4)
Fe1	O2	1.937(2)	N6	C16	1.360(4)
Fe1	O3	2.030(2)	C1	C2	1.407(5)
Fe1	O4	2.109(2)	C1	C6	1.421(4)
Fe1	N1	2.134(3)	C2	C3	1.385(5)
Fe1	N4	2.110(3)	C3	C4	1.395(5)
O1	C1	1.325(4)	C4	C5	1.378(5)
O2	C8	1.330(4)	C5	C6	1.410(4)
O3	C16	1.293(4)	C6	C7	1.442(4)
O4	C15	1.273(4)	C8	C9	1.402(4)
N1	N2	1.381(3)	C8	C13	1.419(4)
N1	C14	1.288(4)	C9	C10	1.388(5)
N2	C15	1.341(4)	C10	C11	1.397(5)
N3	C15	1.328(4)	C11	C12	1.380(5)
N4	N5	1.397(4)	C12	C13	1.403(4)
N4	C7	1.293(4)	C13	C14	1.446(4)

Table S2. Calculated (B3LYP*/6-311++G(d,p,)) and experimental (in brackets) bond lengths of Fe(III) coordinated octahedron for the neutral complexes **(I)**.

Type of Schiff base	Bound	X(S)			X(Se)			X(O)		
		HS	LS	Δ_{HS-LS}, %	HS	LS	Δ_{HS-LS}, %	HS	LS	Δ_{HS-LS}, %
A	Fe-O	1.95740 [1.927(5)]*	1.89800	3.13	1.96533	1.90571	3.13	1.94590 [1.936(3)]	1.86793	4.17
	Fe-**N**	2.24511 [2.111(6)]*	1.95963	14.57	2.25775	1.96962	14.63	2.21148 [2.134(3)]	1.94173	13.89
	Fe-**X**	2.62896 [2.444(7)]*	2.37480	10.70	2.73169	2.48507	9.92	2.22536 [2.110(3)]	2.05905	8.08
B	Fe-O	1.92668 [1.894(5)]*	1.90150	1.32	1.93441	1.90965	1.30	1.90534 [1.889(3)]	1.85875	2.51
	Fe-**N**	2.16541 [2.166(6)]*	1.95201	10.93	2.18245	1.96387	11.13	2.12746 [2.109(3)]	1.91916	10.85
	Fe-**X**	2.43178 [2.370(8)]*	2.29065	6.16	2.54192	2.41432	5.29	2.02928 [2.031(3)]	1.93378	4.94

*- [20, in text]. The bond length increase was calculated in percentage (%) regarding the LS state.

Table S3. The experimental bond lengths of Fe(III) coordinated octahedron for [Fe(Hth5Br-sal)(th5Br-sal)] · H_2O neutral complex (**I**) at a different temperature [21, in text].

Type of Schiff base	Bound	Fe(Hth5Br-sal)(th5Br-sal)] · H_2O		
		HS 303 K	LS 123 K	Δ_{HS-LS}, %
A	Fe-O	1.969(4)	1.941(4)	1.44
	Fe-**N**	2.153(4)	1.952(5)	10.30
	Fe-**S**	2.4845(18)	2.2718(17)	9.36
B	Fe-O	1.952(4)	1.949(4)	0.15
	Fe-**N**	2.131(4)	1.955(4)	9.00
	Fe-**S**	2.4177(19)	2.2463(17)	7.63

The bound length increase was calculated in percentage (%) regarding the LS state.

Table S4. Calculated (B3LYP*/6-311++G(d,p,)) values of electronic energy (E_{el}), zero-point vibration energy (E_{ZPV}) and total energy difference between HS and LS states for the Fe(III) neutral (**I**) and anionic (**II**) complexes (ΔE_0 (HS-LS)). Hammet parameters (σ_p) for substituents R=5OMe-, H-, 5Cl-, $5NO_2$- in the benzene ring of salicylaldehyde.

X	*R*-	E_{el}, a.u.		E_{ZPV}, a.u.		ΔE_{el} (HS-LS), kJ/mol	ΔE_0 (HS-LS), kJ/mol	σ_p
		HS	LS	HS	LS			
S	**(I)** *5OMe-*	-3386.1656059	-3386.169346	0.363799	0.366667	9.82	2.29	-0.27
	(I) *H-*	-3157.18794870	-3157.192377	0.30003	0.30285	11.63	**4.22**	0
	(II) *H-*	-3156.688924	-3156.696489	0.287413	0.290232	19.86	**12.46**	0
	(I) *5Cl-*	-4076.227084	-4076.232219	0.280664	0.283528	13.48	5.96	0.23
	(I) *5NO_2-*	-3566.132347	-3566.139071	0.304185	0.307103	17.65	9.99	0.78
Se	**(I)** *5OMe-*	-7392.544127	-7392.5471797	0.361868	0.364539	8.01	1.00	-0.27
	(I) *H-*	-7163.566561	-7163.57032	0.298087	0.300725	9.87	**2.94**	0
	(II) *H-*	-7163.070181	-7163.076443	0.285641	0.28817	16.44	**9.80**	0
	(I) *5Cl-*	-8082.605652	-8082.610122	0.278735	0.2814	11.74	4.74	0.23
	(I) *5NO_2-*	-7572.511316	-7572.517375	0.3022	0.304906	15.91	8.81	0.78
O	**(I)** *5OMe-*	-2740.3803035	-2511.3985388	0.369209	0.372305	-11.56	-19.69	-0.27
	(I) *H-*	-2511.402611	-2511.398539	0.305479	0.308517	-10.69	**-18.67**	0
	(II) *H-*	-2510.89526	-2510.894173	0.292652	0.295775	-2.85	**-11.05**	0
	(I) *5Cl-*	-3430.442317	-3430.438567	0.286148	0.289209	-9.85	-17.88	0.23
	(I) *5NO_2*	-2920.347477	-2920.344497	0.309703	0.312788	-7.83	-15.93	0.78

ΔE_0 (HS-LS) [**II-I**] = 12.46 - 4.22 = 8.24 (kJ/mol) for X=S
ΔE_0 (HS-LS) [**II-I**] = 9.80 - 2.94 = 6.86 (kJ/mol) for X=Se
ΔE_0 (HS-LS) [**II-I**] = -11.05 - (-18.67) = 7.62 (kJ/mol) for X=O

Table S5. Calculated (B3LYP*/6-311++G(d,p,)) and experimental (in brackets) bond lengths of Fe(III) coordinated octahedron for the anionic complexes **(II)**.

Type of Schiff base	Bound	**X(S)**			**X(Se)**			**X(O)**		
		HS	LS	Δ_{HS-LS}, %	HS	LS	Δ_{HS-LS}, %	HS	LS	Δ_{HS-LS}, %
B	Fe-O	1.97069 [1.968]*	1.92775	2.23	1.97360	1.93371	2.06	1.95334	1.88939	3.38
	Fe-**N**	2.21752 [2.180]*	1.96430	12.89	2.23524	1.97637	13.10	2.18246	1.93507	12.78
	Fe-**X**	2.48896 [2.4087]*	2.30943	7.77	2.60828	2.43271	7.22	2.06707	1.95596	5.68

*- The bond lengths and atomic coordinates for starting geometry of the anionic complexes (**II**) were taken from the single crystal structural data.
1. Meetsma, A. CCDC 787172: Experimental Crystal Structure Determination. **2016** DOI: 10.5517/ccdc.csd.ccvf3n0.

The bond length increase was calculated in percentage (%) regarding the LS state.

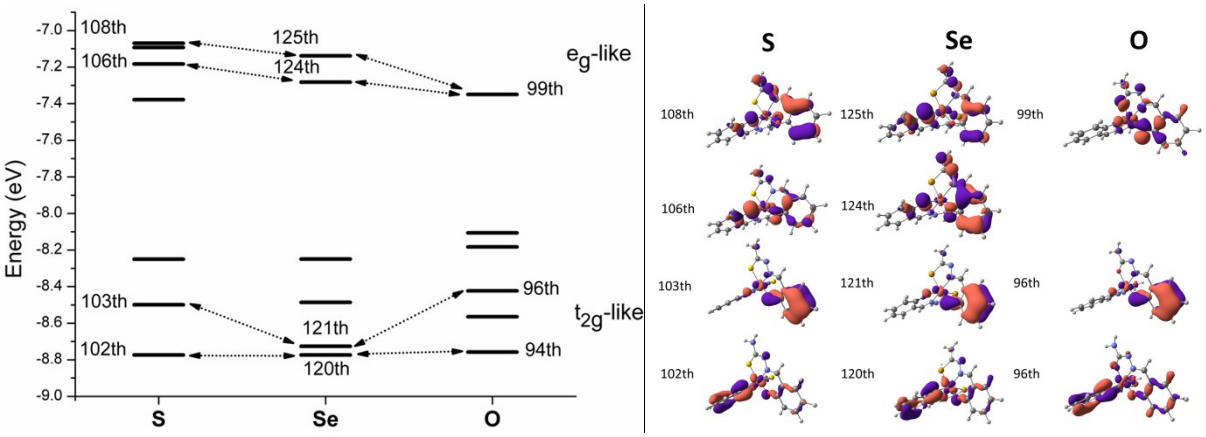

Figure S7. Energy-level diagram of α-spin molecular orbitals (MOs) (B3LYP*/6-311 ++G(d,p)) for Fe (III) neutral complexes: X = O, S, Se (left). The numbers and double arrows mark the corresponding MOs of the complexes containing the contribution of Fe d-orbitals (e_g-like and t_{2g}-like). The surfaces of the selected MOs are represented on the right. A contour of 0.04 eÅ^{-3} has been used for the MOs plots.

The iron e_g-like orbitals were included into: the 108th and 106th MOs for the HS state of the neutral complex with X=S; 125th and 124th MOs for the HS state of the neutral complex with X=Se; 99th MO for the HS state of the neutral complex with X=O. As for the iron t_{2g}-like orbitals, they were found in: the 103th and 102th MOs for the HS state of the neutral complex with X=S; 121th and 120th MOs for the HS state of the neutral complex with X=Se; 96th and 94th MOs for the HS state of the neutral complex with X=O.

Table S6. Optimized parameters of [Fe(Hsem5OMe-sal)(sem5OMe-sal)] neutral complex in the sextet state (**HS**). Units are in **Å.**

Fe	0.041102000	0.690114000	0.091492000
O	-0.593706000	2.103685000	1.416541000
O	0.622170000	2.370763000	-1.259670000
O	1.211659000	-0.456833000	-0.878272000
O	-1.139481000	-0.710013000	0.754814000
N	1.668012000	0.948213000	1.446043000
N	1.444422000	1.800406000	2.494414000
N	-0.125524000	3.277078000	3.313562000
H	0.415705000	3.279065000	4.166818000
H	-1.117332000	3.443355000	3.401324000
C	3.283410000	-0.500279000	0.335745000
C	2.451230000	-0.878257000	-0.757676000
C	2.988213000	-1.749776000	-1.733447000
H	2.346359000	-2.036172000	-2.560680000
C	4.280514000	-2.229504000	-1.641518000
H	4.684588000	-2.900194000	-2.393677000
C	5.101807000	-1.859119000	-0.560860000
C	4.603437000	-1.005353000	0.412637000
H	5.214853000	-0.704010000	1.256387000
C	2.843722000	0.395534000	1.374866000
H	3.555249000	0.648800000	2.164484000
C	0.239341000	2.347640000	2.368477000
N	-1.654778000	1.078850000	-1.265307000
N	-1.436865000	2.094375000	-2.181199000
N	-0.061712000	3.845378000	-2.864140000
H	-0.572045000	3.917288000	-3.733268000
H	0.877927000	4.218062000	-2.868800000
C	-3.256701000	-0.503419000	-0.365989000
C	-2.375960000	-1.075184000	0.606423000
C	-2.906687000	-2.105454000	1.433136000
H	-2.242706000	-2.542974000	2.171424000
C	-4.206519000	-2.535903000	1.305084000
H	-4.597653000	-3.323889000	1.942118000
C	-5.071541000	-1.968699000	0.341661000
C	-4.598408000	-0.966533000	-0.480138000
H	-5.236601000	-0.510794000	-1.230238000
C	-2.838114000	0.542472000	-1.244393000
H	-3.592789000	0.915559000	-1.947919000
C	-0.235373000	2.752408000	-2.059079000
H	-2.243630000	2.573154000	-2.572419000
O	6.366377000	-2.394063000	-0.565337000
C	7.235479000	-2.061737000	0.506890000
H	7.430168000	-0.981582000	0.547594000
H	6.828770000	-2.392499000	1.471931000
H	8.170227000	-2.588926000	0.310219000
O	-6.342304000	-2.487572000	0.315390000
C	-7.254844000	-1.959982000	-0.634123000
H	-7.430546000	-0.887849000	-0.470385000
H	-6.900557000	-2.116539000	-1.662242000
H	-8.189291000	-2.503787000	-0.489007000

Table S7. Optimized parameters of [Fe(Hsem5OMe-sal)(sem5OMe-sal)] neutral complex in the doublet state (**LS**). Units are in **Å**.

Fe	0.011771000	1.167883000	0.042562000
O	-0.667764000	2.537687000	1.239118000
O	0.571384000	2.771889000	-1.131272000
O	0.877514000	0.069263000	-1.174697000
O	-0.704141000	-0.216592000	1.068409000
N	1.491116000	1.235331000	1.263235000
N	1.365006000	2.141873000	2.290098000
N	-0.118297000	3.714167000	3.113933000
H	0.410208000	3.680393000	3.974402000
H	-1.103534000	3.917680000	3.202595000
C	2.782811000	-0.520953000	0.215656000
C	1.943814000	-0.677044000	-0.922160000
C	2.294733000	-1.665835000	-1.869801000
H	1.651855000	-1.779265000	-2.737203000
C	3.409831000	-2.465671000	-1.709500000
H	3.665227000	-3.224341000	-2.443050000
C	4.239504000	-2.309339000	-0.585276000
C	3.925914000	-1.344912000	0.359939000
H	4.549181000	-1.198994000	1.235606000
C	2.536473000	0.470817000	1.232298000
H	3.264709000	0.596912000	2.033917000
C	0.195647000	2.751543000	2.182506000
N	-1.527079000	1.249970000	-1.134770000
N	-1.442662000	2.281634000	-2.060557000
N	-0.226568000	4.138654000	-2.777000000
H	-0.700433000	4.124113000	-3.669395000
H	0.671874000	4.602523000	-2.762176000
C	-2.721388000	-0.630107000	-0.245909000
C	-1.794178000	-0.893119000	0.811749000
C	-2.098292000	-1.982244000	1.677142000
H	-1.398686000	-2.182468000	2.481875000
C	-3.220283000	-2.756760000	1.505556000
H	-3.431091000	-3.587340000	2.172725000
C	-4.130254000	-2.494750000	0.456321000
C	-3.880547000	-1.444431000	-0.401116000
H	-4.560109000	-1.214179000	-1.215038000
C	-2.548022000	0.451552000	-1.163427000
H	-3.325057000	0.611611000	-1.916367000
C	-0.313213000	3.053219000	-1.951609000
H	-2.302697000	2.667525000	-2.436861000
O	5.323084000	-3.153158000	-0.521390000
C	6.190550000	-3.037895000	0.595377000
H	6.653469000	-2.042922000	0.645748000
H	5.663172000	-3.238484000	1.537854000
H	6.968411000	-3.789438000	0.451198000
O	-5.211231000	-3.338468000	0.390341000
C	-6.154792000	-3.127223000	-0.647681000
H	-6.623508000	-2.136514000	-0.569756000
H	-5.692300000	-3.232533000	-1.638641000
H	-6.917956000	-3.896704000	-0.523076000

Table S8. Optimized parameters of [Fe(Hsemsal)(semsal)] neutral complex in the sextet state (**HS**). Units are in **Å.**

Fe	0.041746000	0.368093000	0.089118000
O	-0.652491000	1.717592000	1.436292000
O	0.665561000	2.098931000	-1.162796000
O	1.256522000	-0.729114000	-0.885960000
O	-1.153346000	-1.064731000	0.641658000
N	1.599556000	0.557133000	1.525635000
N	1.330352000	1.369201000	2.596970000
N	-0.274224000	2.822600000	3.394232000
H	0.225383000	2.793789000	4.272094000
H	-1.269547000	2.984513000	3.441546000
C	3.254331000	-0.864935000	0.437782000
C	2.479252000	-1.181218000	-0.721832000
C	3.051006000	-2.012078000	-1.709733000
H	2.447651000	-2.244452000	-2.581816000
C	4.334643000	-2.517795000	-1.565483000
H	4.748198000	-3.157474000	-2.340700000
C	5.098444000	-2.213435000	-0.429135000
C	4.555254000	-1.397455000	0.551476000
H	5.137535000	-1.153245000	1.437029000
C	2.770370000	-0.008002000	1.490325000
H	3.443992000	0.200861000	2.324726000
C	0.136200000	1.925843000	2.436682000
N	-1.610208000	0.819809000	-1.310041000
N	-1.362299000	1.880127000	-2.164162000
N	0.041207000	3.656608000	-2.710203000
H	-0.447690000	3.786345000	-3.584759000
H	0.979938000	4.028431000	-2.663096000
C	-3.225957000	-0.818866000	-0.549610000
C	-2.377097000	-1.438702000	0.430223000
C	-2.922194000	-2.514382000	1.180778000
H	-2.278223000	-2.981332000	1.918913000
C	-4.217569000	-2.947762000	0.976735000
H	-4.601065000	-3.774444000	1.569222000
C	-5.048601000	-2.339043000	0.015175000
C	-4.548337000	-1.291423000	-0.730339000
H	-5.174884000	-0.809354000	-1.478193000
C	-2.786167000	0.274675000	-1.360357000
H	-3.519880000	0.673174000	-2.071864000
C	-0.163732000	2.526103000	-1.969794000
H	-2.154327000	2.375253000	-2.564784000
H	6.101811000	-2.611784000	-0.317313000
H	-6.064002000	-2.689623000	-0.136020000

Table S9. Optimized parameters of [Fe(Hsemsal)(semsal)] neutral complex in the doublet state (**LS**). Units are in **Å**.

Fe	-0.011064000	0.681889000	0.051991000
O	-0.855894000	1.993481000	1.194572000
O	0.616386000	2.320190000	-1.025946000
O	1.009883000	-0.363841000	-1.096511000
O	-0.776105000	-0.751447000	0.973632000
N	1.333288000	0.767254000	1.418961000
N	1.062883000	1.635190000	2.452577000
N	-0.558738000	3.131875000	3.147984000
H	-0.124901000	3.095543000	4.059730000
H	-1.554594000	3.299004000	3.132751000
C	2.796777000	-0.904067000	0.460284000
C	2.078795000	-1.069273000	-0.763241000
C	2.557653000	-2.015334000	-1.696148000
H	2.001657000	-2.129779000	-2.621667000
C	3.692406000	-2.770753000	-1.442006000
H	4.030981000	-3.492952000	-2.180290000
C	4.402033000	-2.608421000	-0.243612000
C	3.951431000	-1.682552000	0.684114000
H	4.493428000	-1.543012000	1.616720000
C	2.406942000	0.046461000	1.472626000
H	3.046347000	0.174299000	2.346435000
C	-0.108021000	2.208089000	2.234721000
N	-1.424023000	0.742805000	-1.278477000
N	-1.265796000	1.786504000	-2.179084000
N	-0.029866000	3.684516000	-2.737273000
H	-0.411313000	3.678252000	-3.672852000
H	0.847969000	4.172409000	-2.619594000
C	-2.653426000	-1.174856000	-0.530435000
C	-1.821863000	-1.445474000	0.610017000
C	-2.177741000	-2.555240000	1.423815000
H	-1.547933000	-2.754948000	2.284528000
C	-3.268558000	-3.347580000	1.130942000
H	-3.502166000	-4.190202000	1.776444000
C	-4.081169000	-3.081500000	0.010625000
C	-3.768227000	-2.008644000	-0.796146000
H	-4.384507000	-1.784754000	-1.664484000
C	-2.417319000	-0.075688000	-1.415256000
H	-3.122212000	0.074045000	-2.238100000
C	-0.178671000	2.587586000	-1.938575000
H	-2.083769000	2.140488000	-2.664517000
H	5.290975000	-3.198216000	-0.043865000
H	-4.936959000	-3.709887000	-0.212153000

Table S10. Optimized parameters of [Fe(Hsem5Cl-sal)(sem5Cl-sal)] neutral complex in the sextet state (**HS**). Units are in **Å.**

Fe	0.041674000	0.750022000	0.084621000
O	-0.658160000	2.147121000	1.375416000
O	0.670465000	2.414803000	-1.234265000
O	1.256736000	-0.395856000	-0.832293000
O	-1.157860000	-0.660115000	0.689365000
N	1.598522000	0.999769000	1.519394000
N	1.325025000	1.854285000	2.552027000
N	-0.285903000	3.330779000	3.286232000
H	0.217662000	3.354270000	4.161670000
H	-1.278103000	3.512342000	3.318075000
C	3.254357000	-0.465644000	0.496172000
C	2.479240000	-0.836499000	-0.646372000
C	3.053375000	-1.712432000	-1.592787000
H	2.453839000	-1.989770000	-2.453881000
C	4.336859000	-2.212015000	-1.433696000
H	4.761909000	-2.886151000	-2.169687000
C	5.083456000	-1.842331000	-0.310181000
C	4.556742000	-0.986084000	0.639584000
H	5.148383000	-0.707926000	1.506193000
C	2.770757000	0.436244000	1.511879000
H	3.442786000	0.681688000	2.337058000
C	0.127269000	2.400195000	2.367590000
N	-1.611237000	1.145508000	-1.337541000
N	-1.354226000	2.165705000	-2.233676000
N	0.065669000	3.904939000	-2.852265000
H	-0.431402000	4.016725000	-3.724467000
H	1.003774000	4.279221000	-2.815599000
C	-3.232001000	-0.450027000	-0.505341000
C	-2.383775000	-1.032646000	0.495631000
C	-2.934731000	-2.073858000	1.290387000
H	-2.294987000	-2.516873000	2.046373000
C	-4.231262000	-2.512580000	1.114273000
H	-4.628960000	-3.309860000	1.733822000
C	-5.045259000	-1.928728000	0.128037000
C	-4.558104000	-0.917418000	-0.668113000
H	-5.193117000	-0.472322000	-1.428534000
C	-2.790260000	0.607137000	-1.364817000
H	-3.522961000	0.978816000	-2.091093000
C	-0.151754000	2.812515000	-2.064358000
H	-2.139459000	2.637512000	-2.673821000
Cl	6.716908000	-2.477523000	-0.105831000
Cl	-6.698481000	-2.501755000	-0.083496000

Table S11. Optimized parameters of [Fe(Hsem5Cl-sal)(sem5Cl-sal)] neutral complex in the doublet state (**LS**). Units are in **Å**.

Fe	0.008902000	1.189657000	0.045254000
O	-0.764101000	2.547209000	1.181888000
O	0.668204000	2.779294000	-1.072932000
O	0.960608000	0.083329000	-1.106013000
O	-0.781389000	-0.199953000	1.012706000
N	1.388149000	1.253336000	1.378188000
N	1.175017000	2.150155000	2.396903000
N	-0.377041000	3.712267000	3.102062000
H	0.084558000	3.692146000	4.000308000
H	-1.362694000	3.930836000	3.103151000
C	2.765114000	-0.487680000	0.418540000
C	2.011121000	-0.651962000	-0.782642000
C	2.434202000	-1.633961000	-1.704597000
H	1.854981000	-1.751385000	-2.614952000
C	3.545697000	-2.428057000	-1.469345000
H	3.849780000	-3.179310000	-2.190409000
C	4.275562000	-2.251910000	-0.290091000
C	3.897622000	-1.299098000	0.637095000
H	4.475133000	-1.170024000	1.547145000
C	2.437646000	0.496565000	1.421644000
H	3.103211000	0.619243000	2.275993000
C	0.016938000	2.758664000	2.197808000
N	-1.433719000	1.277394000	-1.251765000
N	-1.259259000	2.298936000	-2.171811000
N	0.036871000	4.136040000	-2.794120000
H	-0.380945000	4.143714000	-3.713821000
H	0.932346000	4.597067000	-2.705328000
C	-2.710801000	-0.578726000	-0.434805000
C	-1.860390000	-0.859511000	0.687474000
C	-2.236587000	-1.941836000	1.529168000
H	-1.595037000	-2.153718000	2.377916000
C	-3.360897000	-2.701803000	1.285946000
H	-3.618784000	-3.525330000	1.943703000
C	-4.179108000	-2.410455000	0.180511000
C	-3.863657000	-1.370584000	-0.661664000
H	-4.501510000	-1.149584000	-1.512284000
C	-2.460583000	0.496123000	-1.347785000
H	-3.181993000	0.657958000	-2.153325000
C	-0.136649000	3.062287000	-1.973251000
H	-2.070461000	2.662955000	-2.661107000
Cl	5.694453000	-3.257492000	0.014280000
Cl	-5.614931000	-3.387797000	-0.118928000

Table S12. Optimized parameters of [Fe(Hsem5NO$_2$-sal)(sem5NO$_2$-sal)] neutral complex in the sextet state (**HS**). Units are in **Å**.

Fe	0.042253000	0.915373000	0.081372000
O	-0.657961000	2.279869000	1.378473000
O	0.646667000	2.549277000	-1.240068000
O	1.255437000	-0.243924000	-0.844660000
O	-1.138278000	-0.522082000	0.690153000
N	1.601405000	1.150915000	1.509730000
N	1.335088000	2.008063000	2.540400000
N	-0.279841000	3.478119000	3.278010000
H	0.236647000	3.533777000	4.144115000
H	-1.268367000	3.677473000	3.305986000
C	3.241675000	-0.337434000	0.492579000
C	2.462252000	-0.702111000	-0.656199000
C	3.023369000	-1.589558000	-1.606697000
H	2.416829000	-1.855424000	-2.465952000
C	4.294063000	-2.104407000	-1.446752000
H	4.724834000	-2.785015000	-2.171349000
C	5.042291000	-1.739158000	-0.318888000
C	4.529410000	-0.872518000	0.635557000
H	5.139628000	-0.613380000	1.493794000
C	2.767042000	0.573522000	1.505529000
H	3.440902000	0.810456000	2.331080000
C	0.134062000	2.548145000	2.366151000
N	-1.636224000	1.293926000	-1.322901000
N	-1.390607000	2.318259000	-2.213104000
N	0.038825000	4.044191000	-2.847723000
H	-0.496217000	4.204230000	-3.688911000
H	0.973483000	4.426762000	-2.814907000
C	-3.221351000	-0.332897000	-0.486443000
C	-2.352180000	-0.914735000	0.506291000
C	-2.874695000	-1.975652000	1.300970000
H	-2.215302000	-2.410589000	2.044167000
C	-4.161287000	-2.433071000	1.136753000
H	-4.554320000	-3.240035000	1.744273000
C	-4.990213000	-1.847386000	0.161272000
C	-4.530781000	-0.817096000	-0.636885000
H	-5.198042000	-0.391319000	-1.379084000
C	-2.805002000	0.738768000	-1.346496000
H	-3.550499000	1.097982000	-2.065090000
C	-0.182552000	2.957684000	-2.062106000
H	-2.173946000	2.766756000	-2.679760000
N	6.391631000	-2.279181000	-0.144852000
O	7.025949000	-1.941917000	0.856205000
O	6.821637000	-3.042592000	-1.011240000
N	-6.358949000	-2.331154000	-0.013595000
O	-6.739651000	-3.248928000	0.712857000
O	-7.056316000	-1.794193000	-0.877169000

Table S13. Optimized parameters of [Fe(Hsem5NO$_2$-sal)(sem5NO$_2$-sal)] neutral complex in the doublet state (**LS**). Units are in **Å**.

Fe	0.042253000	0.915373000	0.081372000
O	-0.657961000	2.279869000	1.378473000
O	0.646667000	2.549277000	-1.240068000
O	1.255437000	-0.243924000	-0.844660000
O	-1.138278000	-0.522082000	0.690153000
N	1.601405000	1.150915000	1.509730000
N	1.335088000	2.008063000	2.540400000
N	-0.279841000	3.478119000	3.278010000
H	0.236647000	3.533777000	4.144115000
H	-1.268367000	3.677473000	3.305986000
C	3.241675000	-0.337434000	0.492579000
C	2.462252000	-0.702111000	-0.656199000
C	3.023369000	-1.589558000	-1.606697000
H	2.416829000	-1.855424000	-2.465952000
C	4.294063000	-2.104407000	-1.446752000
H	4.724834000	-2.785015000	-2.171349000
C	5.042291000	-1.739158000	-0.318888000
C	4.529410000	-0.872518000	0.635557000
H	5.139628000	-0.613380000	1.493794000
C	2.767042000	0.573522000	1.505529000
H	3.440902000	0.810456000	2.331080000
C	0.134062000	2.548145000	2.366151000
N	-1.636224000	1.293926000	-1.322901000
N	-1.390607000	2.318259000	-2.213104000
N	0.038825000	4.044191000	-2.847723000
H	-0.496217000	4.204230000	-3.688911000
H	0.973483000	4.426762000	-2.814907000
C	-3.221351000	-0.332897000	-0.486443000
C	-2.352180000	-0.914735000	0.506291000
C	-2.874695000	-1.975652000	1.300970000
H	-2.215302000	-2.410589000	2.044167000
C	-4.161287000	-2.433071000	1.136753000
H	-4.554320000	-3.240035000	1.744273000
C	-4.990213000	-1.847386000	0.161272000
C	-4.530781000	-0.817096000	-0.636885000
H	-5.198042000	-0.391319000	-1.379084000
C	-2.805002000	0.738768000	-1.346496000
H	-3.550499000	1.097982000	-2.065090000
C	-0.182552000	2.957684000	-2.062106000
H	-2.173946000	2.766756000	-2.679760000
N	6.391631000	-2.279181000	-0.144852000
O	7.025949000	-1.941917000	0.856205000
O	6.821637000	-3.042592000	-1.011240000
N	-6.358949000	-2.331154000	-0.013595000
O	-6.739651000	-3.248928000	0.712857000
O	-7.056316000	-1.794193000	-0.877169000

Table S14. Optimized parameters of [Fe(Hth5OMe-sal)(th5OMe-sal)] neutral complex in the sextet state (**HS**). Units are in **Å**.

Fe	0.034675000	0.603959000	0.089654000
S	-0.859927000	2.252424000	1.650015000
S	0.888653000	2.470173000	-1.560616000
O	1.177531000	-0.604537000	-0.877388000
O	-1.087010000	-0.876534000	0.713775000
N	1.716774000	0.833052000	1.439966000
N	1.665305000	1.660506000	2.538970000
N	0.497754000	3.204030000	3.742146000
H	1.209939000	3.091200000	4.452337000
H	-0.415508000	3.504481000	4.047675000
C	3.263911000	-0.688450000	0.280536000
C	2.398154000	-1.061009000	-0.786849000
C	2.893655000	-1.961508000	-1.762244000
H	2.229399000	-2.243450000	-2.572880000
C	4.173743000	-2.470049000	-1.684656000
H	4.547218000	-3.162063000	-2.433580000
C	5.030018000	-2.104046000	-0.626533000
C	4.575925000	-1.223631000	0.341367000
H	5.211673000	-0.923410000	1.167298000
C	2.867310000	0.224887000	1.313533000
H	3.607794000	0.451093000	2.083867000
C	0.553110000	2.330466000	2.684813000
N	-1.730094000	0.989888000	-1.236994000
N	-1.686483000	2.043153000	-2.130499000
N	-0.707686000	3.792847000	-3.241470000
H	-1.404316000	3.730443000	-3.973691000
H	0.143697000	4.277121000	-3.488506000
C	-3.240499000	-0.699002000	-0.319143000
C	-2.306435000	-1.284124000	0.595011000
C	-2.770389000	-2.372815000	1.390063000
H	-2.065358000	-2.819042000	2.083547000
C	-4.057293000	-2.840818000	1.285736000
H	-4.400062000	-3.670500000	1.897237000
C	-4.977752000	-2.258514000	0.380677000
C	-4.571658000	-1.204191000	-0.407985000
H	-5.252887000	-0.737084000	-1.111721000
C	-2.890170000	0.390772000	-1.163281000
H	-3.694607000	0.757111000	-1.814278000
C	-0.563715000	2.766778000	-2.346593000
H	-2.572762000	2.367897000	-2.510964000
O	6.278886000	-2.673368000	-0.653072000
C	7.183247000	-2.343518000	0.390502000
H	7.404715000	-1.267868000	0.405819000
H	6.793248000	-2.646785000	1.371396000
H	8.099541000	-2.897043000	0.180253000
O	-6.228784000	-2.821889000	0.377912000
C	-7.198557000	-2.279756000	-0.504775000
H	-7.401125000	-1.223882000	-0.278954000
H	-6.883836000	-2.373044000	-1.553217000
H	-8.107542000	-2.862185000	-0.348752000

Table S15. Optimized parameters of [Fe(Hth5OMe-sal)(th5OMe-sal)] neutral complex in the doublet state (**LS**). Units are in **Å**.

Fe	-0.008226000	-1.038802000	0.045159000
S	0.925793000	-2.617754000	1.430131000
S	-0.888019000	-2.798282000	-1.292853000
O	-0.844384000	0.167314000	-1.153704000
O	0.727292000	0.378603000	1.065304000
N	-1.488533000	-1.025536000	1.317171000
N	-1.490500000	-1.850635000	2.427207000
N	-0.381122000	-3.463539000	3.614679000
H	-1.036030000	-3.278798000	4.363648000
H	0.528372000	-3.804640000	3.888691000
C	-2.753119000	0.751483000	0.215633000
C	-1.918433000	0.892191000	-0.927449000
C	-2.284604000	1.865449000	-1.890661000
H	-1.647031000	1.971417000	-2.762824000
C	-3.403237000	2.658471000	-1.734800000
H	-3.668646000	3.403540000	-2.478984000
C	-4.227305000	2.517575000	-0.602342000
C	-3.902096000	1.572282000	0.355404000
H	-4.518218000	1.437108000	1.237859000
C	-2.500311000	-0.211250000	1.251675000
H	-3.218396000	-0.279470000	2.069323000
C	-0.428885000	-2.595439000	2.550622000
N	1.494000000	-1.056193000	-1.210051000
N	1.459283000	-1.991929000	-2.229632000
N	0.486746000	-3.723422000	-3.398715000
H	1.012557000	-3.484206000	-4.229941000
H	-0.345858000	-4.272290000	-3.562007000
C	2.726117000	0.793535000	-0.251837000
C	1.824404000	1.035538000	0.833826000
C	2.168561000	2.086925000	1.734077000
H	1.489123000	2.271387000	2.559531000
C	3.303075000	2.841732000	1.568030000
H	3.546843000	3.641428000	2.261356000
C	4.188435000	2.600281000	0.490482000
C	3.900466000	1.591045000	-0.401105000
H	4.559569000	1.378639000	-1.236475000
C	2.504526000	-0.232473000	-1.212173000
H	3.250576000	-0.334229000	-2.006959000
C	0.408931000	-2.826027000	-2.369089000
H	2.290450000	-2.093376000	-2.805839000
O	-5.315376000	3.356767000	-0.549610000
C	-6.176832000	3.255686000	0.573044000
H	-6.632465000	2.258485000	0.643197000
H	-5.646315000	3.476552000	1.509287000
H	-6.960557000	3.999181000	0.419462000
O	5.286097000	3.423289000	0.438648000
C	6.209399000	3.229744000	-0.620649000
H	6.654593000	2.225777000	-0.587314000
H	5.735474000	3.383891000	-1.599832000
H	6.992762000	3.975531000	-0.478616000

Table S16. Optimized parameters of [Fe(Hthsal)(thsal)] neutral complex in the sextet state (**HS**). Units are in **Å**.

Fe	-0.034772000	-0.263771000	0.083801000
S	0.890404000	-1.859764000	1.668224000
S	-0.882322000	-2.170196000	-1.515765000
O	-1.196710000	0.910470000	-0.907738000
O	1.073027000	1.243603000	0.659997000
N	-1.702651000	-0.482673000	1.447350000
N	-1.645740000	-1.309960000	2.547422000
N	-0.462391000	-2.844462000	3.749604000
H	-1.179188000	-2.746412000	4.457398000
H	0.454287000	-3.130345000	4.058967000
C	-3.250388000	1.048093000	0.299464000
C	-2.403694000	1.394655000	-0.798882000
C	-2.901192000	2.287099000	-1.777171000
H	-2.247618000	2.541806000	-2.605479000
C	-4.175706000	2.819503000	-1.672700000
H	-4.531880000	3.505936000	-2.436514000
C	-5.010871000	2.484517000	-0.593370000
C	-4.544675000	1.609255000	0.372413000
H	-5.179466000	1.339128000	1.213180000
C	-2.847250000	0.137660000	1.333031000
H	-3.580183000	-0.074276000	2.114407000
C	-0.527622000	-1.966239000	2.696855000
N	1.729023000	-0.675421000	-1.242885000
N	1.693894000	-1.763284000	-2.094099000
N	0.718433000	-3.554791000	-3.139365000
H	1.427714000	-3.533326000	-3.861435000
H	-0.128835000	-4.054762000	-3.368082000
C	3.210905000	1.077095000	-0.403028000
C	2.278465000	1.679741000	0.510000000
C	2.725777000	2.804229000	1.257488000
H	2.018836000	3.256658000	1.945167000
C	4.006335000	3.295519000	1.111399000
H	4.315927000	4.156254000	1.698621000
C	4.921730000	2.702233000	0.215449000
C	4.519331000	1.611668000	-0.523925000
H	5.211275000	1.141569000	-1.219903000
C	2.874350000	-0.050624000	-1.205583000
H	3.680522000	-0.420039000	-1.852446000
C	0.572712000	-2.496927000	-2.285735000
H	2.581943000	-2.094165000	-2.464731000
H	-6.007960000	2.906043000	-0.518195000
H	5.925657000	3.099995000	0.112387000

Table S17. Optimized parameters of [Fe(Hthsal)(thsal)] neutral complex in the doublet state (**LS**). Units are in **Å**.

Fe	-0.003251000	-0.565491000	0.052208000
S	1.077624000	-2.131268000	1.327803000
S	-1.063034000	-2.320643000	-1.146116000
O	-0.964120000	0.643464000	-1.057224000
O	0.861190000	0.860209000	0.959136000
N	-1.318936000	-0.521451000	1.493523000
N	-1.178580000	-1.315702000	2.617949000
N	0.069575000	-2.909791000	3.689180000
H	-0.482142000	-2.705095000	4.512428000
H	1.006282000	-3.248948000	3.850908000
C	-2.716027000	1.225214000	0.507757000
C	-2.013939000	1.360065000	-0.729254000
C	-2.491225000	2.312664000	-1.661793000
H	-1.950037000	2.408691000	-2.598069000
C	-3.600461000	3.096462000	-1.390545000
H	-3.935945000	3.820736000	-2.128517000
C	-4.292685000	2.964258000	-0.176188000
C	-3.846530000	2.036643000	0.749502000
H	-4.371131000	1.919759000	1.695130000
C	-2.335317000	0.287133000	1.528502000
H	-2.951782000	0.239012000	2.426532000
C	-0.115787000	-2.066936000	2.620413000
N	1.333608000	-0.612919000	-1.379825000
N	1.151969000	-1.546258000	-2.386111000
N	0.007825000	-3.263747000	-3.411463000
H	0.447344000	-3.053057000	-4.298283000
H	-0.847101000	-3.799157000	-3.468411000
C	2.702465000	1.216622000	-0.583900000
C	1.936122000	1.493724000	0.600695000
C	2.398865000	2.545219000	1.441523000
H	1.818963000	2.751047000	2.335202000
C	3.527641000	3.273413000	1.132231000
H	3.845166000	4.070920000	1.799044000
C	4.276550000	3.000864000	-0.032553000
C	3.860029000	1.987542000	-0.866618000
H	4.423846000	1.761465000	-1.769488000
C	2.349955000	0.189748000	-1.506435000
H	2.995235000	0.075143000	-2.383433000
C	0.081056000	-2.365885000	-2.384725000
H	1.891843000	-1.646627000	-3.075822000
H	-5.162143000	3.578533000	0.034186000
H	5.163647000	3.580125000	-0.265550000

Table S18. Optimized parameters of [Fe(Hth5Cl-sal)(th5Cl-sal)] neutral complex in the sextet state (**HS**). Units are in **Å**.

Fe	0.034318000	0.673500000	0.081617000
S	-0.925993000	2.303815000	1.601837000
S	0.926122000	2.514921000	-1.549189000
O	1.214465000	-0.543385000	-0.836690000
O	-1.094606000	-0.818420000	0.668305000
N	1.675269000	0.929482000	1.479501000
N	1.597680000	1.797717000	2.543427000
N	0.388889000	3.370694000	3.664624000
H	1.099742000	3.317831000	4.382795000
H	-0.528919000	3.686851000	3.938100000
C	3.236012000	-0.652373000	0.425568000
C	2.412914000	-1.031794000	-0.678634000
C	2.927802000	-1.964330000	-1.609610000
H	2.296090000	-2.247499000	-2.445192000
C	4.192536000	-2.507094000	-1.461176000
H	4.571912000	-3.224227000	-2.181618000
C	4.987884000	-2.127693000	-0.371294000
C	4.523755000	-1.216843000	0.556766000
H	5.149232000	-0.929146000	1.396095000
C	2.818226000	0.299476000	1.416905000
H	3.534227000	0.535769000	2.206537000
C	0.474269000	2.456185000	2.648862000
N	-1.704759000	1.055433000	-1.294765000
N	-1.641657000	2.118511000	-2.173363000
N	-0.621115000	3.868290000	-3.245897000
H	-1.330687000	3.855581000	-3.967519000
H	0.232267000	4.360767000	-3.467400000
C	-3.213349000	-0.660786000	-0.432839000
C	-2.300586000	-1.245776000	0.508900000
C	-2.771068000	-2.347996000	1.276752000
H	-2.082407000	-2.791468000	1.988038000
C	-4.050793000	-2.837477000	1.128918000
H	-4.388574000	-3.678511000	1.725851000
C	-4.931283000	-2.249052000	0.200373000
C	-4.525054000	-1.183169000	-0.565981000
H	-5.210214000	-0.735687000	-1.280031000
C	-2.856150000	0.443369000	-1.261796000
H	-3.648016000	0.801560000	-1.931541000
C	-0.508491000	2.836087000	-2.360827000
H	-2.516716000	2.440473000	-2.581015000
Cl	6.598722000	-2.823247000	-0.190513000
Cl	-6.562901000	-2.891690000	0.029310000

Table S19. Optimized parameters of [Fe(Hth5Cl-sal)(th5Cl-sal)] neutral complex in the doublet state (**LS**). Units are in **Å**.

Fe	-0.005381000	-1.056907000	0.046087000
S	1.035309000	-2.639152000	1.329812000
S	-1.018783000	-2.787301000	-1.215142000
O	-0.924741000	0.176257000	-1.073626000
O	0.821372000	0.356431000	1.009653000
N	-1.371693000	-1.040966000	1.440067000
N	-1.271857000	-1.858013000	2.550142000
N	-0.057739000	-3.467583000	3.633830000
H	-0.647252000	-3.295233000	4.437752000
H	0.866530000	-3.828208000	3.817237000
C	-2.732695000	0.723551000	0.438216000
C	-1.986397000	0.883697000	-0.768658000
C	-2.433096000	1.854738000	-1.697068000
H	-1.862177000	1.973995000	-2.612361000
C	-3.551123000	2.635234000	-1.457847000
H	-3.870953000	3.376212000	-2.183076000
C	-4.271612000	2.463412000	-0.269903000
C	-3.873609000	1.525308000	0.661364000
H	-4.439732000	1.398670000	1.578962000
C	-2.390347000	-0.234700000	1.455357000
H	-3.039147000	-0.301551000	2.328691000
C	-0.205848000	-2.606452000	2.577747000
N	1.382363000	-1.077157000	-1.337252000
N	1.238716000	-1.993729000	-2.364030000
N	0.129084000	-3.692840000	-3.455553000
H	0.623107000	-3.491388000	-4.315056000
H	-0.718425000	-4.233887000	-3.552801000
C	2.717105000	0.738381000	-0.457771000
C	1.907552000	0.994612000	0.700873000
C	2.339794000	2.032369000	1.575241000
H	1.730279000	2.226380000	2.451399000
C	3.476693000	2.769399000	1.327522000
H	3.779283000	3.556973000	2.009972000
C	4.254307000	2.502764000	0.184545000
C	3.884990000	1.510129000	-0.689737000
H	4.489848000	1.309819000	-1.569187000
C	2.401934000	-0.272967000	-1.412917000
H	3.079584000	-0.371759000	-2.266583000
C	0.168216000	-2.813856000	-2.414439000
H	1.998976000	-2.075621000	-3.033770000
Cl	-5.699349000	3.456184000	0.034261000
Cl	5.707304000	3.454122000	-0.116493000

Table S20. Optimized parameters of [Fe(Hth5NO_2-sal)(th5NO_2-sal)] neutral complex in the sextet state (**HS**). Units are in **Å**.

Fe	0.033543000	0.862560000	0.083066000
S	-0.898641000	2.474532000	1.608536000
S	0.892533000	2.645346000	-1.586015000
O	1.185081000	-0.406255000	-0.835279000
O	-1.079794000	-0.639299000	0.713616000
N	1.716481000	1.151531000	1.415579000
N	1.684896000	2.079050000	2.429286000
N	0.506781000	3.697267000	3.519407000
H	1.265475000	3.726168000	4.187984000
H	-0.396158000	4.036454000	3.812971000
C	3.216214000	-0.516477000	0.405346000
C	2.361912000	-0.922786000	-0.673732000
C	2.832718000	-1.911838000	-1.576832000
H	2.175258000	-2.207331000	-2.387413000
C	4.077968000	-2.482873000	-1.425850000
H	4.439025000	-3.241324000	-2.110435000
C	4.900051000	-2.075181000	-0.362209000
C	4.481256000	-1.108850000	0.538291000
H	5.144258000	-0.817377000	1.345625000
C	2.843311000	0.493491000	1.358368000
H	3.584360000	0.750441000	2.117336000
C	0.557796000	2.725448000	2.561693000
N	-1.735142000	1.208835000	-1.267156000
N	-1.693378000	2.270263000	-2.147364000
N	-0.675627000	4.000493000	-3.254959000
H	-1.439618000	4.048813000	-3.916121000
H	0.166949000	4.504898000	-3.489163000
C	-3.196317000	-0.540068000	-0.391084000
C	-2.266034000	-1.101179000	0.558042000
C	-2.704974000	-2.214230000	1.336039000
H	-2.000626000	-2.631731000	2.047212000
C	-3.967119000	-2.737010000	1.193242000
H	-4.297727000	-3.582131000	1.785940000
C	-4.858524000	-2.169344000	0.259347000
C	-4.483469000	-1.092810000	-0.518357000
H	-5.196716000	-0.683539000	-1.226337000
C	-2.869598000	0.571447000	-1.229047000
H	-3.673732000	0.904159000	-1.896345000
C	-0.559845000	2.978670000	-2.367802000
H	-2.572027000	2.568706000	-2.564957000
N	6.223176000	-2.677813000	-0.202964000
O	6.922015000	-2.301005000	0.739613000
O	6.568393000	-3.531007000	-1.022311000
N	-6.202384000	-2.725239000	0.109283000
O	-6.955558000	-2.203889000	-0.716225000
O	-6.507487000	-3.684794000	0.817227000

Table S21. Optimized parameters of [Fe(Hth5NO_2-sal)(th5NO_2-sal)] neutral complex in the doublet state (**LS**). Units are in **Å**.

Fe	-0.003216000	-1.200215000	0.048192000
S	1.047150000	-2.772275000	1.304705000
S	-1.048236000	-2.891353000	-1.207715000
O	-0.919845000	0.072534000	-1.055018000
O	0.838000000	0.221224000	1.011230000
N	-1.357236000	-1.182936000	1.453122000
N	-1.235536000	-1.987739000	2.568779000
N	0.006765000	-3.585618000	3.637457000
H	-0.580663000	-3.437089000	4.447311000
H	0.926292000	-3.967929000	3.797131000
C	-2.747720000	0.558986000	0.447055000
C	-1.994363000	0.746136000	-0.760013000
C	-2.454445000	1.714955000	-1.692442000
H	-1.873657000	1.849310000	-2.598899000
C	-3.588761000	2.462279000	-1.461737000
H	-3.933842000	3.201693000	-2.174727000
C	-4.313745000	2.261441000	-0.276831000
C	-3.901129000	1.327385000	0.660868000
H	-4.485018000	1.199210000	1.565789000
C	-2.385955000	-0.391138000	1.468532000
H	-3.030328000	-0.459250000	2.344670000
C	-0.168569000	-2.734495000	2.583397000
N	1.370590000	-1.215003000	-1.351478000
N	1.196762000	-2.108656000	-2.393245000
N	0.031162000	-3.768304000	-3.487068000
H	0.580910000	-3.636675000	-4.325016000
H	-0.814016000	-4.313182000	-3.578252000
C	2.738417000	0.566974000	-0.451542000
C	1.930777000	0.837784000	0.713699000
C	2.379766000	1.865972000	1.597276000
H	1.768236000	2.064984000	2.470516000
C	3.529431000	2.577286000	1.358370000
H	3.860707000	3.356931000	2.034388000
C	4.299810000	2.292422000	0.212564000
C	3.912356000	1.309530000	-0.674536000
H	4.529014000	1.119679000	-1.546857000
C	2.401149000	-0.428715000	-1.421658000
H	3.073412000	-0.521739000	-2.279652000
C	0.114122000	-2.914280000	-2.436972000
H	1.925290000	-2.157286000	-3.100631000
N	-5.519852000	3.045384000	-0.026685000
O	-5.861924000	3.864714000	-0.882339000
O	-6.133991000	2.847731000	1.024254000
N	5.525328000	3.044819000	-0.044872000
O	6.172924000	2.769157000	-1.057883000
O	5.845078000	3.914169000	0.766151000

Table S22. Optimized parameters of [Fe(Hse5OMe-sal)(se5OMe-sal)] neutral complex in the sextet state (**HS**). Units are in **Å**.

Fe	0.020581000	-0.387728000	-0.122405000
Se	-0.829050000	-2.231686000	-1.663262000
Se	0.985046000	-2.125237000	1.755099000
O	1.050655000	0.994926000	0.747097000
O	-1.152809000	0.984389000	-0.904735000
N	1.816137000	-0.704989000	-1.331778000
N	1.929985000	-1.695702000	-2.285918000
N	1.003889000	-3.455379000	-3.397871000
H	1.821482000	-3.436968000	-3.994975000
H	0.158944000	-3.851355000	-3.780394000
C	3.186896000	1.073535000	-0.312111000
C	2.239271000	1.523435000	0.650658000
C	2.620920000	2.580591000	1.515066000
H	1.896125000	2.919814000	2.248141000
C	3.866763000	3.165256000	1.425950000
H	4.151962000	3.977489000	2.088020000
C	4.803614000	2.723205000	0.469239000
C	4.463740000	1.688885000	-0.385562000
H	5.163420000	1.325337000	-1.130415000
C	2.915435000	-0.003510000	-1.218637000
H	3.724232000	-0.289084000	-1.894640000
C	0.879113000	-2.429493000	-2.498452000
N	-1.757899000	-0.699589000	1.226657000
N	-1.759464000	-1.719924000	2.163610000
N	-0.857095000	-3.392047000	3.441259000
H	-1.633548000	-3.341966000	4.089577000
H	-0.031427000	-3.872104000	3.768662000
C	-3.236546000	1.003783000	0.270211000
C	-2.334935000	1.469145000	-0.741730000
C	-2.798459000	2.515805000	-1.594104000
H	-2.120123000	2.867856000	-2.364134000
C	-4.049691000	3.059793000	-1.446000000
H	-4.391482000	3.858021000	-2.098602000
C	-4.936574000	2.598999000	-0.440995000
C	-4.533915000	1.586012000	0.400132000
H	-5.190762000	1.209705000	1.177627000
C	-2.893057000	-0.047487000	1.160652000
H	-3.685446000	-0.340449000	1.861687000
C	-0.661686000	-2.417896000	2.508239000
H	-2.666441000	-2.000633000	2.531928000
O	6.008550000	3.381551000	0.474568000
C	6.989330000	2.980955000	-0.470353000
H	7.283041000	1.932542000	-0.325477000
H	6.634748000	3.118292000	-1.500755000
H	7.853443000	3.624205000	-0.297882000
O	-6.154189000	3.230567000	-0.405723000
C	-7.089549000	2.812732000	0.575991000
H	-7.361716000	1.756175000	0.447046000
H	-6.700619000	2.966060000	1.591965000
H	-7.975323000	3.432843000	0.432145000

Table S23. Optimized parameters of [Fe(Hse5OMe-sal)(se5OMe-sal)] neutral complex in the doublet state (**LS**). Units are in **Å**.

Fe	-0.011621000	-0.708755000	0.035028000
Se	1.031794000	-2.378212000	1.444798000
Se	-1.017681000	-2.528067000	-1.334869000
O	-0.865815000	0.527347000	-1.133064000
O	0.763897000	0.701943000	1.048447000
N	-1.464584000	-0.683633000	1.355592000
N	-1.487679000	-1.503312000	2.472721000
N	-0.468704000	-3.134723000	3.710302000
H	-1.137989000	-2.925959000	4.440839000
H	0.417802000	-3.512981000	4.008590000
C	-2.731489000	1.120274000	0.285371000
C	-1.925813000	1.259273000	-0.878487000
C	-2.308704000	2.241966000	-1.826820000
H	-1.693887000	2.346802000	-2.715267000
C	-3.414733000	3.044096000	-1.636130000
H	-3.693077000	3.795896000	-2.368820000
C	-4.209894000	2.905490000	-0.482154000
C	-3.868826000	1.952140000	0.460878000
H	-4.461972000	1.818211000	1.359142000
C	-2.463693000	0.150380000	1.308914000
H	-3.165211000	0.090763000	2.141396000
C	-0.463959000	-2.281991000	2.635144000
N	1.453653000	-0.712132000	-1.278058000
N	1.416575000	-1.615984000	-2.329197000
N	0.489942000	-3.308592000	-3.582135000
H	1.043605000	-3.047503000	-4.388629000
H	-0.322495000	-3.875183000	-3.779600000
C	2.714349000	1.132639000	-0.327295000
C	1.851080000	1.361958000	0.791981000
C	2.227667000	2.401529000	1.693848000
H	1.577692000	2.575633000	2.544851000
C	3.356617000	3.156812000	1.497940000
H	3.625946000	3.947255000	2.192461000
C	4.203783000	2.928891000	0.386429000
C	3.884127000	1.932062000	-0.507479000
H	4.513451000	1.730403000	-1.368135000
C	2.458680000	0.122026000	-1.292955000
H	3.178648000	0.040237000	-2.114121000
C	0.392941000	-2.465577000	-2.514183000
H	2.232311000	-1.646650000	-2.936430000
O	-5.287467000	3.755808000	-0.396626000
C	-6.119365000	3.657587000	0.748210000
H	-6.583127000	2.664643000	0.825682000
H	-5.561589000	3.868069000	1.670944000
H	-6.899322000	4.409745000	0.619633000
O	5.299648000	3.752302000	0.307688000
C	6.185777000	3.572000000	-0.785068000
H	6.630516000	2.567272000	-0.780205000
H	5.678707000	3.739368000	-1.745320000
H	6.974540000	4.315185000	-0.660299000

Table S24. Optimized parameters of [Fe(Hsesal)(sesal)] neutral complex in the sextet state (**HS**). Units are in **Å**.

Fe	-0.012959000	-0.027137000	0.118417000
Se	0.830651000	-1.860020000	1.664457000
Se	-1.070402000	-1.771863000	-1.698142000
O	-1.029638000	1.364236000	-0.760446000
O	1.211837000	1.328656000	0.842451000
N	-1.778597000	-0.274495000	1.377158000
N	-1.903436000	-1.263780000	2.332454000
N	-1.003438000	-3.052967000	3.421451000
H	-1.810868000	-3.019318000	4.031626000
H	-0.161924000	-3.469326000	3.789666000
C	-3.099866000	1.567456000	0.405207000
C	-2.177635000	1.962653000	-0.613236000
C	-2.539607000	3.029558000	-1.470299000
H	-1.831788000	3.318702000	-2.240676000
C	-3.751466000	3.682481000	-1.321234000
H	-4.001877000	4.502073000	-1.989929000
C	-4.659548000	3.300616000	-0.318184000
C	-4.328870000	2.254844000	0.525313000
H	-5.021668000	1.944561000	1.304195000
C	-2.845133000	0.479477000	1.304418000
H	-3.641194000	0.235878000	2.011130000
C	-0.869987000	-2.025160000	2.525739000
N	1.720332000	-0.413334000	-1.275848000
N	1.673312000	-1.457055000	-2.185367000
N	0.689632000	-3.132325000	-3.396853000
H	1.460615000	-3.137290000	-4.053263000
H	-0.154445000	-3.602917000	-3.688758000
C	3.245887000	1.297698000	-0.413477000
C	2.388679000	1.803981000	0.624640000
C	2.889403000	2.867278000	1.428381000
H	2.241122000	3.244717000	2.212335000
C	4.145038000	3.393881000	1.213612000
H	4.495547000	4.207234000	1.843858000
C	4.985602000	2.896967000	0.192549000
C	4.533963000	1.865180000	-0.599239000
H	5.168682000	1.467670000	-1.388765000
C	2.861017000	0.226457000	-1.266369000
H	3.627066000	-0.089542000	-1.986274000
C	0.547530000	-2.132246000	-2.484589000
H	2.562438000	-1.764783000	-2.574760000
H	-5.607116000	3.817804000	-0.208343000
H	5.971614000	3.321928000	0.038036000

Table S25. Optimized parameters of [Fe(Hsesal)(sesal)] neutral complex in the doublet state (**LS**). Units are in **Å**.

Fe	-0.032476000	-0.254996000	0.036560000
Se	1.049710000	-2.072906000	1.199744000
Se	-1.538915000	-1.883746000	-1.082993000
O	-0.934911000	1.116589000	-0.938702000
O	1.115920000	1.022521000	0.861694000
N	-1.164169000	-0.070422000	1.630922000
N	-1.046957000	-0.891886000	2.741432000
N	-0.000693000	-2.657944000	3.752020000
H	-0.464493000	-2.377181000	4.607181000
H	0.874481000	-3.148864000	3.856883000
C	-2.401471000	1.884861000	0.821720000
C	-1.832120000	1.957750000	-0.486720000
C	-2.277391000	2.992382000	-1.346188000
H	-1.838586000	3.040277000	-2.338063000
C	-3.229157000	3.910936000	-0.937400000
H	-3.543595000	4.694518000	-1.622211000
C	-3.790295000	3.840728000	0.348489000
C	-3.374506000	2.835783000	1.203993000
H	-3.798924000	2.764698000	2.202958000
C	-2.044394000	0.876178000	1.779876000
H	-2.561311000	0.893189000	2.739654000
C	-0.122779000	-1.798283000	2.689820000
N	1.113528000	-0.418695000	-1.556949000
N	0.725291000	-1.268297000	-2.582422000
N	-0.676242000	-2.776343000	-3.608824000
H	-0.255818000	-2.589463000	-4.510405000
H	-1.582149000	-3.221877000	-3.631708000
C	2.790716000	1.201411000	-0.881894000
C	2.213110000	1.529083000	0.393454000
C	2.906031000	2.480770000	1.195044000
H	2.468481000	2.724067000	2.157663000
C	4.074952000	3.070727000	0.765543000
H	4.571270000	3.795580000	1.405697000
C	4.638098000	2.750177000	-0.489020000
C	3.997388000	1.830416000	-1.287853000
H	4.415658000	1.569426000	-2.257994000
C	2.201464000	0.265055000	-1.776963000
H	2.721135000	0.112664000	-2.728870000
C	-0.420950000	-1.967511000	-2.543306000
H	1.373306000	-1.374154000	-3.359614000
H	-4.536717000	4.561550000	0.665898000
H	5.559043000	3.221076000	-0.815937000

Table S26. Optimized parameters of [Fe(Hse5Cl-sal)(se5Cl-sal)] neutral complex in the sextet state (**HS**). Units are in **Å**.

Fe	0.020776000	-0.462618000	-0.126126000
Se	-0.854441000	-2.285761000	-1.655751000
Se	1.036837000	-2.184641000	1.717825000
O	1.055707000	0.942373000	0.711886000
O	-1.181898000	0.902460000	-0.882214000
N	1.801365000	-0.784701000	-1.356573000
N	1.917436000	-1.813226000	-2.267721000
N	0.982480000	-3.610087000	-3.310625000
H	1.812159000	-3.643203000	-3.889825000
H	0.140662000	-4.030598000	-3.672586000
C	3.143559000	1.064523000	-0.433313000
C	2.215892000	1.512188000	0.556688000
C	2.588596000	2.603515000	1.377992000
H	1.880026000	2.937070000	2.129052000
C	3.811365000	3.233465000	1.227718000
H	4.079938000	4.072081000	1.861728000
C	4.709723000	2.785728000	0.248649000
C	4.387871000	1.720123000	-0.567153000
H	5.092274000	1.378994000	-1.319474000
C	2.882561000	-0.051744000	-1.297741000
H	3.683416000	-0.337285000	-1.982595000
C	0.862231000	-2.546703000	-2.460282000
N	-1.723257000	-0.774577000	1.280540000
N	-1.698066000	-1.801437000	2.208728000
N	-0.743086000	-3.472068000	3.449274000
H	-1.528131000	-3.475319000	4.088385000
H	0.087162000	-3.965062000	3.743348000
C	-3.202687000	0.961658000	0.392796000
C	-2.340230000	1.416683000	-0.662687000
C	-2.818355000	2.476889000	-1.486008000
H	-2.170035000	2.819275000	-2.285544000
C	-4.052485000	3.051422000	-1.278083000
H	-4.395318000	3.860177000	-1.915439000
C	-4.882787000	2.592057000	-0.235484000
C	-4.471828000	1.568407000	0.582396000
H	-5.119057000	1.218390000	1.381268000
C	-2.845133000	-0.103729000	1.267117000
H	-3.614791000	-0.382611000	1.997779000
C	-0.586048000	-2.496103000	2.517584000
H	-2.590864000	-2.072112000	2.616834000
Cl	-6.458265000	3.342912000	0.007451000
Cl	6.266432000	3.595067000	0.066193000

Table S27. Optimized parameters of [Fe(Hse5Cl-sal)(se5Cl-sal)] neutral complex in the doublet state (**LS**). Units are in **Å**.

Fe	-0.008639000	-0.725353000	0.036442000
Se	1.147663000	-2.399735000	1.329907000
Se	-1.157021000	-2.514266000	-1.239830000
O	-0.947696000	0.538857000	-1.046259000
O	0.863147000	0.681282000	0.986034000
N	-1.337022000	-0.697617000	1.482690000
N	-1.252599000	-1.509413000	2.601220000
N	-0.119022000	-3.136805000	3.738863000
H	-0.721033000	-2.939029000	4.528308000
H	0.784590000	-3.534809000	3.944746000
C	-2.701803000	1.094995000	0.519203000
C	-1.991611000	1.253590000	-0.709436000
C	-2.458383000	2.234000000	-1.619474000
H	-1.915643000	2.352088000	-2.551872000
C	-3.559930000	3.023715000	-1.340894000
H	-3.895349000	3.771444000	-2.052101000
C	-4.244816000	2.853830000	-0.130944000
C	-3.827422000	1.907632000	0.782473000
H	-4.365363000	1.782157000	1.717051000
C	-2.341110000	0.128881000	1.520651000
H	-2.968022000	0.070826000	2.410378000
C	-0.220500000	-2.291679000	2.666609000
N	1.331039000	-0.731086000	-1.407150000
N	1.176022000	-1.613298000	-2.465720000
N	0.098360000	-3.272117000	-3.639631000
H	0.622121000	-3.054385000	-4.477587000
H	-0.730942000	-3.833620000	-3.767376000
C	2.700665000	1.076760000	-0.543251000
C	1.937445000	1.320826000	0.649019000
C	2.408057000	2.344862000	1.520809000
H	1.834059000	2.528761000	2.422734000
C	3.537341000	3.080441000	1.238757000
H	3.870115000	3.857415000	1.919351000
C	4.269038000	2.826836000	0.062123000
C	3.862307000	1.848094000	-0.810387000
H	4.431771000	1.657979000	-1.715402000
C	2.345075000	0.082191000	-1.499377000
H	2.990581000	0.003443000	-2.380024000
C	0.127417000	-2.449047000	-2.558508000
H	1.915544000	-1.625324000	-3.164398000
Cl	-5.651900000	3.860153000	0.221635000
Cl	5.712998000	3.777725000	-0.281534000

Table S28. Optimized parameters of [Fe(Hse5NO_2-sal)(se5NO_2-sal)] neutral complex in the sextet state (**HS**). Units are in **Å**.

Fe	0.028911000	-0.650227000	-0.123135000
Se	-0.833159000	-2.412913000	-1.700336000
Se	0.989317000	-2.371984000	1.716633000
O	1.051499000	0.766626000	0.749351000
O	-1.140770000	0.755429000	-0.887607000
N	1.832047000	-0.968280000	-1.307212000
N	1.972789000	-2.014368000	-2.192996000
N	1.046433000	-3.813396000	-3.241422000
H	1.907043000	-3.897248000	-3.767638000
H	0.216358000	-4.243938000	-3.617697000
C	3.133858000	0.915994000	-0.396808000
C	2.190077000	1.359077000	0.589940000
C	2.534631000	2.470456000	1.405374000
H	1.812702000	2.792168000	2.148308000
C	3.738846000	3.122217000	1.254106000
H	4.002193000	3.973661000	1.870595000
C	4.649051000	2.676655000	0.280273000
C	4.356644000	1.591835000	-0.530083000
H	5.085298000	1.272926000	-1.267479000
C	2.900928000	-0.218879000	-1.247712000
H	3.715210000	-0.500765000	-1.917535000
C	0.914606000	-2.732937000	-2.420969000
N	-1.750114000	-0.942672000	1.250801000
N	-1.752434000	-1.977551000	2.168854000
N	-0.817353000	-3.652040000	3.419525000
H	-1.646933000	-3.719059000	3.994913000
H	-0.005076000	-4.176524000	3.708463000
C	-3.173760000	0.838020000	0.360425000
C	-2.282935000	1.294440000	-0.679341000
C	-2.720581000	2.382482000	-1.494937000
H	-2.047592000	2.719526000	-2.275752000
C	-3.941072000	2.979970000	-1.299813000
H	-4.269536000	3.806829000	-1.918906000
C	-4.793994000	2.514779000	-0.276014000
C	-4.421356000	1.464761000	0.537275000
H	-5.105198000	1.133276000	1.311874000
C	-2.854078000	-0.248881000	1.229975000
H	-3.642428000	-0.514688000	1.944667000
C	-0.648965000	-2.680993000	2.492701000
H	-2.648673000	-2.216671000	2.589152000
N	-6.094848000	3.149189000	-0.072266000
O	-6.398580000	4.083871000	-0.813496000
O	-6.816353000	2.714803000	0.828689000
N	5.929034000	3.365132000	0.122051000
O	6.707712000	2.950945000	-0.739004000
O	6.160904000	4.324180000	0.860407000

Table S29. Optimized parameters of [Fe(Hse5NO_2-sal)(se5NO_2-sal)] neutral complex in the doublet state (**LS**). Units are in **Å**.

Fe	-0.005089000	-0.871431000	0.038901000
Se	1.153167000	-2.533071000	1.314658000
Se	-1.167935000	-2.624829000	-1.240328000
O	-0.935684000	0.429172000	-1.035529000
O	0.869577000	0.546638000	0.995375000
N	-1.333451000	-0.845525000	1.484536000
N	-1.235376000	-1.645367000	2.609313000
N	-0.078024000	-3.256618000	3.745536000
H	-0.686084000	-3.082209000	4.535543000
H	0.819857000	-3.673470000	3.936855000
C	-2.722741000	0.923348000	0.506397000
C	-1.995372000	1.110467000	-0.716606000
C	-2.468578000	2.089500000	-1.633164000
H	-1.908096000	2.223563000	-2.552339000
C	-3.589615000	2.845415000	-1.371306000
H	-3.944673000	3.592206000	-2.071655000
C	-4.289008000	2.644998000	-0.170119000
C	-3.863721000	1.701756000	0.751938000
H	-4.427310000	1.574115000	1.669743000
C	-2.349507000	-0.034999000	1.514370000
H	-2.979479000	-0.095643000	2.401392000
C	-0.199519000	-2.422619000	2.672235000
N	1.333054000	-0.867189000	-1.408526000
N	1.160641000	-1.727008000	-2.482827000
N	0.041993000	-3.349058000	-3.668979000
H	0.631852000	-3.206797000	-4.477860000
H	-0.778055000	-3.923802000	-3.794351000
C	2.722410000	0.912683000	-0.514926000
C	1.950426000	1.169365000	0.677481000
C	2.426875000	2.186527000	1.560454000
H	1.842612000	2.374298000	2.454599000
C	3.568679000	2.900365000	1.295241000
H	3.921209000	3.671445000	1.970373000
C	4.303869000	2.630268000	0.122066000
C	3.889355000	1.658741000	-0.764839000
H	4.478941000	1.480514000	-1.658122000
C	2.356171000	-0.068286000	-1.487231000
H	3.004252000	-0.139497000	-2.366058000
C	0.102507000	-2.551732000	-2.578306000
H	1.876254000	-1.706494000	-3.205912000
N	-5.480830000	3.438877000	0.112460000
O	-6.072636000	3.240999000	1.176254000
O	-5.834563000	4.266734000	-0.730306000
N	5.520526000	3.386167000	-0.163326000
O	5.864971000	4.245626000	0.648313000
O	6.137091000	3.123685000	-1.199139000

Table S30. Optimized parameters of $[Fe(semsal)_2]^-$ anionic complex in the sextet state (**HS**). Units are in **Å**.

Fe	0.000095000	0.462537000	0.000080000
O	0.651234000	1.910081000	-1.324386000
O	-0.652099000	1.907657000	1.326375000
O	-1.191156000	-0.877666000	0.774602000
O	1.192461000	-0.875231000	-0.777204000
N	-1.616159000	0.747915000	-1.438791000
N	-1.379524000	1.666017000	-2.430620000
N	0.193131000	3.196632000	-3.152021000
H	-0.317833000	3.180839000	-4.024652000
H	1.193657000	3.313758000	-3.235638000
C	-3.225791000	-0.828902000	-0.504058000
C	-2.406766000	-1.299315000	0.576295000
C	-2.960570000	-2.272017000	1.448286000
H	-2.331139000	-2.621703000	2.261458000
C	-4.246719000	-2.759616000	1.272166000
H	-4.635305000	-3.506046000	1.961966000
C	-5.046740000	-2.301144000	0.214002000
C	-4.527248000	-1.349137000	-0.652831000
H	-5.134107000	-0.982217000	-1.479050000
C	-2.773346000	0.161656000	-1.451143000
H	-3.466999000	0.449665000	-2.247149000
C	-0.175955000	2.203390000	-2.253104000
N	1.615500000	0.745848000	1.439987000
N	1.377991000	1.662013000	2.433482000
N	-0.195570000	3.190730000	3.156829000
H	0.314840000	3.173401000	4.029757000
H	-1.196184000	3.307404000	3.240092000
C	3.226133000	-0.828648000	0.503181000
C	2.407894000	-1.297192000	-0.578594000
C	2.962444000	-2.268332000	-1.451888000
H	2.333614000	-2.616666000	-2.266105000
C	4.248528000	-2.756066000	-1.275768000
H	4.637658000	-3.501248000	-1.966612000
C	5.047802000	-2.299333000	-0.216278000
C	4.527580000	-1.348944000	0.651879000
H	5.133825000	-0.983397000	1.479159000
C	2.772917000	0.160075000	1.451830000
H	3.466083000	0.446872000	2.248691000
C	0.174386000	2.199403000	2.256220000
H	-6.053914000	-2.683217000	0.074319000
H	6.054947000	-2.681486000	-0.076618000

Table S31. Optimized parameters of $[Fe(semsal)_2]^-$ anionic complex in the doublet state (**LS**). Units are in **Å**.

Fe	-0.000056000	0.697337000	-0.000095000
O	0.792641000	2.123320000	-1.078983000
O	-0.793062000	2.122708000	1.079342000
O	-0.935861000	-0.592548000	1.015260000
O	0.936906000	-0.591057000	-1.015945000
N	-1.325056000	0.778759000	-1.408018000
N	-1.082238000	1.713287000	-2.389970000
N	0.453597000	3.346654000	-2.974789000
H	0.067900000	3.264973000	-3.906012000
H	1.448453000	3.524518000	-2.939640000
C	-2.742516000	-0.995801000	-0.567269000
C	-2.005532000	-1.246620000	0.638916000
C	-2.488870000	-2.268179000	1.498674000
H	-1.923506000	-2.449654000	2.408149000
C	-3.622508000	-3.005952000	1.198257000
H	-3.953233000	-3.782497000	1.884995000
C	-4.342391000	-2.759053000	0.018637000
C	-3.894216000	-1.763774000	-0.837707000
H	-4.440793000	-1.556330000	-1.756317000
C	-2.371032000	0.025564000	-1.517217000
H	-3.009288000	0.180636000	-2.389396000
C	0.049816000	2.341089000	-2.104864000
N	1.324950000	0.778748000	1.408265000
N	1.081595000	1.712657000	2.390538000
N	-0.455455000	3.344684000	2.976330000
H	-0.070831000	3.261728000	3.907898000
H	-1.450397000	3.522031000	2.940496000
C	2.743117000	-0.994968000	0.566857000
C	2.006270000	-1.245664000	-0.639391000
C	2.489383000	-2.267274000	-1.499105000
H	1.924020000	-2.448716000	-2.408581000
C	3.623017000	-3.005130000	-1.198688000
H	3.953653000	-3.781722000	-1.885410000
C	4.342925000	-2.758230000	-0.019123000
C	3.894791000	-1.762941000	0.837278000
H	4.441384000	-1.555592000	1.755899000
C	2.371380000	0.026138000	1.517007000
H	3.009678000	0.181217000	2.389165000
C	-0.050777000	2.339949000	2.105708000
H	-5.231626000	-3.335002000	-0.221227000
H	5.232156000	-3.334187000	0.220748000

Table S32. Optimized parameters of $[Fe(thsal)_2]^-$ anionic complex in the sextet state (**HS**). Units are in **Å**.

Fe	0.000000000	0.299869000	0.000000000
S	0.944823000	1.978958000	-1.575721000
S	-0.944824000	1.978954000	1.575724000
O	-1.150162000	-1.117258000	0.743308000
O	1.150164000	-1.117255000	-0.743311000
N	-1.678307000	0.607812000	-1.416280000
N	-1.611922000	1.517413000	-2.449051000
N	-0.432570000	3.119564000	-3.561958000
H	-1.139443000	3.003212000	-4.278655000
H	0.493880000	3.346418000	-3.894634000
C	-3.240825000	-1.007795000	-0.410068000
C	-2.370131000	-1.524918000	0.605566000
C	-2.887491000	-2.529056000	1.469261000
H	-2.220807000	-2.914738000	2.234756000
C	-4.184491000	-2.994811000	1.340600000
H	-4.546212000	-3.763554000	2.020505000
C	-5.037525000	-2.488059000	0.343834000
C	-4.556443000	-1.509599000	-0.511592000
H	-5.201386000	-1.105779000	-1.290178000
C	-2.829958000	0.000859000	-1.348516000
H	-3.569042000	0.298176000	-2.097883000
C	-0.479014000	2.162611000	-2.557515000
N	1.678306000	0.607810000	1.416281000
N	1.611919000	1.517409000	2.449055000
N	0.432567000	3.119560000	3.561962000
H	1.139441000	3.003207000	4.278659000
H	-0.493882000	3.346415000	3.894637000
C	3.240826000	-1.007794000	0.410067000
C	2.370133000	-1.524915000	-0.605569000
C	2.887494000	-2.529052000	-1.469265000
H	2.220810000	-2.914732000	-2.234762000
C	4.184493000	-2.994807000	-1.340604000
H	4.546215000	-3.763548000	-2.020510000
C	5.037527000	-2.488056000	-0.343837000
C	4.556444000	-1.509598000	0.511591000
H	5.201386000	-1.105780000	1.290178000
C	2.829957000	0.000858000	1.348516000
H	3.569041000	0.298174000	2.097885000
C	0.479011000	2.162604000	2.557520000
H	-6.054541000	-2.856189000	0.246071000
H	6.054542000	-2.856186000	-0.246073000

Table S33. Optimized parameters of $[Fe(thsal)_2]^-$ anionic complex in the doublet state (**LS**). Units are in **Å**.

Fe	0.000000000	0.568371000	0.000000000
S	1.131449000	2.194754000	-1.186656000
S	-1.131450000	2.194754000	1.186656000
O	-0.958545000	-0.796098000	0.967287000
O	0.958545000	-0.796097000	-0.967288000
N	-1.253711000	0.577164000	-1.512155000
N	-1.048546000	1.395387000	-2.610013000
N	0.243300000	3.018539000	-3.584762000
H	-0.236502000	2.757123000	-4.437999000
H	1.210163000	3.285026000	-3.710605000
C	-2.729617000	-1.167053000	-0.641816000
C	-2.038118000	-1.413028000	0.592129000
C	-2.585349000	-2.401598000	1.458290000
H	-2.056814000	-2.582530000	2.389634000
C	-3.730858000	-3.105842000	1.133478000
H	-4.111330000	-3.855333000	1.824759000
C	-4.405732000	-2.863144000	-0.076108000
C	-3.897157000	-1.904431000	-0.937403000
H	-4.403694000	-1.701964000	-1.879731000
C	-2.288638000	-0.200888000	-1.612079000
H	-2.873437000	-0.106411000	-2.528988000
C	0.013913000	2.146955000	-2.527328000
N	1.253711000	0.577164000	1.512155000
N	1.048546000	1.395386000	2.610014000
N	-0.243302000	3.018535000	3.584764000
H	0.236499000	2.757118000	4.438002000
H	-1.210166000	3.285020000	3.710608000
C	2.729617000	-1.167053000	0.641815000
C	2.038118000	-1.413027000	-0.592130000
C	2.585350000	-2.401596000	-1.458291000
H	2.056815000	-2.582528000	-2.389635000
C	3.730860000	-3.105840000	-1.133480000
H	4.111331000	-3.855331000	-1.824761000
C	4.405733000	-2.863143000	0.076107000
C	3.897158000	-1.904430000	0.937402000
H	4.403695000	-1.701963000	1.879730000
C	2.288638000	-0.200888000	1.612079000
H	2.873437000	-0.106411000	2.528988000
C	-0.013913000	2.146954000	2.527328000
H	-5.305368000	-3.415474000	-0.331290000
H	5.305370000	-3.415473000	0.331289000

Table S34. Optimized parameters of $[Fe(sesal)_2]^-$ anionic complex in the sextet state (**HS**). Units are in **Å**.

Fe	0.000000000	0.030815000	0.000000000
Se	-1.104461000	1.782720000	1.585596000
Se	1.104461000	1.782721000	-1.585595000
O	1.176805000	-1.395965000	-0.688866000
O	-1.176804000	-1.395966000	0.688865000
N	1.622377000	0.327386000	1.508720000
N	1.557029000	1.240426000	2.542797000
N	0.432372000	2.871843000	3.665735000
H	1.136154000	2.741360000	4.383707000
H	-0.485181000	3.139406000	3.991724000
C	3.198759000	-1.329897000	0.578933000
C	2.375090000	-1.831964000	-0.482237000
C	2.918386000	-2.850545000	-1.313781000
H	2.287801000	-3.223789000	-2.115112000
C	4.195058000	-3.343185000	-1.110769000
H	4.577944000	-4.121952000	-1.767292000
C	5.002277000	-2.851475000	-0.068364000
C	4.496048000	-1.859728000	0.755646000
H	5.105124000	-1.466774000	1.567918000
C	2.761637000	-0.308905000	1.490384000
H	3.472299000	-0.025060000	2.271536000
C	0.454618000	1.921589000	2.658538000
N	-1.622377000	0.327387000	-1.508720000
N	-1.557029000	1.240428000	-2.542796000
N	-0.432372000	2.871843000	-3.665735000
H	-1.136154000	2.741359000	-4.383707000
H	0.485182000	3.139404000	-3.991724000
C	-3.198759000	-1.329897000	-0.578934000
C	-2.375089000	-1.831965000	0.482237000
C	-2.918385000	-2.850546000	1.313780000
H	-2.287800000	-3.223790000	2.115111000
C	-4.195058000	-3.343186000	1.110768000
H	-4.577943000	-4.121953000	1.767291000
C	-5.002277000	-2.851476000	0.068364000
C	-4.496048000	-1.859728000	-0.755646000
H	-5.105124000	-1.466773000	-1.567918000
C	-2.761637000	-0.308904000	-1.490384000
H	-3.472299000	-0.025060000	-2.271536000
C	-0.454619000	1.921591000	-2.658536000
H	6.003803000	-3.241479000	0.086938000
H	-6.003802000	-3.241479000	-0.086938000

Table S35. Optimized parameters of $[Fe(sesal)_2]^-$ anionic complex in the doublet state (**LS**). Units are in **Å**.

Fe	0.000000000	0.259216000	0.000000000
Se	1.343175000	1.967882000	-1.092895000
Se	-1.343175000	1.967882000	1.092895000
O	-1.056642000	-1.112009000	0.861668000
O	1.056642000	-1.112009000	-0.861668000
N	-1.085945000	0.255550000	-1.651293000
N	-0.798009000	1.053120000	-2.749096000
N	0.512497000	2.670999000	-3.700568000
H	0.089810000	2.378037000	-4.574266000
H	1.476939000	2.962961000	-3.775458000
C	-2.638131000	-1.498350000	-0.925441000
C	-2.084957000	-1.736454000	0.377492000
C	-2.719774000	-2.724428000	1.183559000
H	-2.296311000	-2.898697000	2.168280000
C	-3.819494000	-3.434355000	0.737849000
H	-4.270893000	-4.182776000	1.386309000
C	-4.358896000	-3.199912000	-0.540240000
C	-3.763511000	-2.242655000	-1.344650000
H	-4.164325000	-2.046380000	-2.337791000
C	-2.099075000	-0.536060000	-1.846780000
H	-2.584383000	-0.455024000	-2.821087000
C	0.233042000	1.832214000	-2.631520000
N	1.085945000	0.255550000	1.651293000
N	0.798009000	1.053120000	2.749096000
N	-0.512497000	2.670999000	3.700568000
H	-0.089810000	2.378037000	4.574266000
H	-1.476939000	2.962961000	3.775458000
C	2.638131000	-1.498350000	0.925441000
C	2.084957000	-1.736453000	-0.377492000
C	2.719774000	-2.724428000	-1.183559000
H	2.296312000	-2.898697000	-2.168281000
C	3.819494000	-3.434355000	-0.737849000
H	4.270894000	-4.182776000	-1.386309000
C	4.358896000	-3.199912000	0.540240000
C	3.763511000	-2.242655000	1.344650000
H	4.164325000	-2.046380000	2.337791000
C	2.099075000	-0.536060000	1.846780000
H	2.584383000	-0.455024000	2.821087000
C	-0.233042000	1.832214000	2.631520000
H	-5.222888000	-3.757530000	-0.889551000
H	5.222888000	-3.757530000	0.889551000