APPENDIX

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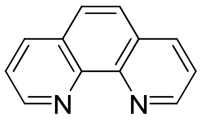
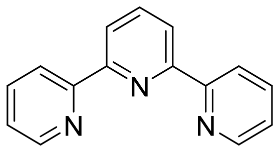
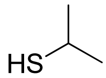
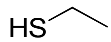
Table A1. Ligands considered in the model compounds

ligands	
	
phen	**terpy**
	
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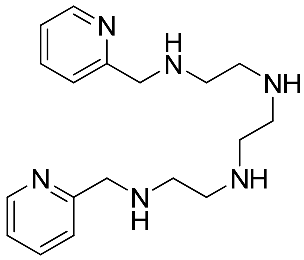
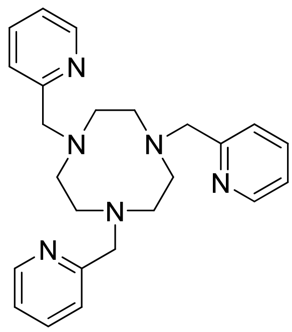
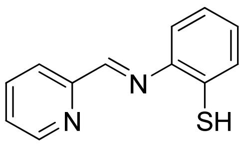
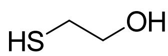
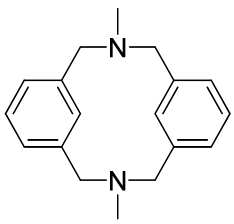
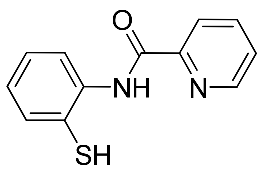
	
bpteta	**Py_3tacn**
	
$PyASH_2$	**HSEtOH**
	
$Py_2(NMe)_2$	**$PypepSH_2$**

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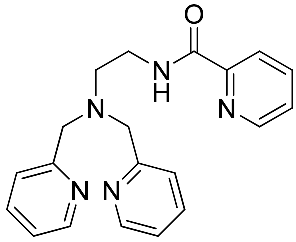
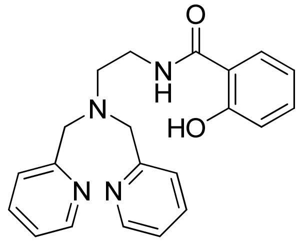
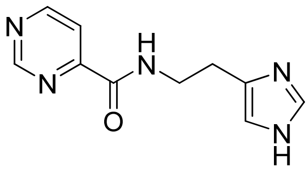
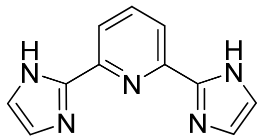
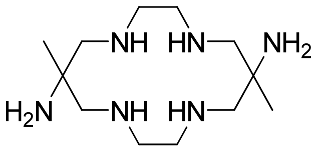
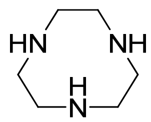
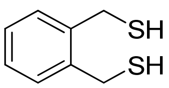
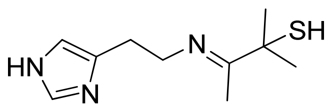
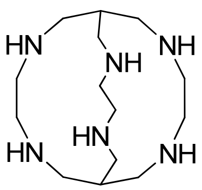
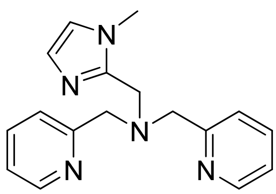
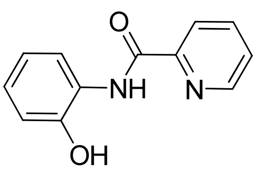
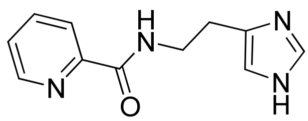
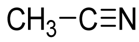
 **PaPy$_3$H**	 **PaPy$_2$OH$_2$**
 **PrpepH**	 **PyIm$_2$H$_2$**
 **diammac**	 **tacn**
 **S$_2$-o-xylH$_2$**	 **DITim**
 **sar**	 **bpia**
 **PypepOH$_2$**	 **PypepH**

Table A1. (*Continued.*)



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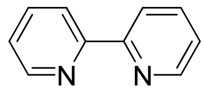

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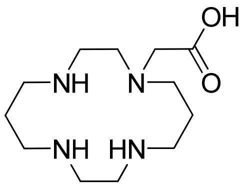

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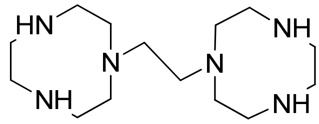
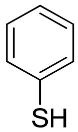
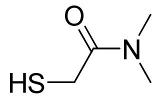


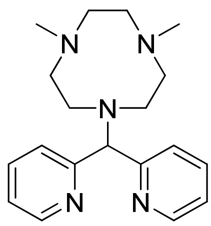
dtne



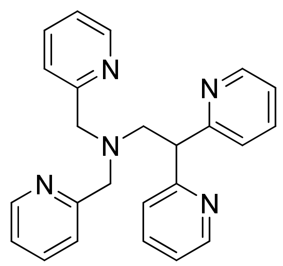
HSPh



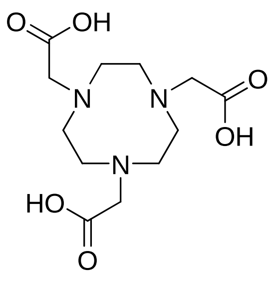
$(CH_3)_2NCOCH_2SH$



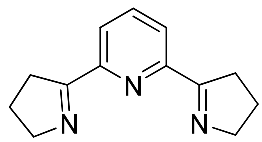
$tacnPy_2$



N_4Py



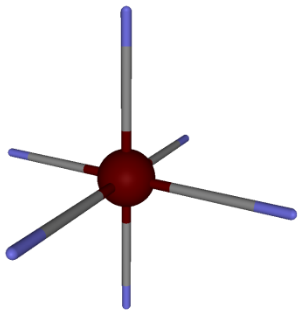
$TCTAH_3$



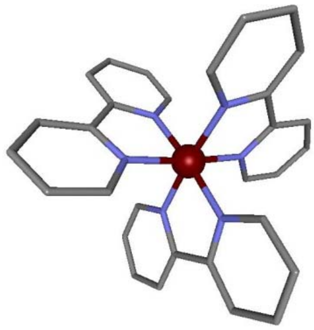
Pyr_2Py

Abbreviation of ligands: **AN** = acetonitrile, **bpy** = 2,2'-bipyridinyl, **phen** = 1,10-phenanthroline, **diammac** = 6,13-dimethyl-1,4,8,11-tetraazacyclotetradecane-6,13-diamine, **sar** = 3,6,10,13,16,19-hexaaza-bicyclo[6.6.6]icosane, **tacn** = 1,4,7-triazonane, **$PyIm_2$** = 2,6-di-(1*H*-imidazol-2-yl)-pyridine-11,16-diide, **PypepO** = *N*-(2-hydroxyphenyl)pyridine-2-carboxamide-7,8-diide, **PypepS** = *N*-(2-mercaptophenyl)pyridine-2-carboxamide-7,8-diide, **PyAS**= 2-[(pyridin-2-ylmethylene)-amino]-benzenethiol-15-ide, **bpteta**=*N*-pyridin-2-ylmethyl-*N'*-(2-{2-[(pyridin-2-ylmethyl)-amino]-ethylamino}-ethyl)-ethane-1,2-diamine, **DITim** = 3-[2-(1*H*-imidazol-4-yl)-ethylimino]-2-methyl-butane-2-thiol-11-ide, **Pypep** = *N*-[2-(1*H*-imidazol-4-yl)ethyl]pyridine-2-carboxamide-8-ide, **Prpep** = pyrimidine-4-carboxylic acid [2-(1*H*-imidazol-4-yl)-ethyl]-amide-8-ide, **$PaPy_3$** = *N*-{2-[bis(pyridin-2-ylmethyl)amino]ethyl}pyridine-2-carboxamide-18-ide, **$PaPy_2O$** = *N*-{2-[bis(pyridin-2-ylmethyl)amino]ethyl}-2-hydroxybenzamide-18,27-diide, **SEt** = ethanethiol-1-ide, **S_2-o-xyl** = 1,2-phenylenedimethanethiol-8,10-diide, **bpia** = (1-methyl-1*H*-imidazol-2-ylmethyl)-bis-pyridin-2-ylmethyl-amine, **$py_2(nMe)_2$** = 3,11-dimethyl-3,11-diaza-tricyclo[11.3.1.$1^{5,9}$]octadeca-1(16),5,7,9(18),13(17),14-hexaene, **SEtOH** = 2-mercapto-ethanole-1-ide, **cyclamAc** = (1,4,8,11tetraaza-Cyclotetradec-1-yl)-acetic acid anion, **dtne** = 1,2-di(1,4,7-triazonan-1-yl)ethane, **terpy** = 2,2':6',2''-terpyridine, **SPh** = benzenethiol-7-ide, **$(CH_3)_2NCOCH_2S$** = *N*,*N*-dimethyl-2-sulfanylacetamide-1-ide, **HS-*i*-Pr** = propane-2-thiol-1-ide, **$tacnPy_2$** = 1-(dipyridin-2-ylmethyl)-4,7-dimethyl-1,4,7-triazonane, **N_4Py** = 2,2-di(pyridin-2-yl)-N,N-bis(pyridin-2-ylmethyl)ethanamine, **Py_3tacn** = 1,4,7-tris(pyridin-2-ylmethyl)-1,4,7-triazonane, **TCTA** = 2,2',2''-(1,4,7-triazonane-1,4,7-triyl)triacetate, **Pyr_2Py** = 2,6-bis(3,4-dihydro-2H-pyrrol-5-yl)pyridine.

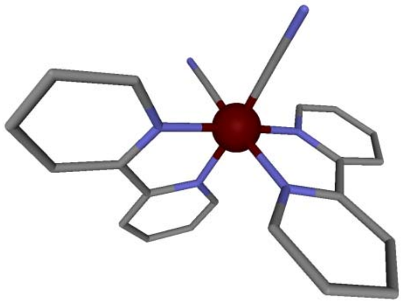
Figure A1. Graphical representation of considered TMCs.



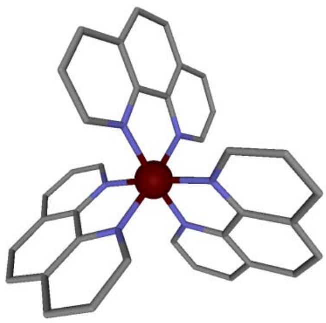
1. $[Fe(CN)_6]^{3-/4-}$



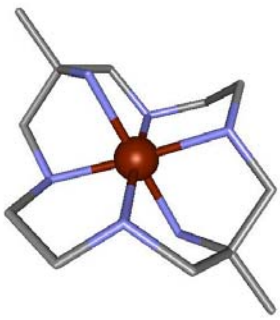
2. $[Fe(bpy)_3]^{3+/2+}$



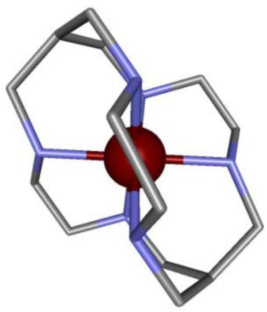
3. $[Fe(bpy)_2(CN)_2]^{1+/0}$



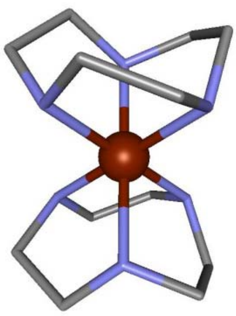
4. $[Fe(phen)_3]^{3+/2+}$



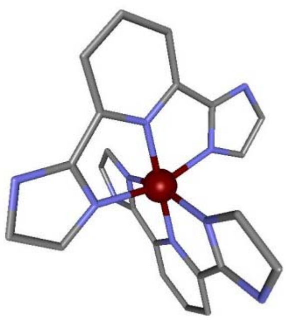
5. $[Fe(diammac)]^{3+/2+}$



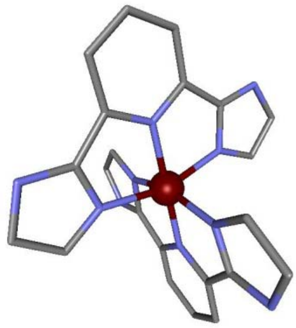
6. $[Fe(sar)]^{3+/2+}$



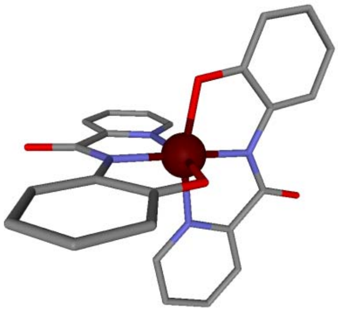
7. $[Fe(tacn)_2]^{3+/2+}$



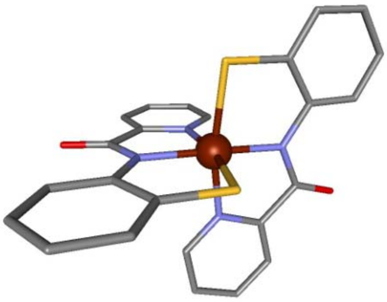
8. $[Fe(PyIm_2H_2)_2]^{3+/2+}$



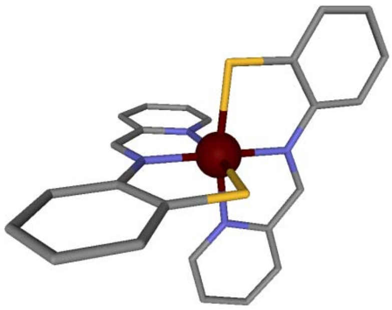
9. $[Fe(PyIm_2)_2]^{1-/2-}$



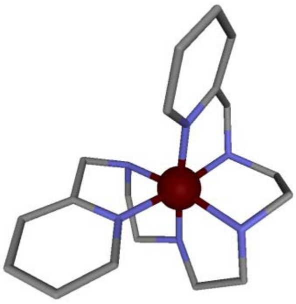
10. $[Fe(PypepO)_2]^{1-/2-}$



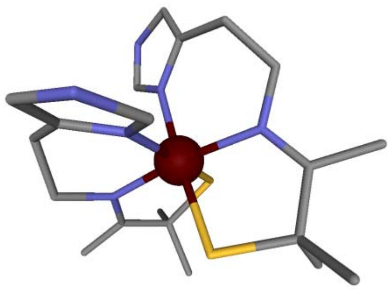
11. $[Fe(PypepS)_2]^{1-/2-}$



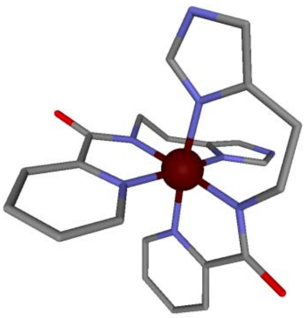
12. $[Fe(PyAS)_2]^{1+/0}$



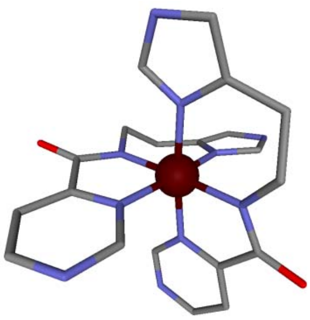
13. $[Fe(bpteta)]^{3+/2+}$



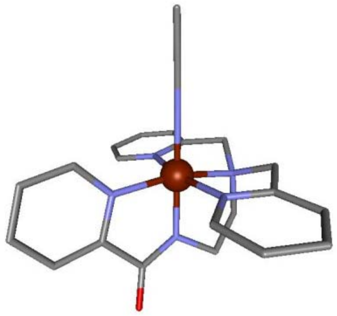
14. $[Fe(DITim)_2]^{1+/0}$



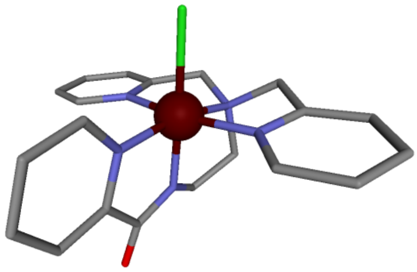
15. $[Fe(Pypep)_2]^{1+/0}$



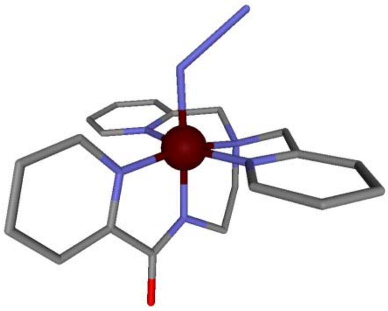
16. $[Fe(Prpep)_2]^{1+/0}$



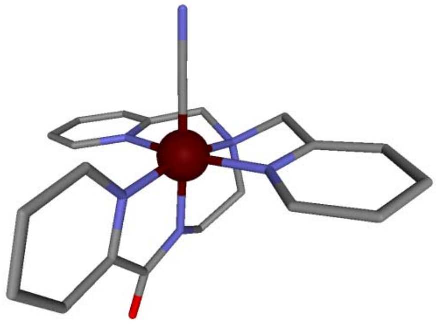
17. $[Fe(PaPy_3)(AN)]^{2+/1+}$



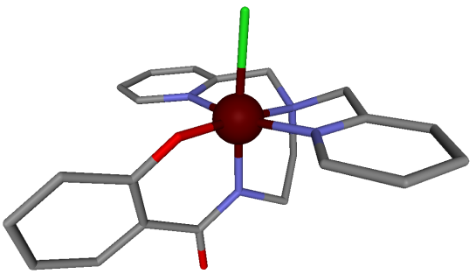
18. $[Fe(PaPy_3)(Cl)]^{1+/0}$



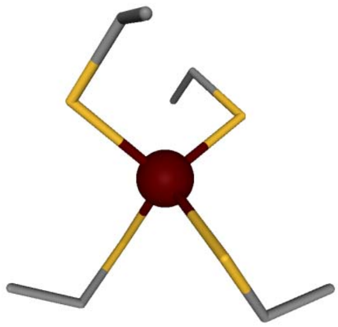
19. $[Fe(PaPy_3)(N_3)]^{1+/0}$



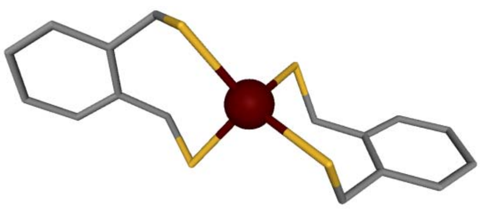
20. $[Fe(PaPy_3)(CN)]^{1+/0}$



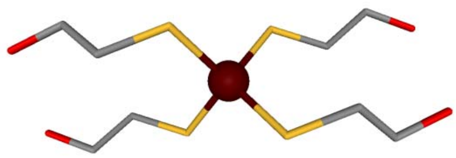
21. $[Fe(PaPy_2O)(Cl)]^{0/1-}$



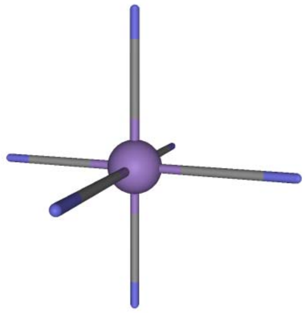
22. $[Fe(SEt)_4]^{1-/2-}$



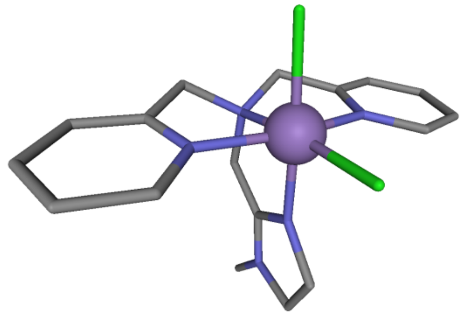
23. $[Fe(S_2\text{-o-xyl})_2]^{1-/2-}$



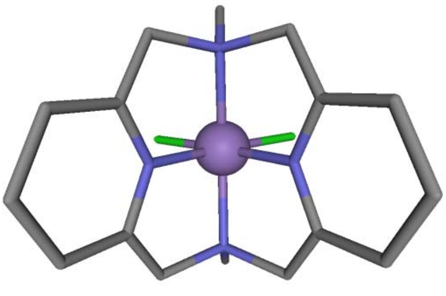
24. $[Fe(SEtOH)_4]^{1-/2-}$



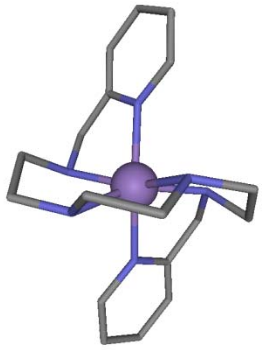
25. $[Mn(CN)_6]^{3-/4-}$



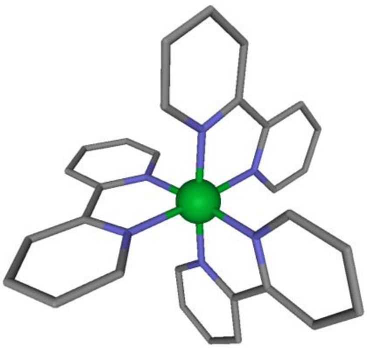
26. $[Mn(bpia)(Cl)_2]^{2+/1+}$



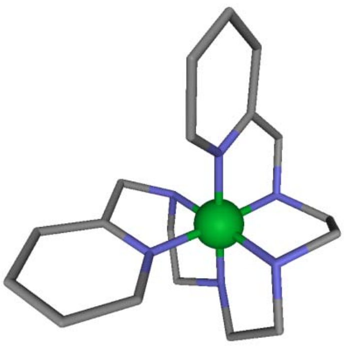
27. $[Mn(py_2(NMe)_2Cl_2]^{2+/1+}$



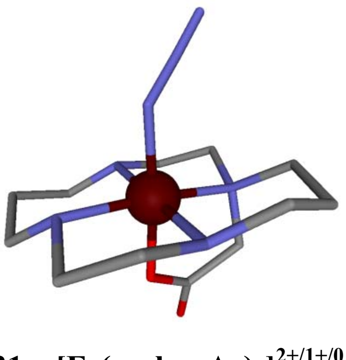
28. $[Mn(bpteta)]^{3+/2+}$



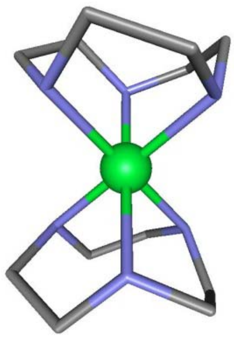
29. $[Ni(bpy)_3]^{3+/2+}$



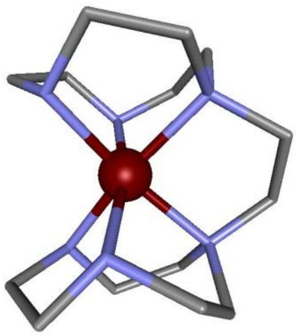
30. $[Ni(bpteta)]^{3+/2+}$



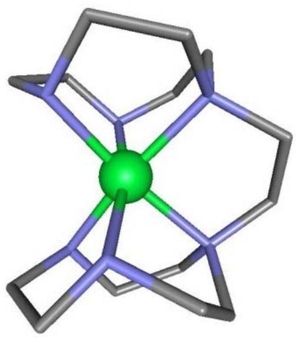
31. $[Fe(cyclamAc)_4]^{2+/1+/0}$



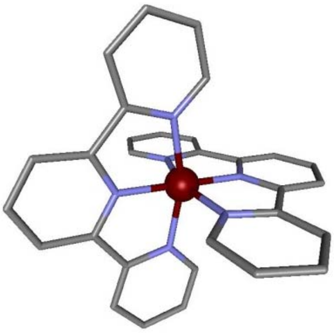
32. $[Ni(tacn)]^{3+/2+}$



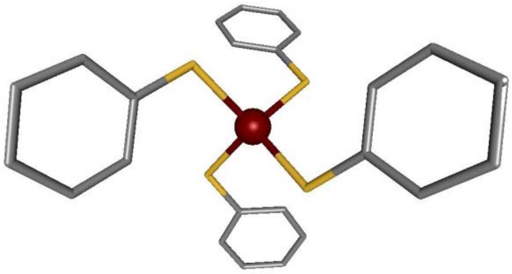
33. [Fe(dtne)]$^{3+/2+}$



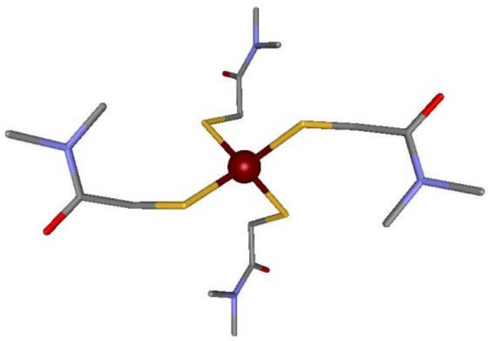
34. [Ni(dtne)]$^{3+/2+}$



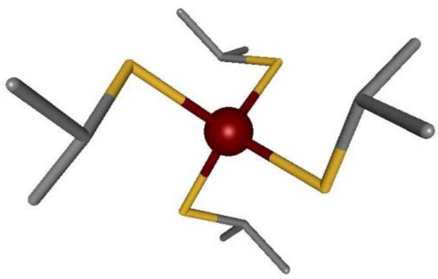
35. [Fe(terpy)]$^{3+/2+}$



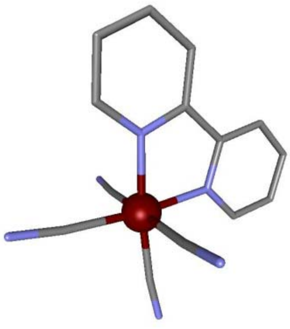
36. [Fe(SPh)$_4$]$^{1-/2-}$



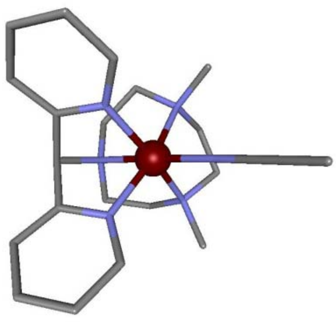
37. [Fe(SCH$_2$CON(CH$_3$)$_2$)$_4$]$^{1-/2-}$



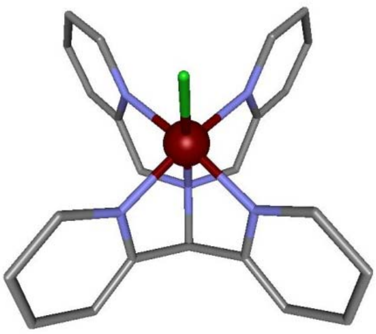
38. [Fe(S-*i*-Pr)$_4$]$^{1-/2-}$



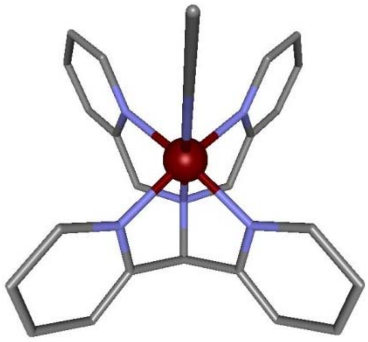
39. [Fe(bpy)(CN)$_4$]$^{3+/2+}$



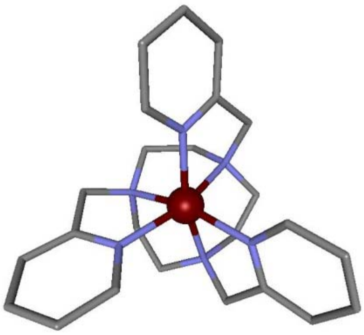
40. [Fe(tacnPy$_2$)(AN)]$^{3+/2+}$



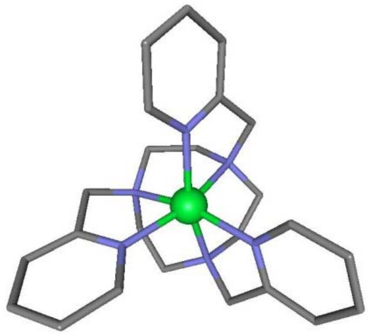
41. $[Fe(N_4Py)(Cl)]^{2+/1+}$



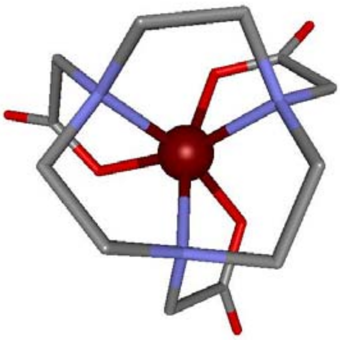
42. $[Fe(N_4Py)(AN)]^{3+/2+}$



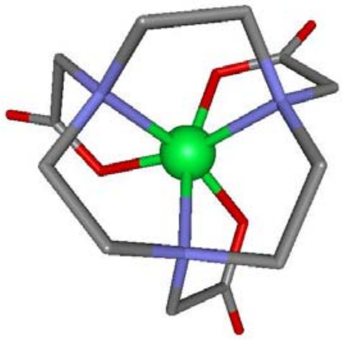
43. $[Fe(Py_3tacn)]^{3+/2+}$



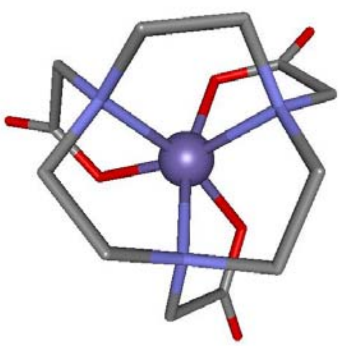
44. $[Ni(Py_3tacn)]^{3+/2+}$



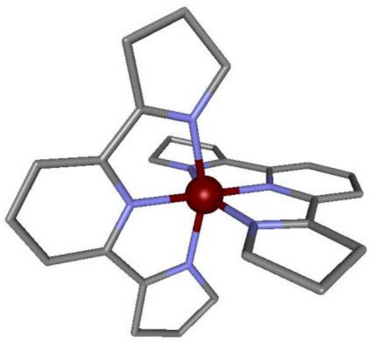
45. $[Fe(TCTA)]^{0/1-}$



46. $[Ni(TCTA)]^{0/1-}$



47. $[Mn(TCTA)]^{0/1-}$



48. $[Fe(Pyr_2Py)_2]^{3+/2+}$

Table A2. Calculated energy terms and correction energies G_X used in the **B4(XQ3)LYP-approach**.

The correction term G_X is calculated from the polynomial Ansatz $G_X(q) = Aq^3 + Bq^2 + Cq + D$, with coefficients A= –0.333, B = 1.545 and C = 21.634 with q given in units of the elementary charge. Coefficient D has been arbitrarily set to 0, since it cancels in the differences $\Delta G_X(q)$. All energies are given in units of kcal·mol^{-1}.

model compound	spin theory	E_0^{B4LYP}	ZPE	$\Delta G_{0\to 298K}$	G_X	G_g^0	ΔG_{sol}^0	G_s^0	solvent	spin exp.
		kcal·mol^{-1}								
training set (1-30)										
1. $[Fe(CN)_6]^{3-}$	1/2	**-422968.677**	30.716	-24.776	-41.790	**-423004.747**	-391.166	**-423395.913**	water	1/2[156]
1. $[Fe(CN)_6]^{3-}$	5/2	-422932.311	27.189	-28.568	-41.790	-422975.700	-376.897	-423352.597	water	
1. $[Fe(CN)_6]^{4-}$	0	**-422781.253**	29.923	-24.518	-40.250	**-422816.365**	-691.396	**-423507.761**	water	
1. $[Fe(CN)_6]^{4-}$	2	-422762.843	25.296	-30.994	-40.250	-422809.058	-652.496	-423461.554	water	
2. $[Fe(bpy)_3]^{3+}$	1/2	**-999027.385**	305.553	-36.564	69.513	**-998688.570**	-289.042 -277.750 -269.011	**-998977.612** **-998966.320** **-998957.581**	water AN DMF	
2. $[Fe(bpy)_3]^{3+}$	5/2	-999011.164	303.291	-39.582	69.513	-998677.629	-284.619 -273.782 -265.380	-998962.248 -998951.411 -998943.009	water AN DMF	
2. $[Fe(bpy)_3]^{2+}$	0	**-999287.743**	304.955	-36.900	46.586	**-998972.901**	-131.410 -125.920 -120.611	**-999104.311** **-999098.821** **-999093.512**	water AN DMF	0[131]
2. $[Fe(bpy)_3]^{2+}$	2	-999274.269	305.537	-40.206	46.586	-998962.151	-128.616 -123.431 -118.463	-999090.767 -999085.582 -999080.614	water AN DMF	
3. $[Fe(bpy)_2(CN)_2]^{1+}$	1/2	**-807249.991**	213.887	-33.331	22.750	**-807046.589**	-62.248	**-807108.837**	water	
3. $[Fe(bpy)_2(CN)_2]^{1+}$	5/2	-807224.000	211.151	-37.192	22.750	-807027.195	-58.889	-807086.084	water	
3. $[Fe(bpy)_2(CN)_2]^{0}$	0	**-807372.425**	212.874	-33.025	0	**-807192.576**	-38.159	**-807230.735**	water	
3. $[Fe(bpy)_2(CN)_2]^{0}$	2	-807351.434	209.900	-38.188	0	-807179.722	-35.741	-807215.463	water	
4. $[Fe(phen)_3]^{3+}$	1/2	**-1141007.360**	329.319	-36.195	69.513	**-1140644.410**	-278.196 -267.357 -258.528	**-1140922.606** **-1140911.767** **-1140902.938**	water AN DMF	
4. $[Fe(phen)_3]^{3+}$	5/2	-1140992.474	327.512	-38.780	69.513	-1140633.916	-274.108 -263.819 -255.380	-1140908.024 -1140897.735 -1140889.296	water AN DMF	
4. $[Fe(phen)_3]^{2+}$	0	**-1141263.367**	328.962	-36.295	46.586	**-1140923.913**	-126.922 -121.599 -116.153	**-1141050.835** **-1141045.512** **-1141040.066**	water AN DMF	0[131]
4. $[Fe(phen)_3]^{2+}$	2	-1141251.233	326.959	-40.942	46.586	-1140918.429	-124.844 -119.708 -114.449	-1141043.273 -1141038.137 -1141032.878	water AN DMF	
5. $[Fe(diammac)]^{3+}$	1/2	**-575266.075**	287.584	-26.351	69.513	**-574935.016**	-370.965	**-575305.981**	water	1/2[142]
5. $[Fe(diammac)]^{3+}$	5/2	-575241.637	284.192	-28.560	69.513	-574916.179	-366.310	-575282.489	water	
5. $[Fe(diammac)]^{2+}$	0	**-575547.596**	286.805	-26.187	46.586	**-575240.191**	-167.401	**-575407.592**	water	
5. $[Fe(diammac)]^{2+}$	2	-575534.502	283.524	-28.136	46.586	-575232.327	-165.555	-575397.882	water	
6. $[Fe(sar)]^{3+}$	1/2	-623291.056	315.435	-26.143	69.513	**-622931.938**	-362.269	**-623294.207**	water	1/2[157]
6. $[Fe(sar)]^{3+}$	5/2	-623279.636	309.797	-27.884	69.513	-622927.897	-356.576	-623284.473	water	
6. $[Fe(sar)]^{2+}$	0	-623569.230	311.811	-26.271	46.586	**-623236.903**	-163.102	**-623400.005**	water	0[132]
6. $[Fe(sar)]^{2+}$	2	-623561.594	309.353	-29.469	46.586	-623234.923	-160.454	-623395.377	water	
7. $[Fe(tacn)_2]^{3+}$	1/2	-575239.218	290.071	-27.468	69.513	**-574906.789**	-370.195	**-575276.984**	water	1/2[158]
7. $[Fe(tacn)_2]^{3+}$	5/2	-575227.461	286.972	-28.085	69.513	-574898.748	-363.689	-575262.437	water	
7. $[Fe(tacn)_2]^{2+}$	0	-575528.743	289.313	-25.182	46.586	**-575217.825**	-166.030	**-575383.855**	water	0[158] spin-crossover[130]
7. $[Fe(tacn)_2]^{2+}$	2	-575521.891	286.534	-27.604	46.586	-575216.174	-163.698	-575379.872	water	20%HS at 278K
8. $[Fe(PyIm_2H_2)_2]^{3+}$	1/2	-943739.682	248.616	-32.582	69.513	**-943453.822**	-301.079	**-943754.901**	AN	
8. $[Fe(PyIm_2H_2)_2]^{3+}$	5/2	-943729.681	246.486	-36.054	69.513	-943449.423	-296.312	-943745.735	AN	

8. [Fe(PyIm$_2$H$_2$)$_2$]$^{2+}$	0	-943997.880	247.748	-32.669	46.586	**-943736.014**	-142.874	**-943878.888**	AN	spin-crossover[106]
8. [Fe(PyIm$_2$H$_2$)$_2$]$^{2+}$	2	-943991.721	245.703	-36.635	46.586	-943735.866	-140.476	-943876.342	AN	spin-crossover
9. [Fe(PyIm$_2$)$_2$]$^{1-}$	1/2	-942865.117	214.817	-32.740	-19.670	**-942702.796**	-49.817	**-942752.613**	AN	1/2[106]
9. [Fe(PyIm$_2$)$_2$]$^{1-}$	5/2	-942849.953	212.387	-36.161	-19.670	-942693.483	-48.560	-942742.043	AN	
9. [Fe(PyIm$_2$)$_2$]$^{2-}$	0	-942844.031	212.615	-33.103	-34.265	**-942698.944**	-146.626	**-942845.570**	AN	
9. [Fe(PyIm$_2$)$_2$]$^{2-}$	2	-942838.205	220.310	-35.915	-34.265	-942688.235	-143.464	-942831.699	AN	
10. [Fe(PypepO)$_2$]$^{1-}$	1/2	-973923.604	225.576	-34.001	-19.670	-973751.699	-50.837	-973802.536	DMF	
10. [Fe(PypepO)$_2$]$^{1-}$	5/2	-973922.527	224.303	-36.071	-19.670	**-973753.965**	-48.775	**-973802.740**	DMF	5/2[109]
10. [Fe(PypepO)$_2$]$^{2-}$	0	-973889.930	223.475	-33.572	-34.265	-973734.292	-147.865	-973882.157	DMF	
10. [Fe(PypepO)$_2$]$^{2-}$	2	-973893.968	223.115	-36.472	-34.265	**-973741.590**	-145.804	**-973887.394**	DMF	
11. [Fe(PypepS)$_2$]$^{1-}$	1/2	-1377584.387	223.127	-35.402	-19.670	**-1377416.418**	-48.364	**-1377464.782**	DMF	1/2[110]
11. [Fe(PypepS)$_2$]$^{1-}$	5/2	-1377574.917	221.733	-38.050	-19.670	-1377410.990	-46.455	-1377457.445	DMF	
11. [Fe(PypepS)$_2$]$^{2-}$	0	-1377552.206	221.234	-35.215	-34.265	**-1377400.612**	-144.017	**-1377544.629**	DMF	
11. [Fe(PypepS)$_2$]$^{2-}$	2	-1377549.342	229.245	-37.126	-34.265	-1377391.648	-142.205	-1377533.853	DMF	
12. [Fe(PyAS)$_2$]$^{1+}$	1/2	-1284506.117	233.311	-34.210	22.750	**-1284284.170**	-40.029	**-1284324.199**	DMF	1/2[110, 133]
12. [Fe(PyAS)$_2$]$^{1+}$	5/2	-1284486.294	232.616	-36.983	22.750	-1284267.815	-38.579	-1284306.394	DMF	
12. [Fe(PyAS)$_2$]^{0}	0	-1284615.904	231.975	-34.197	0	**-1284418.126**	-13.209	**-1284431.335**	DMF	0[133]
12. [Fe(PyAS)$_2$]^{0}	2	-1284604.601	230.346	-36.775	0	-1284411.030	-14.694	-1284425.724	DMF	
13. [Fe(bpteta)]$^{3+}$	1/2	-716508.728	294.967	-30.055	69.513	**-716173.990**	-326.462	**-716500.452**	AN	
13. [Fe(bpteta)]$^{3+}$	5/2	-716496.873	292.202	-32.686	69.513	-716167.531	-320.844	-716488.375	AN	
13. [Fe(bpteta)]$^{2+}$	0	-716784.698	294.258	-29.925	46.586	**-716473.578**	-147.516	**-716621.094**	AN	0[101]
13. [Fe(bpteta)]$^{2+}$	2	-716779.594	291.992	-33.080	46.586	**-716473.895**	-144.220	-716618.115	AN	
14. [Fe(DITim)$_2$]$^{1+}$	1/2	-1262996.034	323.839	-36.550	22.750	**-1262685.899**	-48.253	**-1262734.152**	AN	1/2[107]
14. [Fe(DITim)$_2$]$^{1+}$	5/2	-1262983.878	322.014	-39.134	22.750	-1262678.152	-47.174	-1262725.326	AN	
14. [Fe(DITim)$_2$]^{0}	0	-1263079.595	322.005	-36.166	0	-1262793.756	-23.292	-1262817.048	AN	
14. [Fe(DITim)$_2$]^{0}	2	-1263082.536	320.408	-39.309	0	**-1262801.437**	-22.391	**-1262823.828**	AN	
15. [Fe(Pypep)$_2$]$^{1+}$	1/2	-971149.214	273.960	-34.766	22.750	**-970887.174**	-61.860	**-970949.034**	water	1/2[114]
							-54.608	**-970941.782**	AN	
							-49.562	**-970936.736**	DMF	
15. [Fe(Pypep)$_2$]$^{1+}$	5/2	-971137.124	271.564	-37.806	22.750	-970880.520	-57.417	-970937.937	water	
							-52.086	-970932.606	AN	
							-47.535	-970928.055	DMF	
15. [Fe(Pypep)$_2$]^{0}	0	-971253.083	271.926	-34.775	0	**-971015.932**	-33.393	**-971049.325**	water	
							-25.058	**-971040.990**	AN	
							-21.777	**-971037.709**	DMF	
15. [Fe(Pypep)$_2$]^{0}	2	-971246.671	269.760	-38.762	0	**-971015.673**	-30.282	-971045.955	water	
							-24.491	-971040.164	AN	
							-21.255	-971036.928	DMF	
16. [Fe(Prpep)$_2$]$^{1+}$	1/2	-991166.855	258.928	-34.819	22.750	**-990919.900**	-65.774	**-990985.674**	AN	1/2[105]
16. [Fe(Prpep)$_2$]$^{1+}$	5/2	-991157.207	256.592	-37.839	22.750	-990915.608	-63.712	-990979.320	AN	
16. [Fe(Prpep)$_2$]^{0}	0	-991280.643	257.152	-35.014	0	**-991058.505**	-34.655	**-991093.160**	AN	0[105]
16. [Fe(Prpep)$_2$]^{0}	2	-991274.852	255.081	-38.769	0	**-991058.540**	-33.841	-991092.381	AN	
17. [Fe(PaPy$_3$)(AN)]$^{2+}$	1/2	-856424.397	269.432	-34.807	46.586	**-856142.985**	-141.658	**-856284.643**	AN	1/2[113]
17. [Fe(PaPy$_3$)(AN)]$^{2+}$	5/2	-856410.532	266.800	-38.234	46.586	-856135.179	-136.722	-856271.901	AN	
17. [Fe(PaPy$_3$)(AN)]$^{1+}$	0	-856607.403	268.355	-35.027	22.750	**-856351.229**	-47.296	**-856398.525**	AN	
17. [Fe(PaPy$_3$)(AN)]$^{1+}$	2	-856596.955	266.235	-38.490	22.750	-856346.364	-45.751	-856392.115	AN	
18. [Fe(PaPy$_3$)(Cl)]$^{1+}$	1/2	-1061605.435	241.645	-31.852	22.750	**-1061372.796**	-50.240	**-1061423.036**	AN	1/2[113]
18. [Fe(PaPy$_3$)(Cl)]$^{1+}$	5/2	-1061596.339	238.218	-36.025	22.750	-1061371.300	-47.917	-1061419.217	AN	
18. [Fe(PaPy$_3$)(Cl)]^{0}	0	-1061714.598	238.712	-32.218	0	-1061508.104	-23.311	**-1061531.415**	AN	
18. [Fe(PaPy$_3$)(Cl)]^{0}	2	-1061711.734	237.179	-35.110	0	**-1061509.665**	-20.481	-1061530.146	AN	
19. [Fe(PaPy$_3$)(N$_3$)]$^{1+}$	1/2	-876245.479	247.793	-33.300	22.750	**-876008.140**	-49.403	**-876057.543**	AN	1/2[113]
19. [Fe(PaPy$_3$)(N$_3$)]$^{1+}$	5/2	-876237.371	245.489	-36.605	22.750	-876005.641	-47.227	-876052.868	AN	

19. $[Fe(PaPy_3)(N_3)]^{0}$	0	-876353.663	246.345	-33.138	0	-876140.456	-20.558	**-876161.014**	AN	
19. $[Fe(PaPy_3)(N_3)]^{0}$	2	-876350.081	244.428	-36.660	0	**-876142.313**	-18.665	-876160.978	AN	
20. $[Fe(PaPy_3)(CN)]^{1+}$	1/2	-831850.633	245.264	-32.501	22.750	**-831615.024**	-53.572	**-831668.596**	AN	1/2[113]
20. $[Fe(PaPy_3)(CN)]^{1+}$	5/2	-831831.641	242.036	-36.179	22.750	-831602.938	-52.025	-831654.963	AN	
20. $[Fe(PaPy_3)(CN)]^{0}$	0	-831964.838	243.129	-32.402	0	**-831754.111**	-24.154	**-831778.265**	AN	
20. $[Fe(PaPy_3)(CN)]^{0}$	2	-831951.488	240.736	-35.425	0	-831746.177	-22.543	-831768.720	AN	
21. $[Fe(PaPy_2O)(Cl)]^{0}$	1/2	-1098138.006	246.809	-32.348	0	-1097923.545	-19.690	-1097943.235	DMF	
21. $[Fe(PaPy_2O)(Cl)]^{0}$	5/2	-1098139.280	240.111	-35.572	0	**-1097934.741**	-18.363	**-1097953.104**	DMF	5/2[109]
21. $[Fe(PaPy_2O)(Cl)]^{1-}$	0	-1098170.194	239.863	-33.238	-19.670	-1097983.325	-58.4354	-1098041.760	DMF	
21. $[Fe(PaPy_2O)(Cl)]^{1-}$	2	-1098173.782	239.343	-36.784	-19.670	**-1097990.979**	-58.727	**-1098049.706**	DMF	
22. $[Fe(SEt)_4]^{1-}$	1/2	-1267958.227	169.214	-33.884	-19.670	-1267842.653	-44.297	-1267886.950	AN	
22. $[Fe(SEt)_4]^{1-}$	5/2	-1267982.269	169.423	-37.457	-19.670	**-1267870.059**	-44.330	**-1267914.389**	AN	
22. $[Fe(SEt)_4]^{2-}$	1	-1267920.915	168.103	-35.347	-34.265	-1267822.584	-157.088	-1267979.672	AN	
22. $[Fe(SEt)_4]^{2-}$	2	-1267937.481	168.081	-37.144	-34.265	**-1267840.969**	-157.843	**-1267998.812**	AN	
23. $[Fe(S_2\text{-o-xyl})_2]^{1-}$	1/2	-1455772.122	175.797	-31.929	-19.670	-1455648.010	-46.8206	-1455694.831	DMF	
23. $[Fe(S_2\text{-o-xyl})_2]^{1-}$	5/2	-1455792.775	175.801	-34.020	-19.670	**-1455670.750**	-45.682	**-1455716.432**	DMF	5/2[119]
23. $[Fe(S_2\text{-o-xyl})_2]^{2-}$	1	-1455740.110	174.325	-33.041	-34.265	-1455633.251	-156.083	-1455789.334	DMF	
23. $[Fe(S_2\text{-o-xyl})_2]^{2-}$	2	-1455757.616	174.335	-32.838	-34.265	**-1455650.544**	-151.448	**-1455801.992**	DMF	2[119]
24. $[Fe(SEtOH)_4]^{1-}$	1/2	-1455150.782	199.247	-31.109	-19.670	-1455002.314	-67.591	-1455069.905	water	
24. $[Fe(SEtOH)_4]^{1-}$	5/2	-1455184.195	181.058	-37.270	-19.670	**-1455060.077**	-71.019	**-1455131.096**	water	
24. $[Fe(SEtOH)_4]^{2-}$	1	-1455121.202	179.726	-36.807	-34.265	-1455012.548	-193.297	-1455205.845	water	
24. $[Fe(SEtOH)_4]^{2-}$	2	-1455152.063	184.696	-36.953	-34.265	**-1455038.585**	-192.200	**-1455230.785**	water	2[116]
25. $[Mn(CN)_6]^{3-}$	1	-410796.584	29.926	-25.536	-41.790	**-410834.204**	-387.796	**-411222.000**	water	
25. $[Mn(CN)_6]^{3-}$	2	-410785.090	27.734	-28.103	-41.790	-410827.469	-378.415	-411205.884	water	
25. $[Mn(CN)_6]^{4-}$	1/2	-410596.184	29.143	-25.527	-40.250	-410633.085	-684.755	**-411317.840**	water	1/2[134]
25. $[Mn(CN)_6]^{4-}$	5/2	-410610.090	24.959	-31.493	-40.250	**-410657.141**	-644.859	-411302.000	water	
26. $[Mn(bpia)(Cl)_2]^{2+}$	3/2	-1218510.028	219.534	-32.223	46.586	-1218275.930	-148.358	-1218424.288	AN	
26. $[Mn(bpia)(Cl)_2]^{1+}$	1	-1218712.370	219.410	-32.187	22.750	-1218502.301	-53.875	-1218556.176	AN	
26. $[Mn(bpia)(Cl)_2]^{1+}$	2	-1218721.892	218.100	-33.268	22.750	**-1218514.214**	-51.7645	**-1218565.979**	AN	
26. $[Mn(bpia)(Cl)_2]^{0}$	1/2	-1218807.941	217.959	-33.104	0	-1218623.086	-30.876	-1218653.962	AN	
26. $[Mn(bpia)(Cl)_2]^{0}$	5/2	-1218836.455	216.463	-35.572	0	**-1218655.564**	-32.4007	**-1218687.965**	AN	
27. $[Mn(py_2(NMe)_2Cl_2)]^{2+}$	3/2	-1161166.129	220.361	-30.417	46.586	-1160929.398	-155.520	-1161084.918	AN	
27. $[Mn(py_2(NMe)_2Cl_2)]^{1+}$	1	-1161374.181	220.552	-30.508	22.750	-1161161.291	-55.1719	-1161216.463	AN	
27. $[Mn(py_2(NMe)_2Cl_2)]^{1+}$	2	-1161384.281	219.020	-32.461	22.750	**-1161174.876**	-51.597	**-1161226.473**	AN	
27. $[Mn(py_2(NMe)_2Cl_2)]^{0}$	1/2	-1161478.947	218.848	-30.563	0	-1161290.662	-28.761	-1161319.423	AN	
27. $[Mn(py_2(NMe)_2Cl_2)]^{0}$	5/2	-1161498.449	217.150	-31.728	0	**-1161313.027**	-30.344	**-1161343.371**	AN	5/2[100]
28. $[Mn(bpteta)]^{3+}$	1	-704328.626	293.769	-31.137	69.513	-703996.168	-321.128	-704317.296	AN	
28. $[Mn(bpteta)]^{3+}$	2	-704335.910	292.402	-32.070	69.513	**-704005.752**	-318.342	-704324.094	AN	
28. $[Mn(bpteta)]^{2+}$	1/2	-704586.822	299.437	-30.857	46.586	-704271.455	-144.365	-704415.820	AN	
28. $[Mn(bpteta)]^{2+}$	5/2	-704620.010	291.438	-33.603	46.586	**-704315.388**	-143.191	-704458.579	AN	5/2[101]
29. $[Ni(bpy)_3]^{3+}$	1/2	-1027612.871	304.240	-37.504	69.513	**-1027276.309**	-288.437	-1027564.746	water	
29. $[Ni(bpy)_3]^{3+}$	3/2	-1027597.725	301.911	-39.759	69.513	-1027265.747	-283.266	-1027549.013	water	
29. $[Ni(bpy)_3]^{2+}$	1	-1027890.244	303.452	-39.419	46.586	**-1027579.424**	-129.731	-1027709.155	water	
30. $[Ni(bpteta)]^{3+}$	1/2	-745103.784	293.570	-30.746	69.513	**-744771.134**	-325.745	-745096.879	AN	
30. $[Ni(bpteta)]^{3+}$	3/2	-745077.519	290.270	-32.711	69.513	-744750.134	-315.733	-745065.867	AN	
30. $[Ni(bpteta)]^{2+}$	1	-745395.996	292.715	-31.884	46.586	**-745088.378**	-146.632	-745235.010	AN	1[101]
prediction set (31-48)										

31. [Fe(cyclamAc)(N$_3$)]$^{2+}$	1	-701330.364	256.999	-30.144	46.586	**-701056.923**	-167.315	**-701224.238**	AN	
31. [Fe(cyclamAc)(N$_3$)]$^{2+}$	2	-701313.303	254.637	-31.438	46.586	-701043.518	-164.125	-701207.643	AN	
31. [Fe(cyclamAc)(N$_3$)]$^{1+}$	1/2	-701557.877	258.049	-29.033	22.750	**-701306.111**	-56.615	**-701362.726**	AN	1/2[145]
31. [Fe(cyclamAc)(N$_3$)]$^{1+}$	5/2	-701542.408	254.887	-32.128	22.750	-701296.899	-55.866	-701352.765	AN	
31. [Fe(cyclamAc)(N$_3$)]^{0}	0	-701669.143	255.994	-29.580	0	**-701442.729**	-19.810	**-701462.539**	AN	0[145]
31. [Fe(cyclamAc)(N$_3$)]^{0}	2	-701663.717	253.401	-32.101	0	-701442.417	-19.420	-701461.837	AN	
32. [Ni(tacn)]$^{3+}$	1/2	-603838.888	288.387	-26.556	69.513	**-603507.544**	-369.019	**-603876.563**	water	1/2[117]
32. [Ni(tacn)]$^{3+}$	3/2	-603809.570	286.921	-27.130	69.513	-603480.266	-365.770	-603846.036	water	
32. [Ni(tacn)]$^{2+}$	1	-604141.954	287.768	-26.884	46.586	**-603834.484**	-164.730	**-603999.214**	water	
33. [Fe(dtne)]$^{3+}$	1/2	-623262.235	311.814	-26.751	69.513	-622907.659	-360.350	**-623268.009**	water	1/2[146]
33. [Fe(dtne)]$^{3+}$	5/2	-623258.123	309.483	-28.544	69.513	**-622907.671**	-355.150	-623262.821	water	
33. [Fe(dtne)]$^{2+}$	0	-623545.643	311.429	-26.361	46.586	-623213.989	-161.440	-623375.429	water	
33. [Fe(dtne)]$^{2+}$	2	-623547.026	309.403	-28.568	46.586	**-623219.605**	-158.680	**-623378.285**	water	
34. [Ni(dtne)]$^{3+}$	1/2	-651860.085	310.472	-26.724	69.513	**-651506.824**	-360.034	**-651866.858**	water	
34. [Ni(dtne)]$^{3+}$	3/2	-651846.366	308.583	-28.766	69.513	-651497.036	-353.310	-651850.346	water	
34. [Ni(dtne)]$^{2+}$	1	-652162.844	310.039	-27.836	46.586	**-651834.055**	-160.050	**-651994.105**	water	1[146]
35. [Fe(terpy)$_2$]$^{3+}$	1/2	-998303.812	292.794	-35.324	69.513	**-997976.829**	-270.350	**-998247.179**	DMF	
35. [Fe(terpy)$_2$]$^{3+}$	5/2	-998293.814	289.712	-38.248	69.513	-997972.837	-266.800	-998239.637	DMF	
35. [Fe(terpy)$_2$]$^{2+}$	0	-998569.395	291.313	-35.138	46.586	**-998266.634**	-121.530	**-998388.164**	DMF	
35. [Fe(terpy)$_2$]$^{2+}$	2	-998555.049	289.157	-39.560	46.586	-998258.866	-119.250	-998378.116	DMF	
36. [Fe(SPh)$_4$]$^{1-}$	1/2	-1646491.370	231.134	-41.931	-19.670	-1646321.837	-43.71	-1646365.547	AN	
36. [Fe(SPh)$_4$]$^{1-}$	5/2	-1646508.710	230.484	-43.501	-19.670	**-1646341.397**	-43.35	**-1646384.747**	AN	
36. [Fe(SPh)$_4$]$^{2-}$	1	-1646475.610	229.890	-43.341	-34.265	-1646323.326	-137.39	-1646460.716	AN	
36. [Fe(SPh)$_4$]$^{2-}$	2	-1646495.010	228.975	-43.463	-34.265	**-1646343.763**	-137.68	**-1646481.443**	AN	
37. [Fe(SCH$_2$CON(CH$_3$)$_2$)$_4$]$^{1-}$	1/2	-1784947.463	310.343	-49.167	-19.670	-1784705.957	-50.733 -49.403	-1784756.690 -1784755.360	AN DMF	
37. [Fe(SCH$_2$CON(CH$_3$)$_2$)$_4$]$^{1-}$	5/2	-1784964.787	308.613	-51.020	-19.670	**-1784726.864**	-48.978 -47.594	**-1784775.842** **-1784774.458**	AN DMF	
37. [Fe(SCH$_2$CON(CH$_3$)$_2$)$_4$]$^{2-}$	1	-1784929.981	307.615	-48.501	-34.265	-1784705.132	-139.201 -137.523	-1784844.333 -1784842.655	AN DMF	
37. [Fe(SCH$_2$CON(CH$_3$)$_2$)$_4$]$^{2-}$	2	-1784952.198	307.096	-51.149	-34.265	**-1784730.516**	-138.455 -136.441	**-1784868.971** **-1784866.957**	AN DMF	
38. [Fe(S-*i*-Pr)$_4$]$^{1-}$	1/2	-1365486.520	239.758	-37.753	-19.670	-1365304.185	-42.840	-1365347.025	AN	
38. [Fe(S-*i*-Pr)$_4$]$^{1-}$	5/2	-1365511.678	239.880	-41.083	-19.670	**-1365332.551**	-42.818	**-1365375.369**	AN	
38. [Fe(S-*i*-Pr)$_4$]$^{2-}$	1	-1365453.305	283.404	-37.741	-34.265	-1365241.907	-149.016	-1365390.923	AN	
38. [Fe(S-*i*-Pr)$_4$]$^{2-}$	2	-1365469.848	238.400	-41.235	-34.265	**-1365306.948**	-149.880	**-1365456.828**	AN	
39. [Fe(bpy)(CN)$_4$]$^{3+}$	1/2	-615233.248	122.642	-30.131	69.513	**-615160.407**	-82.749	**-615243.156**	water	
39. [Fe(bpy)(CN)$_4$]$^{3+}$	5/2	-615206.415	119.485	-33.825	69.513	-615140.425	-74.900	-615215.325	water	
39. [Fe(bpy)(CN)$_4$]$^{2+}$	0	-615215.939	120.810	-29.920	46.586	**-615159.314**	-200.870	**-615360.184**	water	
39. [Fe(bpy)(CN)$_4$]$^{2+}$	2	-615188.138	117.277	-33.969	46.586	-615139.095	-199.798	-615338.893	water	
40. [Fe(tacnPy$_2$)(AN)]$^{3+}$	1/2	-788192.942	316.283	-32.647	69.513	**-787839.793**	-305.298	**-788145.091**	AN	
40. [Fe(tacnPy$_2$)(AN)]$^{3+}$	5/2	-788179.616	312.923	-35.395	69.513	-787832.575	-300.986	-788133.561	AN	
40. [Fe(tacnPy$_2$)(AN)]$^{2+}$	0	-788465.568	315.147	-32.498	46.586	**-788136.333**	-136.942	**-788273.275**	AN	0[112]
40. [Fe(tacnPy$_2$)(AN)]$^{2+}$	2	-788455.336	312.777	-35.856	46.586	-788131.829	-134.462	-788266.291	AN	
41. [Fe(N$_4$Py)(Cl)]$^{2+}$	1/2	-1085939.903	263.119	-31.984	46.586	-1085662.182	-139.806	-1085801.988	AN	
41. [Fe(N$_4$Py)(Cl)]$^{2+}$	5/2	-1085936.276	254.981	-35.177	46.586	**-1085669.886**	-136.901	**-1085806.787**	AN	
41. [Fe(N$_4$Py)(Cl)]$^{1+}$	0	-1086129.799	263.805	-31.955	22.750	-1085875.199	-47.711	-1085922.910	AN	
41. [Fe(N$_4$Py)(Cl)]$^{1+}$	2	-1086129.194	253.814	-35.027	22.750	**-1085887.657**	-45.560	**-1085933.217**	AN	2[111]
42. [Fe(N$_4$Py)(AN)]$^{3+}$	1/2	-880698.030	285.342	-34.723	69.513	**-880377.898**	-290.331	**-880668.229**	AN	
42. [Fe(N$_4$Py)(AN)]$^{3+}$	5/2	-880681.975	283.102	-37.522	69.513	-880366.882	-286.482	-880653.364	AN	
42. [Fe(N$_4$Py)(AN)]$^{2+}$	0	-880964.934	285.053	-34.770	46.586	**-880668.065**	-131.806	**-880799.871**	AN	0[111]

42. [Fe(N$_4$Py)(AN)]$^{2+}$	2	-880952.988	282.981	-38.420	46.586	-880661.841	-129.527	-880791.368	AN	
43. [Fe(Py$_3$tacn)]$^{3+}$	1/2	-859179.690	330.931	-31.619	69.513	**-858810.865**	-302.070	**-859112.935**	AN	1/2[118]
43. [Fe(Py$_3$tacn)]$^{3+}$	5/2	-859169.010	328.415	-34.048	69.513	-858805.130	-296.883	-859102.013	AN	
43. [Fe(Py$_3$tacn)]$^{2+}$	0	-859446.270	329.952	-31.562	46.586	**-859101.294**	-138.213	**-859239.507**	AN	0[118]
43. [Fe(Py$_3$tacn)]$^{2+}$	2	-859437.519	327.501	-34.046	46.586	-859097.478	-132.977	-859230.455	AN	
44. [Ni(Py$_3$tacn)]$^{3+}$	1/2	-887771.766	329.336	-31.652	69.513	**-887404.569**	-301.650	**-887706.219**	AN	
44. [Ni(Py$_3$tacn)]$^{3+}$	3/2	-887749.278	326.769	-33.588	69.513	-887386.584	-301.902	-887688.486	AN	
44. [Ni(Py$_3$tacn)]$^{2+}$	1	-888055.739	328.598	-33.393	46.586	**-887713.948**	-134.151	**-887848.099**	AN	1[118]
45. [Fe(TCTA)]^{0}	1/2	-750241.605	202.766	-27.715	0.000	-750066.554	-39.970	-750106.524	water	
45. [Fe(TCTA)]^{0}	5/2	-750252.301	201.044	-29.647	0.000	**-750080.904**	-34.433	**-750115.337**	water	5/2[147]
45. [Fe(TCTA)]$^{1-}$	0	-750284.950	200.640	-27.944	-19.670	-750131.924	-76.677	-750208.601	water	
45. [Fe(TCTA)]$^{1-}$	2	-750299.368	199.236	-30.576	-19.670	**-750150.378**	-72.485	**-750222.863**	water	2[147]
46. [Ni(TCTA)]^{0}	1/2	-778833.123	201.245	-28.526	0.000	**-778660.404**	-38.962	**-778699.366**	water	
46. [Ni(TCTA)]^{0}	3/2	-778822.309	200.216	-29.439	0.000	-778651.532	-36.759	-778688.291	water	
46. [Ni(TCTA)]$^{1-}$	1	-778906.548	199.778	-29.499	-19.670	**-778755.939**	-75.129	**-778831.068**	water	1[147]
47. [Mn(TCTA)]^{0}	1	-738076.480	211.350	-27.708	0.000	-737892.838	-39.943	-737932.781	water	
47. [Mn(TCTA)]^{0}	2	-738083.395	200.810	-29.728	0.000	**-737912.313**	-36.229	**-737948.542**	water	2[147]
47. [Mn(TCTA)]$^{1-}$	1/2	-738104.833	203.770	-28.170	-19.670	-737948.903	-77.145	-738026.048	water	
47. [Mn(TCTA)]$^{1-}$	5/2	-738145.483	199.114	-30.830	-19.670	**-737996.869**	-71.304	**-738068.173**	water	5/2[147]
48. [Fe(Pyr$_2$Py)$_2$]$^{3+}$	1/2	-906529.945	334.302	-35.282	69.513	**-906161.412**	-286.304	**-906447.716**	AN	
48. [Fe(Pyr$_2$Py)$_2$]$^{3+}$	5/2	-906516.925	331.648	-38.289	69.513	-906154.053	-281.656	-906435.709	AN	
48. [Fe(Pyr$_2$Py)$_2$]$^{2+}$	0	-906796.987	333.723	-35.524	46.586	**-906452.202**	-127.524	**-906579.726**	AN	0[148]
48. [Fe(Pyr$_2$Py)$_2$]$^{2+}$	2	-906783.355	331.708	-38.881	46.586	-906443.942	-125.153	-906569.095	AN	

Table A3. Energy terms calculated using the B3LYP functional.

model compound	spin theory	E_{0K}	ZPE	$\Delta G_{0\rightarrow 298K}$	G^0_g	ΔG^0_{sol}	G^0_s	solvent	spin exp.
		kcal·mol^{-1}							
training set (1-30)									
1. $[Fe(CN)_6]^{3-}$	1/2	-427118.723	30.716	-24.776	**-427112.783**	-391.166	**-427503.949**	water	1/2[156]
1. $[Fe(CN)_6]^{3-}$	5/2	-427091.900	27.189	-28.568	-427093.279	-376.897	-427470.176	water	
1. $[Fe(CN)_6]^{4-}$	0	-426935.791	29.923	-24.518	-426930.386	-691.396	**-427621.782**	water	
1. $[Fe(CN)_6]^{4-}$	2	-426931.033	25.296	-30.994	**-426936.731**	-652.496	-427589.227	water	
2. $[Fe(bpy)_3]^{3+}$	1/2	-1009807.281	305.553	-36.564	**-1009538.292**	-289.042	**-1009827.334**	water	
						-277.750	**-1009816.042**	AN	
						-269.011	**-1009807.303**	DMF	
2. $[Fe(bpy)_3]^{3+}$	5/2	-1009800.229	303.291	-39.582	-1009536.520	-284.619	-1009821.139	water	
						-273.782	-1009810.302	AN	
						-265.380	-1009801.900	DMF	
2. $[Fe(bpy)_3]^{2+}$	0	-1010083.604	304.955	-36.900	**-1009815.549**	-131.410	**-1009946.959**	water	0[131]
						-125.920	**-1009941.469**	AN	
						-120.611	**-1009936.160**	DMF	
2. $[Fe(bpy)_3]^{2+}$	2	-1010080.497	305.537	-40.206	-1009815.166	-128.616	-1009943.782	water	
						-123.431	-1009938.597	AN	
						-118.463	-1009933.629	DMF	
3. $[Fe(bpy)_2(CN)_2]^{1+}$	1/2	-815822.852	213.887	-33.331	**-815642.296**	-62.248	**-815704.544**	water	
3. $[Fe(bpy)_2(CN)_2]^{1+}$	5/2	-815805.808	211.151	-37.192	-815631.849	-58.889	-815690.738	water	
3. $[Fe(bpy)_2(CN)_2]^{0}$	0	-815955.948	212.874	-33.025	**-815776.099**	-38.159	**-815814.257**	water	
3. $[Fe(bpy)_2(CN)_2]^{0}$	2	-815947.321	209.900	-38.188	-815775.609	-35.741	-815811.350	water	
4. $[Fe(phen)_3]^{3+}$	1/2	-1153351.101	329.319	-36.195	**-1153057.977**	-278.196	**-1153336.173**	water	
						-267.357	**-1153325.334**	AN	
						-258.528	**-1153316.505**	DMF	
4. $[Fe(phen)_3]^{3+}$	5/2	-1153345.767	327.512	-38.780	-1153057.035	-274.108	-1153331.143	water	
						-263.819	-1153320.854	AN	
						-255.380	-1153312.415	DMF	
4. $[Fe(phen)_3]^{2+}$	0	-1153622.663	328.962	-36.295	-1153329.996	-126.922	-1153456.918	water	0[131]
						-121.599	-1153451.595	AN	
						-116.153	-1153446.149	DMF	
4. $[Fe(phen)_3]^{2+}$	2	-1153621.835	326.959	-40.942	**-1153335.818**	-124.844	**-1153460.662**	water	
						-119.708	**-1153455.526**	AN	
						-114.449	**-1153450.267**	DMF	
5. $[Fe(diammac)]^{3+}$	1/2	-581483.147	287.584	-26.351	**-581221.914**	-370.965	**-581592.879**	water	1/2[142]
5. $[Fe(diammac)]^{3+}$	5/2	-581466.626	284.192	-28.560	-581210.994	-366.310	-581577.304	water	
5. $[Fe(diammac)]^{2+}$	0	-581784.327	286.805	-26.187	**-581523.709**	-167.401	**-581691.110**	water	
5. $[Fe(diammac)]^{2+}$	2	-581778.886	283.524	-28.136	-581523.498	-165.555	-581689.053	water	
6. $[Fe(sar)]^{3+}$	1/2	-630065.130	315.435	-26.143	-629775.838	-362.269	**-630138.107**	water	1/2[157]
6. $[Fe(sar)]^{3+}$	5/2	-630061.179	309.353	-29.469	**-629779.266**	-356.576	-630135.842	water	
6. $[Fe(sar)]^{2+}$	0	-630362.706	311.811	-26.271	-630077.166	-163.102	-630240.268	water	0[132]
6. $[Fe(sar)]^{2+}$	2	-630363.438	309.353	-29.469	**-630083.554**	-160.454	**-630244.008**	water	
7. $[Fe(tacn)_2]^{3+}$	1/2	-581455.505	290.071	-27.468	-581192.902	-370.195	**-581563.097**	water	1/2[158]
7. $[Fe(tacn)_2]^{3+}$	5/2	-581452.097	286.972	-28.085	**-581193.210**	-363.689	-581556.899	water	
7. $[Fe(tacn)_2]^{2+}$	0	-581764.886	289.313	-25.182	-581500.755	-166.030	-581666.785	water	0[158], spin-crossover[130]
7. $[Fe(tacn)_2]^{2+}$	2	-581765.698	286.534	-27.604	**-581506.768**	-163.698	**-581670.466**	water	20%HS at 278K
8. $[Fe(PyIm_2H_2)_2]^{3+}$	1/2	-953663.900	248.616	-32.582	-953447.866	-301.079	-953748.945	AN	
8. $[Fe(PyIm_2H_2)_2]^{3+}$	5/2	-953663.609	246.486	-36.054	**-953453.177**	-296.312	**-953749.489**	AN	
8. $[Fe(PyIm_2H_2)_2]^{2+}$	0	-953938.257	247.748	-32.669	-953723.178	-142.874	-953866.052	AN	spin-crossover[106]
8. $[Fe(PyIm_2H_2)_2]^{2+}$	2	-953943.093	245.703	-36.635	**-953734.025**	-140.476	**-953874.501**	AN	spin-crossover
9. $[Fe(PyIm_2)_2]^{1-}$	1/2	-952761.409	214.817	-32.740	-952579.332	-49.817	**-952629.149**	AN	1/2[106]
9. $[Fe(PyIm_2)_2]^{1-}$	5/2	-952756.244	212.387	-36.161	**-952580.018**	-48.560	-952628.578	AN	
9. $[Fe(PyIm_2)_2]^{2-}$	0	-952749.651	212.615	-33.103	-952570.139	-146.626	**-952716.765**	AN	
9. $[Fe(PyIm_2)_2]^{2-}$	2	-952755.413	220.310	-35.915	**-952571.018**	-143.464	-952714.482	AN	
10. $[Fe(PypepO)_2]^{1-}$	1/2	-984034.685	225.576	-34.001	-983843.110	-50.837	-983893.947	DMF	

10. $[Fe(PypepO)_2]^{1-}$	5/2	-984043.407	224.303	-36.071	**-983855.175**	-48.775	**-983903.950**	DMF	5/2[109]
10. $[Fe(PypepO)_2]^{2-}$	0	-984008.551	223.475	-33.572	-983818.648	-147.865	-983966.513	DMF	
10. $[Fe(PypepO)_2]^{2-}$	2	-984024.497	223.115	-36.472	**-983837.854**	-145.804	**-983983.658**	DMF	
11. $[Fe(PypepS)_2]^{1-}$	1/2	-1389378.087	223.127	-35.402	-1389190.362	-48.364	-1389238.726	DMF	1/2[110]
11. $[Fe(PypepS)_2]^{1-}$	5/2	-1389377.337	221.733	-38.050	**-1389193.654**	-46.455	**-1389240.109**	DMF	
11. $[Fe(PypepS)_2]^{2-}$	0	-1389354.084	221.234	-35.215	-1389168.065	-144.017	-1389312.082	DMF	
11. $[Fe(PypepS)_2]^{2-}$	2	-1389363.010	229.245	-37.126	**-1389170.891**	-142.205	**-1389313.096**	DMF	
12. $[Fe(PyAS)_2]^{1+}$	1/2	-1295486.279	233.311	-34.210	**-1295287.178**	-40.029	**-1295327.207**	DMF	1/2[110, 133]
12. $[Fe(PyAS)_2]^{1+}$	5/2	-1295477.182	232.616	-36.983	-1295281.549	-38.579	-1295320.128	DMF	
12. $[Fe(PyAS)_2]^{0}$	0	-1295608.267	231.975	-34.197	-1295410.489	-13.209	-1295423.698	DMF	0[133]
12. $[Fe(PyAS)_2]^{0}$	2	-1295608.193	230.346	-36.775	**-1295414.622**	-14.694	**-1295429.316**	DMF	
13. $[Fe(bpteta)]^{3+}$	1/2	-724246.458	295.060	-30.032	-723981.430	-326.365	-723981.430	AN	
13. $[Fe(bpteta)]^{3+}$	5/2	-724242.935	292.219	-32.763	**-723983.479**	-320.634	**-723983.479**	AN	
13. $[Fe(bpteta)]^{2+}$	0	-724541.049	294.192	-30.020	-724276.877	-147.422	-724276.877	AN	0[101]
13. $[Fe(bpteta)]^{2+}$	2	-724545.310	291.862	-33.322	**-724286.770**	-144.464	**-724286.770**	AN	
14. $[Fe(DITim)_2]^{1+}$	1/2	-1273854.471	323.839	-36.550	-1273567.182	-48.253	**-1273615.435**	AN	1/2[107]
14. $[Fe(DITim)_2]^{1+}$	5/2	-1273850.878	322.014	-39.134	**-1273567.998**	-47.174	**-1273615.172**	AN	
14. $[Fe(DITim)_2]^{0}$	0	-1273950.514	322.005	-36.166	-1273664.675	-23.292	-1273687.967	AN	
14. $[Fe(DITim)_2]^{0}$	2	-1273963.301	320.408	-39.309	**-1273682.202**	-22.391	**-1273704.593**	AN	
15. $[Fe(Pypep)_2]^{1+}$	1/2	-981330.407	273.960	-34.766	-981091.213	-61.860	**-981153.073**	water	1/2[114]
						-54.608	**-981145.821**	AN	
						-49.562	**-981140.775**	DMF	
15. $[Fe(Pypep)_2]^{1+}$	5/2	-981326.430	271.564	-37.806	**-981092.672**	-57.417	-981150.089	water	
						-52.086	-981144.758	AN	
						-47.535	-981140.207	DMF	
15. $[Fe(Pypep)_2]^{0}$	0	-981447.243	271.926	-34.775	-981210.092	-33.393	-981243.485	water	
						-25.058	-981235.150	AN	
						-21.777	-981231.869	DMF	
15. $[Fe(Pypep)_2]^{0}$	2	-981450.282	269.760	-38.762	**-981219.284**	-30.282	**-981249.566**	water	
						-24.491	**-981243.775**	AN	
						-21.255	**-981240.539**	DMF	
16. $[Fe(Prpep)_2]^{1+}$	1/2	-1001453.280	258.928	-34.819	-1001229.171	-65.774	-1001294.945	AN	1/2[105]
16. $[Fe(Prpep)_2]^{1+}$	5/2	-1001452.094	256.592	-37.839	**-1001233.341**	-63.712	**-1001297.053**	AN	
16. $[Fe(Prpep)_2]^{0}$	0	-1001580.729	257.152	-35.014	-1001358.591	-34.655	-1001393.246	AN	0[105]
16. $[Fe(Prpep)_2]^{0}$	2	-1001584.419	255.081	-38.769	**-1001368.107**	-33.841	**-1001401.948**	AN	
17. $[Fe(PaPy_3)(AN)]^{2+}$	1/2	-865530.091	269.432	-34.807	-865295.466	-141.658	**-865437.124**	AN	1/2[113]
17. $[Fe(PaPy_3)(AN)]^{2+}$	5/2	-865524.233	266.800	-38.234	**-865295.667**	-136.722	-865432.389	AN	
17. $[Fe(PaPy_3)(AN)]^{1+}$	0	-865727.999	268.355	-35.027	-865494.671	-47.296	-865541.967	AN	
17. $[Fe(PaPy_3)(AN)]^{1+}$	2	-865728.081	266.235	-38.490	**-865500.336**	-45.751	**-865546.087**	AN	
18. $[Fe(PaPy_3)(Cl)]^{1+}$	1/2	-1071203.949	241.645	-31.852	-1070994.156	-50.240	-1071044.396	AN	1/2[113]
18. $[Fe(PaPy_3)(Cl)]^{1+}$	5/2	-1071202.905	238.218	-36.025	**-1071000.712**	-47.917	**-1071048.629**	AN	
18. $[Fe(PaPy_3)(Cl)]^{0}$	0	-1071326.166	238.712	-32.218	-1071119.672	-23.311	-1071142.983	AN	
18. $[Fe(PaPy_3)(Cl)]^{0}$	2	-1071333.820	237.179	-35.110	**-1071131.751**	-20.481	**-1071152.232**	AN	
19. $[Fe(PaPy_3)(N_3)]^{1+}$	1/2	-885449.369	247.793	-33.300	-885234.876	-49.403	-885284.279	AN	1/2[113]
19. $[Fe(PaPy_3)(N_3)]^{1+}$	5/2	-885449.365	245.489	-36.605	**-885240.481**	-47.227	**-885287.708**	AN	
19. $[Fe(PaPy_3)(N_3)]^{0}$	0	-885570.722	246.345	-33.138	-885357.515	-20.558	-885378.073	AN	
19. $[Fe(PaPy_3)(N_3)]^{0}$	2	-885577.686	244.428	-36.660	**-885369.918**	-18.665	**-885388.583**	AN	
20. $[Fe(PaPy_3)(CN)]^{1+}$	1/2	-840654.959	245.264	-32.501	**-840442.196**	-53.572	**-840495.768**	AN	1/2[113]
20. $[Fe(PaPy_3)(CN)]^{1+}$	5/2	-840643.936	242.036	-36.179	-840438.079	-52.025	-840490.104	AN	
20. $[Fe(PaPy_3)(CN)]^{0}$	0	-840781.588	243.129	-32.402	-840570.861	-24.154	-840595.015	AN	
20. $[Fe(PaPy_3)(CN)]^{0}$	2	-840779.270	240.736	-35.425	**-840573.959**	-22.543	**-840596.502**	AN	
21. $[Fe(PaPy_2O)(Cl)]^{0}$	1/2	-1108089.012	246.809	-32.348	-1107874.551	-19.690	-1107894.241	DMF	
21. $[Fe(PaPy_2O)(Cl)]^{0}$	5/2	-1108098.254	240.111	-35.572	**-1107893.715**	-18.363	**-1107912.078**	DMF	5/2[109]

21. $[Fe(PaPy_2O)(Cl)]^{1-}$	0	-1108132.440	239.863	-33.238	-1107925.815	-58.4354	-1107984.250	DMF	
21. $[Fe(PaPy_2O)(Cl)]^{1-}$	2	-1108145.595	239.343	-36.784	**-1107943.036**	-58.727	**-1108001.763**	DMF	
22. $[Fe(SEt)_4]^{1-}$	1/2	-1276018.976	169.214	-33.884	-1275883.646	-44.297	-1275927.943	AN	
22. $[Fe(SEt)_4]^{1-}$	5/2	-1276051.332	169.420	-37.572	**-1275919.484**	-44.330	**-1275963.813**	AN	
22. $[Fe(SEt)_4]^{2-}$	1	-1275996.296	168.103	-35.347	-1275863.540	-157.088	-1276020.628	AN	
22. $[Fe(SEt)_4]^{2-}$	2	-1276019.346	168.105	-36.707	**-1275887.948**	-157.843	**-1276045.791**	AN	
23. $[Fe(S_2\text{-o-xyl})_2]^{1-}$	1/2	-1465844.032	175.797	-31.929	-1465700.164	-46.8206	-1465746.985	DMF	
23. $[Fe(S_2\text{-o-xyl})_2]^{1-}$	5/2	-1465872.805	175.801	-34.020	**-1465731.024**	-45.682	**-1465776.706**	DMF	5/2[119]
23. $[Fe(S_2\text{-o-xyl})_2]^{2-}$	1	-1465826.482	174.325	-33.041	-1465685.198	-156.083	-1465841.281	DMF	
23. $[Fe(S_2\text{-o-xyl})_2]^{2-}$	2	-1465850.651	174.335	-32.838	**-1465709.154**	-151.448	**-1465860.602**	DMF	2[119]
24. $[Fe(SEtOH)_4]^{1-}$	1/2	-1464864.505	199.247	-31.109	-1464696.367	-67.591	-1464763.958	water	
24. $[Fe(SEtOH)_4]^{1-}$	5/2	-1464902.965	181.058	-37.270	**-1464759.177**	-71.019	**-1464830.196**	water	
24. $[Fe(SEtOH)_4]^{2-}$	1	-1464846.134	179.726	-36.807	-1464703.215	-193.297	-1464896.512	water	
24. $[Fe(SEtOH)_4]^{2-}$	2	-1464882.736	184.696	-36.953	**-1464734.993**	-192.200	**-1464927.193**	water	2[116]
25. $[Mn(CN)_6]^{3-}$	1	-414871.350	29.926	-25.536	**-414866.960**	-387.796	**-415254.756**	water	
25. $[Mn(CN)_6]^{3-}$	2	-414865.626	27.734	-28.103	-414865.995	-378.415	-415244.410	water	
25. $[Mn(CN)_6]^{4-}$	1/2	-414674.424	29.143	-25.527	-414670.808	-684.755	-415355.563	water	1/2[134]
25. $[Mn(CN)_6]^{4-}$	5/2	-414705.714	24.959	-31.493	**-414712.248**	-644.859	**-415357.107**	water	
26. $[Mn(bpia)(Cl)_2]^{2+}$	3/2	-1228186.353	219.534	-32.223	-1227999.042	-148.358	-1228147.400	AN	
26. $[Mn(bpia)(Cl)_2]^{1+}$	1	-1228407.145	219.410	-32.187	-1228219.922	-53.875	-1228273.797	AN	
26. $[Mn(bpia)(Cl)_2]^{1+}$	2	-1228421.990	218.100	-33.268	**-1228237.158**	-51.7645	**-1228288.923**	AN	
26. $[Mn(bpia)(Cl)_2]^{0}$	1/2	-1228513.396	217.959	-33.104	-1228328.541	-30.876	-1228359.417	AN	
26. $[Mn(bpia)(Cl)_2]^{0}$	5/2	-1228557.770	216.463	-35.572	**-1228376.879**	-32.4007	**-1228409.280**	AN	
27. $[Mn(py_2(NMe)_2Cl_2)]^{2+}$	3/2	-1170269.815	220.361	-30.417	-1170079.871	-155.520	-1170235.391	AN	
27. $[Mn(py_2(NMe)_2Cl_2)]^{1+}$	1	-1170496.916	220.552	-30.508	-1170306.872	-55.1719	-1170362.044	AN	
27. $[Mn(py_2(NMe)_2Cl_2)]^{1+}$	2	-1170512.529	219.020	-32.461	**-1170325.970**	-51.597	**-1170377.567**	AN	
27. $[Mn(py_2(NMe)_2Cl_2)]^{0}$	1/2	-1170610.881	218.848	-30.563	-1170422.596	-28.761	-1170451.357	AN	
27. $[Mn(py_2(NMe)_2Cl_2)]^{0}$	5/2	-1170647.503	217.150	-31.728	**-1170462.081**	-30.344	**-1170492.425**	AN	5/2[100]
28. $[Mn(bpteta)]^{3+}$	1	-711989.878	293.769	-31.137	-711727.246	-321.128	-712048.374	AN	
28. $[Mn(bpteta)]^{3+}$	2	-712002.500	292.402	-32.070	**-711742.168**	-318.342	**-712060.510**	AN	
28. $[Mn(bpteta)]^{2+}$	1/2	-712265.800	299.437	-30.857	-711997.220	-144.365	-712141.585	AN	
28. $[Mn(bpteta)]^{2+}$	5/2	-712312.470	291.438	-33.603	**-712054.635**	-143.191	**-712197.826**	AN	5/2[101]
29. $[Ni(bpy)_3]^{3+}$	1/2	-1038563.008	304.240	-37.504	**-1038296.272**	-288.437	**-1038584.709**	water	
29. $[Ni(bpy)_3]^{3+}$	3/2	-1038551.766	301.911	-39.759	-1038289.614	-283.266	-1038572.880	water	
29. $[Ni(bpy)_3]^{2+}$	1	-1038864.354	303.452	-39.419	**-1038600.321**	-129.731	**-1038730.052**	water	
30. $[Ni(bpteta)]^{3+}$	1/2	-753011.608	293.570	-30.746	**-752748.784**	-325.745	**-753074.529**	AN	
30. $[Ni(bpteta)]^{3+}$	3/2	-752989.893	290.270	-32.711	-752732.334	-315.733	-753048.067	AN	
30. $[Ni(bpteta)]^{2+}$	1	-753328.570	292.715	-31.884	**-753067.739**	-146.632	**-753214.371**	AN	1[101]
prediction set (31-48)									
31. $[Fe(cyclamAc)(N_3)]^{2+}$	1	-708608.426	256.999	-30.144	**-708381.571**	-167.315	**-708548.886**	AN	
31. $[Fe(cyclamAc)(N_3)]^{2+}$	2	-708594.663	254.637	-31.438	-708371.464	-164.125	-708535.589	AN	
31. $[Fe(cyclamAc)(N_3)]^{1+}$	1/2	-708858.059	258.049	-29.033	**-708629.043**	-56.615	**-708685.658**	AN	1/2[145]
31. $[Fe(cyclamAc)(N_3)]^{1+}$	5/2	-708851.032	254.887	-32.128	-708628.273	-55.866	-708684.139	AN	
31. $[Fe(cyclamAc)(N_3)]^{0}$	0	-708986.011	255.994	-29.580	-708759.597	-19.81	-708779.407	AN	0[145]
31. $[Fe(cyclamAc)(N_3)]^{0}$	2	-708988.617	253.401	-32.101	**-708767.317**	-19.42	**-708786.737**	AN	
32. $[Ni(tacn)]^{3+}$	1/2	-610224.978	288.387	-26.556	**-609963.147**	-369.019	**-610332.166**	water	1/2[117]
32. $[Ni(tacn)]^{3+}$	3/2	-610197.786	286.921	-27.130	-609937.995	-365.770	-610303.765	water	
32. $[Ni(tacn)]^{2+}$	1	-610553.804	287.768	-26.884	**-610292.920**	-164.730	**-610457.650**	water	

33. [Fe(dtne)]$^{3+}$	1/2	-630034.536	311.814	-26.751	-629749.473	-360.350	-630109.823	water	1/2[146]
33. [Fe(dtne)]$^{3+}$	5/2	-630038.718	309.483	-28.544	**-629757.779**	-355.150	**-630112.929**	water	
33. [Fe(dtne)]$^{2+}$	0	-630337.880	311.429	-26.361	-630052.812	-161.440	-630214.252	water	
33. [Fe(dtne)]$^{2+}$	2	-630348.101	309.403	-28.568	**-630067.266**	-158.680	**-630225.946**	water	
34. [Ni(dtne)]$^{3+}$	1/2	-658802.115	310.472	-26.724	**-658518.367**	-360.034	**-658878.401**	water	
34. [Ni(dtne)]$^{3+}$	3/2	-658795.660	308.583	-28.766	-658515.843	-353.310	-658869.153	water	
34. [Ni(dtne)]$^{2+}$	1	-659130.706	310.039	-27.836	**-658848.503**	-160.050	**-659008.553**	water	1[146]
35. [Fe(terpy)$_2$]$^{3+}$	1/2	-1009049.626	292.794	-35.324	-1008792.156	-270.350	-1009062.506	DMF	
35. [Fe(terpy)$_2$]$^{3+}$	5/2	-1009049.174	289.712	-38.248	**-1008797.710**	-266.800	**-1009064.510**	DMF	
35. [Fe(terpy)$_2$]$^{2+}$	0	-1009329.952	291.313	-35.138	-1009073.777	-121.530	-1009195.307	DMF	
35. [Fe(terpy)$_2$]$^{2+}$	2	-1009327.792	289.157	-39.560	**-1009078.195**	-119.250	**-1009197.445**	DMF	
36. [Fe(SPh)$_4$]$^{1-}$	1/2	-1658709.869	231.134	-41.931	-1658520.666	-43.71	-1658564.376	AN	
36. [Fe(SPh)$_4$]$^{1-}$	5/2	-1658734.844	230.484	-43.501	**-1658547.861**	-43.35	**-1658591.211**	AN	
36. [Fe(SPh)$_4$]$^{2-}$	1	-1658709.779	229.890	-43.341	-1658523.230	-137.39	-1658660.620	AN	
36. [Fe(SPh)$_4$]$^{2-}$	2	-1658734.611	228.975	-43.463	**-1658549.099**	-137.68	**-1658686.779**	AN	
37. [Fe(SCH$_2$CON(CH$_3$)$_2$)$_4$]$^{1-}$	1/2	-1798273.998	310.343	-49.167	-1798012.822	-50.733 -49.403	-1798063.555 -1798062.225	AN DMF	
37. [Fe(SCH$_2$CON(CH$_3$)$_2$)$_4$]$^{1-}$	5/2	-1798298.673	308.613	-51.020	**-1798041.080**	-48.978 -47.594	**-1798090.058** **-1798088.674**	AN DMF	
37. [Fe(SCH$_2$CON(CH$_3$)$_2$)$_4$]$^{2-}$	1	-1798271.321	307.615	-48.501	-1798012.207	-139.201 -137.523	-1798151.408 -1798149.730	AN DMF	
37. [Fe(SCH$_2$CON(CH$_3$)$_2$)$_4$]$^{2-}$	2	-1798300.324	307.096	-51.149	**-1798044.377**	-138.455 -136.441	**-1798182.832** **-1798180.818**	AN DMF	
38. [Fe(S-*i*-Pr)$_4$]$^{1-}$	1/2	-1374728.655	239.758	-37.753	-1374526.650	-42.840	-1374569.490	AN	
38. [Fe(S-*i*-Pr)$_4$]$^{1-}$	5/2	-1374762.231	239.880	-41.083	**-1374563.434**	-42.818	**-1374606.252**	AN	
38. [Fe(S-*i*-Pr)$_4$]$^{2-}$	1	-1374710.039	283.404	-37.741	-1374464.376	-149.016	-1374613.392	AN	
38. [Fe(S-*i*-Pr)$_4$]$^{2-}$	2	-1374733.302	238.400	-41.235	**-1374536.137**	-149.880	**-1374686.017**	AN	
39. [Fe(bpy)(CN)$_4$]$^{3+}$	1/2	-621596.265	122.642	-30.131	**-621503.754**	-82.749	**-621586.503**	water	
39. [Fe(bpy)(CN)$_4$]$^{3+}$	5/2	-621578.950	119.485	-33.825	-621493.290	-74.900	-621568.190	water	
39. [Fe(bpy)(CN)$_4$]$^{2+}$	0	-621584.615	120.810	-29.920	**-621493.725**	-200.870	**-621694.595**	water	
39. [Fe(bpy)(CN)$_4$]$^{2+}$	2	-621570.135	117.277	-33.969	-621486.827	-199.798	-621686.625	water	
40. [Fe(tacnPy$_2$)(AN)]$^{3+}$	1/2	-796740.909	316.283	-32.647	-796457.273	-305.298	**-796762.571**	AN	
40. [Fe(tacnPy$_2$)(AN)]$^{3+}$	5/2	-796736.168	312.923	-35.395	**-796458.640**	-300.986	-796759.626	AN	
40. [Fe(tacnPy$_2$)(AN)]$^{2+}$	0	-797031.002	315.147	-32.498	-796748.353	-136.942	-796885.295	AN	0[112]
40. [Fe(tacnPy$_2$)(AN)]$^{2+}$	2	-797031.058	312.777	-35.856	**-796754.137**	-134.462	**-796888.599**	AN	
41. [Fe(N$_4$Py)(Cl)]$^{2+}$	1/2	-1095911.753	263.119	-31.984	-1095680.618	-139.806	-1095820.424	AN	
41. [Fe(N$_4$Py)(Cl)]$^{2+}$	5/2	-1095916.943	254.981	-35.177	**-1095697.139**	-136.901	**-1095834.040**	AN	
41. [Fe(N$_4$Py)(Cl)]$^{1+}$	0	-1096116.448	263.805	-31.955	-1095884.598	-47.711	-1095932.309	AN	
41. [Fe(N$_4$Py)(Cl)]$^{1+}$	2	-1096125.618	253.814	-35.027	**-1095906.831**	-45.560	**-1095952.391**	AN	2[111]
42. [Fe(N$_4$Py)(AN)]$^{3+}$	1/2	-890176.791	285.342	-34.723	**-889926.172**	-290.331	**-890216.503**	AN	
42. [Fe(N$_4$Py)(AN)]$^{3+}$	5/2	-890169.393	283.102	-37.522	-889923.813	-286.482	-890210.295	AN	
42. [Fe(N$_4$Py)(AN)]$^{2+}$	0	-890460.110	285.053	-34.770	-890209.827	-131.806	-890341.633	AN	0[111]
42. [Fe(N$_4$Py)(AN)]$^{2+}$	2	-890458.938	282.981	-38.420	**-890214.377**	-129.527	**-890343.904**	AN	
43. [Fe(Py$_3$tacn)]$^{3+}$	1/2	-868512.672	330.931	-31.619	-868213.360	-302.070	**-868515.430**	AN	1/2[118]
43. [Fe(Py$_3$tacn)]$^{3+}$	5/2	-868510.439	328.415	-34.048	**-868216.072**	-296.883	-868512.955	AN	
43. [Fe(Py$_3$tacn)]$^{2+}$	0	-868796.724	329.952	-31.562	-868498.334	-138.213	-868636.547	AN	0[118]
43. [Fe(Py$_3$tacn)]$^{2+}$	2	-868797.617	327.501	-34.046	**-868504.162**	-132.977	**-868637.139**	AN	
44. [Ni(Py$_3$tacn)]$^{3+}$	1/2	-897274.859	329.336	-31.652	**-896977.175**	-301.650	**-897278.825**	AN	
44. [Ni(Py$_3$tacn)]$^{3+}$	3/2	-897253.457	326.769	-33.588	-896960.276	-301.902	-897262.178	AN	
44. [Ni(Py$_3$tacn)]$^{2+}$	1	-897583.446	328.598	-33.393	**-897288.241**	-134.151	**-897422.392**	AN	1[118]
45. [Fe(TCTA)]^{0}	1/2	-757757.241	202.766	-27.715	-757582.190	-39.970	-757622.160	water	
45. [Fe(TCTA)]^{0}	5/2	-757776.720	201.044	-29.647	**-757605.323**	-34.433	**-757639.756**	water	5/2[147]

45. $[Fe(TCTA)]^{1-}$	0	-757815.821	200.640	-27.944	-757643.125	-76.677	-757719.802	water	
45. $[Fe(TCTA)]^{1-}$	2	-757839.808	199.236	-30.576	**-757671.148**	-72.485	**-757743.633**	water	2[147]
46. $[Ni(TCTA)]^{0}$	1/2	-786518.253	201.245	-28.526	**-786345.534**	-38.962	**-786384.496**	water	
46. $[Ni(TCTA)]^{0}$	3/2	-786508.093	200.216	-29.439	-786337.316	-36.759	-786374.075	water	
46. $[Ni(TCTA)]^{1-}$	1	-786614.039	199.778	-29.499	**-786443.760**	-75.129	**-786518.889**	water	1[147]
47. $[Mn(TCTA)]^{0}$	1	-745516.936	211.350	-27.708	-745333.294	-39.943	-745373.237	water	
47. $[Mn(TCTA)]^{0}$	2	-745528.989	200.810	-29.728	**-745357.907**	-36.229	**-745394.136**	water	2[147]
47. $[Mn(TCTA)]^{1-}$	1/2	-745558.854	203.770	-28.170	-745383.254	-77.145	-745460.399	water	
47. $[Mn(TCTA)]^{1-}$	5/2	-745613.124	199.114	-30.830	**-745444.840**	-71.304	**-745516.144**	water	5/2[147]
48. $[Fe(Pyr_2Py)_2]^{3+}$	1/2	-916378.941	334.302	-35.282	-916079.921	-286.304	**-916366.225**	AN	
48. $[Fe(Pyr_2Py)_2]^{3+}$	5/2	-916376.445	331.648	-38.289	**-916083.086**	-281.656	-916364.742	AN	
48. $[Fe(Pyr_2Py)_2]^{2+}$	0	-916661.059	333.723	-35.524	-916362.860	-127.524	-916490.384	AN	0[148]
48. $[Fe(Pyr_2Py)_2]^{2+}$	2	-916660.255	331.708	-38.881	**-916367.428**	-125.153	**-916492.581**	AN	

Table A4. Energy terms calculated using B3LYP*[21] functional.

model compound	spin theory	E_{0K}^{B3LYP*}	ZPE	$\Delta G_{0\rightarrow298K}$	G_g^0	ΔG_{sol}^0	G_s^0	solvent	spin exp.
		kcal·mol^{-1}							
4. $[Fe(phen)_3]^{3+}$	1/2	-1152639.701	329.319	-36.195	**-1152346.577**	-278.196	**-1152624.773**	water	
						-267.357	**-1152613.934**	AN	
						-258.528	**-1152605.105**	DMF	
4. $[Fe(phen)_3]^{3+}$	5/2	-1152629.493	327.512	-38.780	-1152340.761	-274.108	-1152614.869	water	
						-263.819	-1152604.580	AN	
						-255.380	-1152596.141	DMF	
4. $[Fe(phen)_3]^{2+}$	0	-1152910.281	328.962	-36.295	**-1152617.614**	-126.922	**-1152744.536**	water	0[131]
						-121.599	**-1152739.213**	AN	
						-116.153	**-1152733.767**	DMF	
4. $[Fe(phen)_3]^{2+}$	2	-1152903.371	326.959	-40.942	-1152617.354	-124.844	-1152742.198	water	
						-119.708	-1152737.062	AN	
						-114.449	-1152731.803	DMF	
6. $[Fe(sar)]^{3+}$	1/2	-629683.527	315.435	-26.143	-629394.235	-362.269	**-629756.504**	water	1/2[157]
6. $[Fe(sar)]^{3+}$	5/2	-629675.859	309.353	-29.469	**-629395.975**	-356.576	-629752.551	water	
6. $[Fe(sar)]^{2+}$	0	-629978.950	311.811	-26.271	-629693.410	-163.102	**-629856.512**	water	0[132]
6. $[Fe(sar)]^{2+}$	2	-629975.246	309.353	-29.469	**-629695.362**	-160.454	-629855.816	water	
10. $[Fe(PypepO)_2]^{1-}$	1/2	-983467.879	225.576	-34.001	-983276.304	-50.837	-983327.141	DMF	
10. $[Fe(PypepO)_2]^{1-}$	5/2	-983472.053	224.303	-36.071	**-983283.821**	-48.775	**-983332.596**	DMF	5/2[109]
10. $[Fe(PypepO)_2]^{2-}$	0	-983442.261	223.475	-33.572	-983252.358	-147.865	-983400.223	DMF	
10. $[Fe(PypepO)_2]^{2-}$	2	-983452.145	223.115	-36.472	**-983265.502**	-145.804	**-983411.306**	DMF	
11. $[Fe(PypepS)_2]^{1-}$	1/2	-1388736.116	223.127	-35.402	**-1388548.391**	-48.364	**-1388596.755**	DMF	1/2[110]
11. $[Fe(PypepS)_2]^{1-}$	5/2	-1388730.741	221.733	-38.050	-1388547.058	-46.455	-1388593.513	DMF	
11. $[Fe(PypepS)_2]^{2-}$	0	-1388710.976	221.234	-35.215	**-1388524.957**	-144.017	**-1388668.974**	DMF	
11. $[Fe(PypepS)_2]^{2-}$	2	-1388714.330	229.245	-37.126	-1388522.211	-142.205	-1388664.416	DMF	
12. $[Fe(PyAS)_2]^{1+}$	1/2	-1294887.804	233.311	-34.210	**-1294688.703**	-40.029	**-1294728.732**	DMF	1/2[110, 133]
12. $[Fe(PyAS)_2]^{1+}$	5/2	-1294872.087	232.616	-36.983	-1294676.454	-38.579	-1294715.033	DMF	
12. $[Fe(PyAS)_2]^{0}$	0	-1295009.190	231.975	-34.197	**-1294811.412**	-13.209	**-1294824.621**	DMF	0[133]
12. $[Fe(PyAS)_2]^{0}$	2	-1295002.400	230.346	-36.775	-1294808.829	-14.694	-1294823.523	DMF	
13. $[Fe(bpteta)]^{3+}$	1/2	-723807.718	294.967	-30.055	**-723542.806**	-326.462	**-723869.268**	AN	
13. $[Fe(bpteta)]^{3+}$	5/2	-723799.955	292.202	-32.686	-723540.439	-320.844	-723861.283	AN	
13. $[Fe(bpteta)]^{2+}$	0	-724100.637	294.258	-29.925	-723836.304	-147.516	-723983.820	AN	0[101]
13. $[Fe(bpteta)]^{2+}$	2	-724099.836	291.992	-33.080	**-723840.924**	-144.220	**-723985.144**	AN	
16. $[Fe(Prpep)_2]^{1+}$	1/2	-1000876.945	258.928	-34.819	**-1000652.836**	-65.774	**-1000718.610**	AN	1/2[105]
16. $[Fe(Prpep)_2]^{1+}$	5/2	-1000870.821	256.592	-37.839	-1000652.068	-63.712	-1000715.780	AN	
16. $[Fe(Prpep)_2]^{0}$	0	-1001003.055	257.152	-35.014	-1000780.917	-34.655	-1000815.572	AN	0[105]
16. $[Fe(Prpep)_2]^{0}$	2	-1001001.074	255.081	-38.769	**-1000784.762**	-33.841	**-1000818.603**	AN	
18. $[Fe(PaPy_3)(Cl)]^{1+}$	1/2	-1070675.861	241.645	-31.852	-1070466.068	-50.240	**-1070516.308**	AN	1/2[113]
18. $[Fe(PaPy_3)(Cl)]^{1+}$	5/2	-1070670.307	238.218	-36.025	**-1070468.114**	-47.917	-1070516.031	AN	
18. $[Fe(PaPy_3)(Cl)]^{0}$	0	-1070797.059	238.712	-32.218	-1070590.565	-23.311	-1070613.876	AN	
18. $[Fe(PaPy_3)(Cl)]^{0}$	2	-1070798.734	237.179	-35.110	**-1070596.665**	-20.481	**-1070617.146**	AN	
19. $[Fe(PaPy_3)(N_3)]^{1+}$	1/2	-884932.935	247.793	-33.300	-884718.442	-49.403	**-884767.845**	AN	1/2[113]
19. $[Fe(PaPy_3)(N_3)]^{1+}$	5/2	-884928.440	245.489	-36.605	**-884719.556**	-47.227	-884766.783	AN	
19. $[Fe(PaPy_3)(N_3)]^{0}$	0	-885053.438	246.345	-33.138	-884840.231	-20.558	-884860.789	AN	
19. $[Fe(PaPy_3)(N_3)]^{0}$	2	-885054.511	244.428	-36.660	**-884846.743**	-18.665	**-884865.408**	AN	
25. $[Mn(CN)_6]^{3-}$	1	-414651.498	29.926	-25.536	**-414647.108**	-387.796	**-415034.904**	water	
25. $[Mn(CN)_6]^{3-}$	2	-414641.871	27.734	-28.103	-414642.240	-378.415	-415020.655	water	
25. $[Mn(CN)_6]^{4-}$	1/2	-414454.931	29.143	-25.527	-414451.315	-684.755	**-415136.070**	water	1/2[134]
25. $[Mn(CN)_6]^{4-}$	5/2	-414476.707	24.959	-31.493	**-414483.241**	-644.859	-415128.100	water	

Table A5. Energy terms calculated using X3LYP[125] functional.

model compound	spin theory	E_{0K}^{X3LYP}	ZPE	$\Delta G_{0\rightarrow 298K}$	G_g^0	ΔG_{sol}^0	G_s^0	solvent	spin exp.
		kcal·mol^{-1}							
4. [Fe(phen)$_3$]$^{3+}$	1/2	-1152828.312	329.319	-36.195	**-1152535.188**	-278.196	**-1152813.384**	water	
						-267.357	**-1152802.545**	AN	
						-258.528	**-1152793.716**	DMF	
4. [Fe(phen)$_3$]$^{3+}$	5/2	-1152824.370	327.512	-38.780	**-1152535.638**	-274.108	-1152809.746	water	
						-263.819	-1152799.457	AN	
						-255.380	-1152791.018	DMF	
4. [Fe(phen)$_3$]$^{2+}$	0	-1153098.949	328.962	-36.295	-1152806.282	-126.922	-1152933.204	water	0[131]
						-121.599	-1152927.881	AN	
						-116.153	-1152922.435	DMF	
4. [Fe(phen)$_3$]$^{2+}$	2	-1153099.334	326.959	-40.942	**-1152813.317**	-124.844	**-1152938.161**	water	
						-119.708	**-1152933.025**	AN	
						-114.449	**-1152927.766**	DMF	
6. [Fe(sar)]$^{3+}$	1/2	-629764.907	315.435	-26.143	-629475.615	-362.269	-629837.884	water	1/2[157]
6. [Fe(sar)]$^{3+}$	5/2	-629762.082	309.353	-29.469	**-629482.198**	-356.576	**-629838.774**	water	
6. [Fe(sar)]$^{2+}$	0	-630061.939	311.811	-26.271	-629776.399	-163.102	-629939.501	water	0 [132]
6. [Fe(sar)]$^{2+}$	2	-630063.731	309.353	-29.469	**-629783.847**	-160.454	**-629944.301**	water	
10. [Fe(PypepO)$_2$]$^{1-}$	1/2	-983612.177	225.576	-34.001	-983420.602	-50.837	-983471.439	DMF	
10. [Fe(PypepO)$_2$]$^{1-}$	5/2	-983622.383	224.303	-36.071	**-983434.151**	-48.775	**-983482.926**	DMF	5/2[109]
10. [Fe(PypepO)$_2$]$^{2-}$	0	-983584.992	223.475	-33.572	-983395.089	-147.865	-983542.954	DMF	
10. [Fe(PypepO)$_2$]$^{2-}$	2	-983602.690	223.115	-36.472	**-983416.047**	-145.804	**-983561.851**	DMF	
11. [Fe(PypepS)$_2$]$^{1-}$	1/2	-1388928.270	223.127	-35.402	-1388740.545	-48.364	-1388788.909	DMF	1/2[110]
11. [Fe(PypepS)$_2$]$^{1-}$	5/2	-1388928.897	221.733	-38.050	**-1388745.214**	-46.455	**-1388791.669**	DMF	
11. [Fe(PypepS)$_2$]$^{2-}$	0	-1388903.273	221.234	-35.215	-1388717.254	-144.017	-1388861.271	DMF	
11. [Fe(PypepS)$_2$]$^{2-}$	2	-1388913.800	229.245	-37.126	**-1388721.681**	-142.205	**-1388863.886**	DMF	
12. [Fe(PyAS)$_2$]$^{1+}$	1/2	-1295064.469	233.311	-34.210	**-1294865.368**	-40.029	**-1294905.397**	DMF	1/2[110, 133]
12. [Fe(PyAS)$_2$]$^{1+}$	5/2	-1295056.854	232.616	-36.983	-1294861.221	-38.579	-1294899.800	DMF	
12. [Fe(PyAS)$_2$]^{0}	0	-1295185.522	231.975	-34.197	-1294987.744	-13.209	-1295000.953	DMF	0[133]
12. [Fe(PyAS)$_2$]^{0}	2	-1295187.096	230.346	-36.775	**-1294993.525**	-14.694	**-1295008.219**	DMF	
13. [Fe(bpteta)]$^{3+}$	1/2	-723908.905	294.967	-30.055	-723643.993	-326.462	**-723970.455**	AN	
13. [Fe(bpteta)]$^{3+}$	5/2	-723906.593	292.202	-32.686	**-723647.077**	-320.844	-723967.921	AN	
13. [Fe(bpteta)]$^{2+}$	0	-724202.636	294.258	-29.925	-723938.303	-147.516	-724085.819	AN	0[101]
13. [Fe(bpteta)]$^{2+}$	2	-724208.055	291.992	-33.080	**-723949.143**	-144.220	**-724093.363**	AN	
16. [Fe(Prpep)$_2$]$^{1+}$	1/2	-1001023.633	258.928	-34.819	-1000799.524	-65.774	-1000865.298	AN	1/2[105]
16. [Fe(Prpep)$_2$]$^{1+}$	5/2	-1001023.653	256.592	-37.839	**-1000804.900**	-63.712	**-1000868.612**	AN	
16. [Fe(Prpep)$_2$]^{0}	0	-1001150.145	257.152	-35.014	-1000928.007	-34.655	-1000962.662	AN	0[105]
16. [Fe(Prpep)$_2$]^{0}	2	-1001154.852	255.081	-38.769	**-1000938.540**	-33.841	**-1000972.381**	AN	
18. [Fe(PaPy$_3$)(Cl)]$^{1+}$	1/2	-1070822.305	241.645	-31.852	-1070612.512	-50.240	-1070662.752	AN	1/2[113]
18. [Fe(PaPy$_3$)(Cl)]$^{1+}$	5/2	-1070822.445	238.218	-36.025	**-1070620.252**	-47.917	**-1070668.169**	AN	
18. [Fe(PaPy$_3$)(Cl)]^{0}	0	-1070943.738	238.712	-32.218	-1070737.244	-23.311	-1070760.555	AN	
18. [Fe(PaPy$_3$)(Cl)]^{0}	2	-1070952.528	237.179	-35.110	**-1070750.459**	-20.481	**-1070770.940**	AN	
19. [Fe(PaPy$_3$)(N$_3$)]$^{1+}$	1/2	-885058.930	247.793	-33.300	-884844.437	-49.403	-884893.840	AN	1/2[113]
19. [Fe(PaPy$_3$)(N$_3$)]$^{1+}$	5/2	-885060.009	245.489	-36.605	**-884851.125**	-47.227	**-884898.352**	AN	
19. [Fe(PaPy$_3$)(N$_3$)]^{0}	0	-885179.519	246.345	-33.138	-884966.312	-20.558	-884986.870	AN	
19. [Fe(PaPy$_3$)(N$_3$)]^{0}	2	-885187.498	244.428	-36.660	**-884979.730**	-18.665	**-884998.395**	AN	
25. [Mn(CN)$_6$]$^{3-}$	1	-414696.245	29.926	-25.536	**-414691.855**	-387.796	**-415079.651**	water	
25. [Mn(CN)$_6$]$^{3-}$	2	-414691.292	27.734	-28.103	-414691.661	-378.415	-415070.076	water	
25. [Mn(CN)$_6$]$^{4-}$	1/2	-414498.124	29.143	-25.527	-414494.508	-684.755	-415179.263	water	1/2[134]
25. [Mn(CN)$_6$]$^{4-}$	5/2	-414530.959	24.959	-31.493	**-414537.493**	-644.859	**-415182.352**	water	

Table A6. Energy terms calculated using B4LYP functional with â$_0$=0.15

model compound	spin theory	E_{0K}^{B4LYP}	ZPE	$\Delta G_{0\rightarrow 298K}$	G_g^0	ΔG_{sol}^0	G_s^0	solvent	spin exp.
		kcal·mol^{-1}							
4. [Fe(phen)$_3$]$^{3+}$	1/2	-1145629.054	329.319	-36.195	**-1145335.930**	-278.196	**-1145614.126**	water	
						-267.357	**-1145603.287**	AN	
						-258.528	**-1145594.458**	DMF	
4. [Fe(phen)$_3$]$^{3+}$	5/2	-1145617.817	327.512	-38.780	-1145329.085	-274.108	-1145603.193	water	
						-263.819	-1145592.904	AN	
						-255.380	-1145584.465	DMF	
4. [Fe(phen)$_3$]$^{2+}$	0	-1145890.388	328.962	-36.295	**-1145597.721**	-126.922	**-1145724.643**	water	0[131]
						-121.599	**-1145719.320**	AN	
						-116.153	**-1145713.874**	DMF	
4. [Fe(phen)$_3$]$^{2+}$	2	-1145882.812	326.959	-40.942	-1145596.795	-124.844	-1145721.639	water	
						-119.708	-1145716.503	AN	
						-114.449	-1145711.244	DMF	
6. [Fe(sar)]$^{3+}$	1/2	-625826.988	315.435	-26.143	-625537.696	-362.269	**-625899.965**	water	1/2[157]
6. [Fe(sar)]$^{3+}$	5/2	-625818.302	309.353	-29.469	**-625538.418**	-356.576	-625894.994	water	
6. [Fe(sar)]$^{2+}$	0	-626112.219	311.811	-26.271	-625826.679	-163.102	**-625989.781**	water	0[132]
6. [Fe(sar)]$^{2+}$	2	-626107.704	309.353	-29.469	**-625827.820**	-160.454	-625988.274	water	
10. [Fe(PypepO)$_2$]$^{1-}$	1/2	-977708.764	225.576	-34.001	-977517.189	-50.837	-977568.026	DMF	
10. [Fe(PypepO)$_2$]$^{1-}$	5/2	-977711.466	224.303	-36.071	**-977523.234**	-48.775	**-977572.009**	DMF	5/2[109]
10. [Fe(PypepO)$_2$]$^{2-}$	0	-977677.618	223.475	-33.572	-977487.715	-147.865	-977635.580	DMF	
10. [Fe(PypepO)$_2$]$^{2-}$	2	-977686.135	223.115	-36.472	**-977499.492**	-145.804	**-977645.296**	DMF	
11. [Fe(PypepS)$_2$]$^{1-}$	1/2	-1382000.138	223.127	-35.402	**-1381812.413**	-48.364	**-1381860.777**	DMF	1/2[110]
11. [Fe(PypepS)$_2$]$^{1-}$	5/2	-1381993.897	221.733	-38.050	-1381810.214	-46.455	-1381856.669	DMF	
11. [Fe(PypepS)$_2$]$^{2-}$	0	-1381970.303	221.234	-35.215	**-1381784.284**	-144.017	**-1381928.301**	DMF	
11. [Fe(PypepS)$_2$]$^{2-}$	2	-1381972.224	229.245	-37.126	-1381780.105	-142.205	-1381922.310	DMF	
12. [Fe(PyAS)$_2$]$^{1+}$	1/2	-1288616.891	233.311	-34.210	**-1288417.790**	-40.029	**-1288457.819**	DMF	1/2[110, 133]
12. [Fe(PyAS)$_2$]$^{1+}$	5/2	-1288601.234	232.616	-36.983	-1288405.601	-38.579	-1288444.180	DMF	
12. [Fe(PyAS)$_2$]^{0}	0	-1288731.522	231.975	-34.197	**-1288533.744**	-13.209	**-1288546.953**	DMF	0[133]
12. [Fe(PyAS)$_2$]^{0}	2	-1288724.323	230.346	-36.775	-1288530.752	-14.694	-1288545.446	DMF	
13. [Fe(bpteta)]$^{3+}$	1/2	-719405.451	294.967	-30.055	**-719140.539**	-326.462	**-719467.001**	AN	
13. [Fe(bpteta)]$^{3+}$	5/2	-719396.768	292.202	-32.686	-719137.252	-320.844	-719458.096	AN	
13. [Fe(bpteta)]$^{2+}$	0	-719688.161	294.258	-29.925	-719423.828	-147.516	-719571.344	AN	0[101]
13. [Fe(bpteta)]$^{2+}$	2	-719686.677	291.992	-33.080	**-719427.765**	-144.220	**-719571.985**	AN	
16. [Fe(Prpep)$_2$]$^{1+}$	1/2	-995017.914	258.928	-34.819	**-994793.805**	-65.774	**-994859.579**	AN	1/2[105]
16. [Fe(Prpep)$_2$]$^{1+}$	5/2	-995011.394	256.592	-37.839	-994792.641	-63.712	-994856.353	AN	
16. [Fe(Prpep)$_2$]^{0}	0	-995136.269	257.152	-35.014	-994914.131	-34.655	-994948.786	AN	0[105]
16. [Fe(Prpep)$_2$]^{0}	2	-995134.257	255.081	-38.769	**-994917.945**	-33.841	**-994951.786**	AN	
18. [Fe(PaPy$_3$)(Cl)]$^{1+}$	1/2	-1065199.184	241.645	-31.852	-1064989.391	-50.240	**-1065039.631**	AN	1/2[113]
18. [Fe(PaPy$_3$)(Cl)]$^{1+}$	5/2	-1065193.065	238.218	-36.025	**-1064990.872**	-47.917	-1065038.789	AN	
18. [Fe(PaPy$_3$)(Cl)]^{0}	0	-1065312.879	238.712	-32.218	-1065106.385	-23.311	-1065129.696	AN	
18. [Fe(PaPy$_3$)(Cl)]^{0}	2	-1065314.053	237.179	-35.110	**-1065111.984**	-20.481	**-1065132.465**	AN	
19. [Fe(PaPy$_3$)(N$_3$)]$^{1+}$	1/2	-879691.167	247.793	-33.300	-879476.674	-49.403	**-879526.077**	AN	1/2[113]
19. [Fe(PaPy$_3$)(N$_3$)]$^{1+}$	5/2	-879686.126	245.489	-36.605	**-879477.242**	-47.227	-879524.469	AN	
19. [Fe(PaPy$_3$)(N$_3$)]^{0}	0	-879803.986	246.345	-33.138	-879590.779	-20.558	-879611.337	AN	
19. [Fe(PaPy$_3$)(N$_3$)]^{0}	2	-879804.431	244.428	-36.660	**-879596.663**	-18.665	**-879615.328**	AN	
25. [Mn(CN)$_6$]$^{3-}$	1	-412321.483	29.926	-25.536	**-412317.093**	-387.796	**-412704.889**	water	
25. [Mn(CN)$_6$]$^{3-}$	2	-412312.057	27.734	-28.103	-412312.426	-378.415	-412690.841	water	
25. [Mn(CN)$_6$]$^{4-}$	1/2	-412122.048	29.143	-25.527	-412118.432	-684.755	**-412803.187**	water	1/2[134]
25. [Mn(CN)$_6$]$^{4-}$	5/2	-412142.576	24.959	-31.493	**-412149.110**	-644.859	-412793.969	water	

Table A7. Energy terms calculated using B4LYP functional with â₀=0.13

model compound	spin theory	E_{0K}^{B4LYP}	ZPE	$\Delta G_{0\rightarrow 298K}$	G_g^0	ΔG_{sol}^0	G_s^0	solvent	spin exp.
		kcal·mol^{-1}							
4. [Fe(phen)$_3$]$^{3+}$	1/2	-1142546.978	329.319	-36.195	**-1142253.854**	-278.196	**-1142532.050**	water	
						-267.357	**-1142521.211**	AN	
						-258.528	**-1142512.382**	DMF	
4. [Fe(phen)$_3$]$^{3+}$	5/2	-1142533.296	327.512	-38.780	-1142244.564	-274.108	-1142518.672	water	
						-263.819	-1142508.383	AN	
						-255.380	-1142499.944	DMF	
4. [Fe(phen)$_3$]$^{2+}$	0	-1142804.694	328.962	-36.295	**-1142512.027**	-126.922	**-1142638.949**	water	0[131]
						-121.599	**-1142633.626**	AN	
						-116.153	**-1142628.180**	DMF	
4. [Fe(phen)$_3$]$^{2+}$	2	-1142794.106	326.959	-40.942	-1142508.089	-124.844	-1142632.933	water	
						-119.708	-1142627.797	AN	
						-114.449	-1142622.538	DMF	
6. [Fe(sar)]$^{3+}$	1/2	-624135.746	315.435	-26.143	**-623846.454**	-362.269	**-624208.723**	water	1/2[157]
6. [Fe(sar)]$^{3+}$	5/2	-624125.252	309.353	-29.469	-623845.368	-356.576	-624201.944	water	
6. [Fe(sar)]$^{2+}$	0	-624416.254	311.811	-26.271	**-624130.714**	-163.102	**-624293.816**	water	0 [132]
6. [Fe(sar)]$^{2+}$	2	-624409.689	309.353	-29.469	-624129.805	-160.454	-624290.259	water	
10. [Fe(PypepO)$_2$]$^{1-}$	1/2	-975184.377	225.576	-34.001	-974992.802	-50.837	-975043.639	DMF	
10. [Fe(PypepO)$_2$]$^{1-}$	5/2	-975184.651	224.303	-36.071	**-974996.419**	-48.775	**-975045.194**	DMF	5/2[109]
10. [Fe(PypepO)$_2$]$^{2-}$	0	-975151.589	223.475	-33.572	-974961.686	-147.865	-975109.551	DMF	
10. [Fe(PypepO)$_2$]$^{2-}$	2	-975157.145	223.115	-36.472	**-974970.502**	-145.804	**-975116.306**	DMF	
11. [Fe(PypepS)$_2$]$^{1-}$	1/2	-1379055.359	223.127	-35.402	**-1378867.634**	-48.364	**-1378915.998**	DMF	1/2[110]
11. [Fe(PypepS)$_2$]$^{1-}$	5/2	-1379046.980	221.733	-38.050	-1378863.297	-46.455	-1378909.752	DMF	
11. [Fe(PypepS)$_2$]$^{2-}$	0	-1379023.920	221.234	-35.215	**-1378837.901**	-144.017	**-1378981.918**	DMF	
11. [Fe(PypepS)$_2$]$^{2-}$	2	-1379022.710	229.245	-37.126	-1378830.591	-142.205	-1378972.796	DMF	
12. [Fe(PyAS)$_2$]$^{1+}$	1/2	-1285875.063	233.311	-34.210	**-1285675.962**	-40.029	**-1285715.991**	DMF	1/2[110, 133]
12. [Fe(PyAS)$_2$]$^{1+}$	5/2	-1285857.296	232.616	-36.983	-1285661.663	-38.579	-1285700.242	DMF	
12. [Fe(PyAS)$_2$]^{0}	0	-1285986.893	231.975	-34.197	**-1285789.115**	-13.209	**-1285802.324**	DMF	0[133]
12. [Fe(PyAS)$_2$]^{0}	2	-1285977.012	230.346	-36.775	-1285783.441	-14.694	-1285798.135	DMF	
13. [Fe(bpteta)]$^{3+}$	1/2	-717473.635	294.967	-30.055	**-717208.723**	-326.462	**-717535.185**	AN	
13. [Fe(bpteta)]$^{3+}$	5/2	-717462.840	292.202	-32.686	-717203.324	-320.844	-717524.168	AN	
13. [Fe(bpteta)]$^{2+}$	0	-717751.846	294.258	-29.925	-717487.513	-147.516	**-717635.029**	AN	0[101]
13. [Fe(bpteta)]$^{2+}$	2	-717747.951	291.992	-33.080	**-717489.039**	-144.220	-717633.259	AN	
16. [Fe(Prpep)$_2$]$^{1+}$	1/2	-992449.649	258.928	-34.819	**-992225.540**	-65.774	**-992291.314**	AN	1/2[105]
16. [Fe(Prpep)$_2$]$^{1+}$	5/2	-992441.090	256.592	-37.839	-992222.337	-63.712	-992286.049	AN	
16. [Fe(Prpep)$_2$]^{0}	0	-992564.940	257.152	-35.014	-992342.802	-34.655	**-992377.457**	AN	0[105]
16. [Fe(Prpep)$_2$]^{0}	2	-992560.411	255.081	-38.769	**-992344.099**	-33.841	**-992377.940**	AN	
18. [Fe(PaPy$_3$)(Cl)]$^{1+}$	1/2	-1062802.579	241.645	-31.852	**-1062592.786**	-50.240	**-1062643.026**	AN	1/2[113]
18. [Fe(PaPy$_3$)(Cl)]$^{1+}$	5/2	-1062794.477	238.218	-36.025	-1062592.284	-47.917	-1062640.201	AN	
18. [Fe(PaPy$_3$)(Cl)]^{0}	0	-1062913.220	238.712	-32.218	-1062706.726	-23.311	-1062730.037	AN	
18. [Fe(PaPy$_3$)(Cl)]^{0}	2	-1062911.733	237.179	-35.110	**-1062709.664**	-20.481	**-1062730.145**	AN	
19. [Fe(PaPy$_3$)(N$_3$)]$^{1+}$	1/2	-877393.317	247.793	-33.300	**-877178.824**	-49.403	**-877228.227**	AN	1/2[113]
19. [Fe(PaPy$_3$)(N$_3$)]$^{1+}$	5/2	-877386.187	245.489	-36.605	-877177.303	-47.227	-877224.530	AN	
19. [Fe(PaPy$_3$)(N$_3$)]^{0}	0	-877502.965	246.345	-33.138	-877289.758	-20.558	-877310.316	AN	
19. [Fe(PaPy$_3$)(N$_3$)]^{0}	2	-877500.734	244.428	-36.660	**-877292.966**	-18.665	**-877311.631**	AN	
25. [Mn(CN)$_6$]$^{3-}$	1	-411304.440	29.926	-25.536	**-411300.050**	-387.796	**-411687.846**	water	
25. [Mn(CN)$_6$]$^{3-}$	2	-411293.686	27.734	-28.103	-411294.055	-378.415	-411672.470	water	
25. [Mn(CN)$_6$]$^{4-}$	1/2	-411104.348	29.143	-25.527	-411100.732	-684.755	**-411785.487**	water	1/2[134]
25. [Mn(CN)$_6$]$^{4-}$	5/2	-411120.460	24.959	-31.493	**-411126.994**	-644.859	-411771.853	water	

Table A8. Energy terms calculated using B4LYP functional with â$_0$=0.11

model compound	spin theory	E_{0K}^{B4LYP}	ZPE	$\Delta G_{0\rightarrow 298K}$	G_g^0	ΔG_{sol}^0	G_s^0	solvent	spin exp.
		kcal·mol^{-1}							
4. [Fe(phen)$_3$]$^{3+}$	1/2	-1139468.710	329.319	-36.195	**-1139175.586**	-278.196 -267.357 -258.528	**-1139453.782** **-1139442.943** **-1139434.114**	water AN DMF	
4. [Fe(phen)$_3$]$^{3+}$	5/2	-1139452.610	327.512	-38.780	-1139163.878	-274.108 -263.819 -255.380	-1139437.986 -1139427.697 -1139419.258	water AN DMF	
4. [Fe(phen)$_3$]$^{2+}$	0	-1139722.698	328.962	-36.295	**-1139430.031**	-126.922 -121.599 -116.153	**-1139556.953** **-1139551.630** **-1139546.184**	water AN DMF	0[131]
4. [Fe(phen)$_3$]$^{2+}$	2	-1139709.347	326.959	-40.942	-1139423.330	-124.844 -119.708 -114.449	-1139548.174 -1139543.038 -1139537.779	water AN DMF	
6. [Fe(sar)]$^{3+}$	1/2	-622446.918	315.435	-26.143	**-622157.626**	-362.269	**-622519.895**	water	1/2[157]
6. [Fe(sar)]$^{3+}$	5/2	-622434.590	309.353	-29.469	-622154.706	-356.576	-622511.282	water	
6. [Fe(sar)]$^{2+}$	0	-622722.949	311.811	-26.271	**-622437.409**	-163.102	**-622600.511**	water	0[132]
6. [Fe(sar)]$^{2+}$	2	-622714.107	309.353	-29.469	-622434.223	-160.454	-622594.677	water	
10. [Fe(PypepO)$_2$]$^{1-}$	1/2	-972663.655	225.576	-34.001	-972472.080	-50.837	**-972522.917**	DMF	
10. [Fe(PypepO)$_2$]$^{1-}$	5/2	-972661.260	224.303	-36.071	**-972473.028**	-48.775	-972521.803	DMF	5/2[109]
10. [Fe(PypepO)$_2$]$^{2-}$	0	-972629.192	223.475	-33.572	-972439.289	-147.865	-972587.154	DMF	
10. [Fe(PypepO)$_2$]$^{2-}$	2	-972631.682	223.115	-36.472	**-972445.039**	-145.804	**-972590.843**	DMF	
11. [Fe(PypepS)$_2$]$^{1-}$	1/2	-1376114.327	223.127	-35.402	**-1375926.602**	-48.364	**-1375974.966**	DMF	1/2[110]
11. [Fe(PypepS)$_2$]$^{1-}$	5/2	-1376103.756	221.733	-38.050	-1375920.073	-46.455	-1375966.528	DMF	
11. [Fe(PypepS)$_2$]$^{2-}$	0	-1376080.754	221.234	-35.215	**-1375894.735**	-144.017	**-1376038.752**	DMF	
11. [Fe(PypepS)$_2$]$^{2-}$	2	-1376077.001	229.245	-37.126	-1375884.882	-142.205	-1376027.087	DMF	
12. [Fe(PyAS)$_2$]$^{1+}$	1/2	-1283136.629	233.311	-34.210	**-1282937.528**	-40.029	**-1282977.557**	DMF	1/2[110, 133]
12. [Fe(PyAS)$_2$]$^{1+}$	5/2	-1283116.641	232.616	-36.983	-1282921.008	-38.579	-1282959.587	DMF	
12. [Fe(PyAS)$_2$]^{0}	0	-1283245.793	231.975	-34.197	**-1283048.015**	-13.209	**-1283061.224**	DMF	0[133]
12. [Fe(PyAS)$_2$]^{0}	2	-1283233.158	230.346	-36.775	-1283039.587	-14.694	-1283054.281	DMF	
13. [Fe(bpteta)]$^{3+}$	1/2	-715544.408	294.967	-30.055	**-715279.496**	-326.462	**-715605.958**	AN	
13. [Fe(bpteta)]$^{3+}$	5/2	-715531.550	292.202	-32.686	-715272.034	-320.844	-715592.878	AN	
13. [Fe(bpteta)]$^{2+}$	0	-715818.243	294.258	-29.925	**-715553.910**	-147.516	**-715701.426**	AN	0[101]
13. [Fe(bpteta)]$^{2+}$	2	-715811.912	291.992	-33.080	-715553.000	-144.220	-715697.220	AN	
16. [Fe(Prpep)$_2$]$^{1+}$	1/2	-989884.881	258.928	-34.819	**-989660.772**	-65.774	**-989726.546**	AN	1/2 [105]
16. [Fe(Prpep)$_2$]$^{1+}$	5/2	-989874.184	256.592	-37.839	-989655.431	-63.712	-989719.143	AN	
16. [Fe(Prpep)$_2$]^{0}	0	-989997.200	257.152	-35.014	**-989775.062**	-34.655	**-989809.717**	AN	0[105]
16. [Fe(Prpep)$_2$]^{0}	2	-989990.168	255.081	-38.769	-989773.856	-33.841	-989807.697	AN	
18. [Fe(PaPy$_3$)(Cl)]$^{1+}$	1/2	-1060409.034	241.645	-31.852	**-1060199.241**	-50.240	**-1060249.481**	AN	1/2[113]
18. [Fe(PaPy$_3$)(Cl)]$^{1+}$	5/2	-1060398.951	238.218	-36.025	-1060196.758	-47.917	-1060244.675	AN	
18. [Fe(PaPy$_3$)(Cl)]^{0}	0	-1060516.804	238.712	-32.218	-1060310.310	-23.311	**-1060333.621**	AN	
18. [Fe(PaPy$_3$)(Cl)]^{0}	2	-1060512.564	237.179	-35.110	**-1060310.495**	-20.481	-1060330.976	AN	
19. [Fe(PaPy$_3$)(N$_3$)]$^{1+}$	1/2	-875098.428	247.793	-33.300	**-874883.935**	-49.403	**-874933.338**	AN	1/2 [113]
19. [Fe(PaPy$_3$)(N$_3$)]$^{1+}$	5/2	-875089.324	245.489	-36.605	-874880.440	-47.227	-874927.667	AN	
19. [Fe(PaPy$_3$)(N$_3$)]^{0}	0	-875205.156	246.345	-33.138	-874991.949	-20.558	-875012.507	AN	
19. [Fe(PaPy$_3$)(N$_3$)]^{0}	2	-875200.261	244.428	-36.660	**-874992.493**	-18.665	**-875011.158**	AN	
25. [Mn(CN)$_6$]$^{3-}$	1	-410289.113	29.926	-25.536	**-410284.723**	-387.796	**-410672.519**	water	
25. [Mn(CN)$_6$]$^{3-}$	2	-410276.930	27.734	-28.103	-410277.299	-378.415	-410655.714	water	
25. [Mn(CN)$_6$]$^{4-}$	1/2	-410088.466	29.143	-25.527	-410084.850	-684.755	**-410769.605**	water	1/2 [134]
25. [Mn(CN)$_6$]$^{4-}$	5/2	-410100.178	24.959	-31.493	**-410106.712**	-644.859	-410751.571	water	

Table A9. Solvation energies and their differences for the states described in the paragraph 4.1.1

model compound	$\Delta G^{\circ}_{sol,LS}$	$\Delta G^{\circ}_{sol,HS}$	$\Delta G^{\circ}_{sol,(HS;LS)}$	$\Delta G^{\circ}_{sol,(LS;HS)}$	$\Delta\Delta G^{\circ}_{sol,LS/HS}$	$\Delta\Delta G^{\circ,structure}_{sol,LS/HS}$	$\Delta\Delta G^{\circ,charge}_{sol,LS/HS}$	solvent
	kcal·mol⁻¹							
training set (1-30)								
1. $[Fe(CN)_6]^{3-}$	-391.166	-376.897	-376.317	-392.313	14.269	14.849	-0.580	water
1. $[Fe(CN)_6]^{4-}$	-691.396	-652.496	-649.040	-699.968	38.900	42.356	-3.456	water
2. $[Fe(bpy)_3]^{3+}$	-289.042	-284.619	-285.301	-288.406	4.423	3.741	0.682	water
	-277.750	-273.782	-274.285	-277.285	3.968	3.465	0.503	AN
	-269.011	-265.380	-265.642	-268.789	3.631	3.369	0.262	DMF
2. $[Fe(bpy)_3]^{2+}$	-131.410	-128.616	-129.450	-130.474	2.794	1.960	0.834	water
	-125.920	-123.431	-124.137	-120.611	2.489	1.783	0.706	AN
	-120.611	-118.463	-118.802	-120.257	2.148	1.809	0.339	DMF
3. $[Fe(bpy)_2(CN)_2]^{1+}$	-62.248	-58.889	-63.660	-58.433	3.359	-1.412	4.771	water
3. $[Fe(bpy)_2(CN)_2]^{0}$	-38.159	-35.741	-42.639	-35.645	2.418	-4.481	6.898	water
4. $[Fe(phen)_3]^{3+}$	-278.196	-274.108	-274.872	-277.438	4.088	3.324	0.764	water
	-267.357	-263.819	-264.431	-266.747	3.538	2.926	0.612	AN
	-258.528	-255.380	-255.670	-258.229	3.148	2.858	0.290	DMF
4. $[Fe(phen)_3]^{2+}$	-126.922	-124.844	-125.080	-126.582	2.078	1.842	0.236	water
	-121.599	-119.708	-119.958	-121.285	1.891	1.641	0.250	AN
	-116.153	-114.449	-114.490	-116.043	1.704	1.663	0.041	DMF
5. $[Fe(diammac)]^{3+}$	-370.965	-366.310	-365.334	-372.043	4.655	5.631	-0.976	water
5. $[Fe(diammac)]^{2+}$	-167.401	-165.555	-164.927	-168.223	1.846	2.474	-0.628	water
6. $[Fe(sar)]^{3+}$	-362.269	-356.576	-355.299	-363.785	5.693	6.970	-1.277	water
6. $[Fe(sar)]^{2+}$	-163.102	-160.454	-159.820	-164.563	2.648	3.282	-0.634	water
7. $[Fe(tacn)_2]^{3+}$	-370.195	-363.689	-365.291	-373.252	6.506	4.904	1.602	water
7. $[Fe(tacn)_2]^{2+}$	-166.030	-163.698	-162.676	-166.891	2.332	3.354	-1.022	water
8. $[Fe(PyIm_2H_2)_2]^{3+}$	-301.079	-296.312	-297.205	-300.292	4.767	3.874	0.893	AN
8. $[Fe(PyIm_2H_2)_2]^{2+}$	-142.874	-140.476	-141.426	-142.091	2.398	1.448	0.950	AN
9. $[Fe(PyIm_2)_2]^{1-}$	-49.817	-48.560	-49.046	-49.403	1.258	0.771	0.487	AN
9. $[Fe(PyIm_2)_2]^{2-}$	-146.626	-143.464	-143.436	-146.730	3.162	3.190	-0.028	AN
10. $[Fe(PypepO)_2]^{1-}$	-50.837	-48.776	-49.212	-51.357	2.061	1.625	0.437	DMF
10. $[Fe(PypepO)_2]^{2-}$	-147.865	-145.804	-143.704	-152.239	2.061	4.161	-2.100	DMF
11. $[Fe(PypepS)_2]^{1-}$	-48.364	-46.455	-46.951	-48.320	1.909	1.413	0.496	DMF
11. $[Fe(PypepS)_2]^{2-}$	-144.017	-142.205	-139.442	-147.811	1.812	4.575	-2.763	DMF
12. $[Fe(PyAS)_2]^{1+}$	-40.029	-38.580	-40.684	-38.414	1.450	-0.655	2.104	DMF
12. $[Fe(PyAS)_2]^{0}$	-13.209	-14.694	-13.542	-15.700	-1.484	-0.332	-1.152	DMF
13. $[Fe(bpteta)]^{3+}$	-326.462	-320.844	-321.035	-326.261	5.618	5.427	0.191	AN
13. $[Fe(bpteta)]^{2+}$	-147.516	-144.220	-144.412	-147.199	3.296	3.104	0.192	AN
14. $[Fe(DITim)_2]^{1+}$	-48.253	-47.241	-48.288	-48.024	1.012	-0.034	1.046	AN
14. $[Fe(DITim)_2]^{0}$	-23.490	-22.391	-23.372	-22.871	1.099	0.118	0.981	AN
15. $[Fe(Pypep)_2]^{1+}$	-60.127	-57.417	-60.070	-58.282	2.710	0.057	2.653	water
	-54.608	-52.086	-54.307	-53.056	2.522	0.301	2.221	AN
	-49.562	-47.535	-49.521	-48.260	2.027	0.041	1.986	DMF
15. $[Fe(Pypep)_2]^{0}$	-31.049	-30.282	-32.177	-29.363	0.767	-1.127	1.894	water
	-25.058	-24.491	-26.192	-23.494	0.567	-1.134	1.701	AN
	-21.778	-21.255	-22.807	-20.352	0.522	-1.029	1.551	DMF
16. $[Fe(Prpep)_2]^{1+}$	-65.774	-63.712	-65.415	-64.110	2.061	0.358	1.703	AN
16. $[Fe(Prpep)_2]^{0}$	-34.655	-33.841	-35.655	-33.038	0.814	-1.000	1.814	AN
17. $[Fe(PaPy_3)(AN)]^{2+}$	-141.658	-136.722	-138.181	-140.063	4.936	3.477	1.459	AN
17. $[Fe(PaPy_3)(AN)]^{1+}$	-47.296	-45.751	-46.905	-46.855	1.545	0.392	1.153	AN
18. $[Fe(PaPy_3)(Cl)]^{1+}$	-50.239	-48.444	-52.967	-47.715	1.794	-2.729	4.523	AN
18. $[Fe(PaPy_3)(Cl)]^{0}$	-23.311	-21.823	-27.158	-20.539	1.487	-3.848	5.335	AN
19. $[Fe(PaPy_3)(N_3)]^{1+}$	-49.403	-47.227	-51.437	-46.522	2.177	-2.034	4.210	AN
19. $[Fe(PaPy_3)(N_3)]^{0}$	-20.559	-18.665	-23.157	-17.882	1.893	-2.598	4.491	AN
20. $[Fe(PaPy_3)(CN)]^{1+}$	-53.573	-52.025	-56.210	-50.633	1.547	-2.637	4.185	AN
20. $[Fe(PaPy_3)(CN)]^{0}$	-24.154	-22.543	-28.230	-20.433	1.611	-4.076	5.687	AN
21. $[Fe(PaPy_2O)(Cl)]^{0}$	-19.690	-18.363	-21.222	-17.945	1.327	-1.532	2.859	DMF
21. $[Fe(PaPy_2O)(Cl)]^{1-}$	-58.435	-58.727	-58.544	-59.835	-0.291	-0.109	-0.182	DMF
22. $[Fe(SEt)_4]^{1-}$	-44.297	-44.330	-43.971	-44.466	-0.033	0.326	-0.359	AN
22. $[Fe(SEt)_4]^{2-}$	-157.088	-157.843	-155.863	-159.462	-0.755	1.225	-1.980	AN
23. $[Fe(S_2\text{-}o\text{-}xyl)_2]^{1-}$	-46.821	-45.682	-45.315	-47.233	1.138	1.506	-0.367	DMF
23. $[Fe(S_2\text{-}o\text{-}xyl)_2]^{2-}$	-156.083	-151.448	-150.756	-157.760	4.635	5.327	-0.692	DMF
24. $[Fe(SEtOH)_4]^{1-}$	-67.591	-71.019	-78.416	-61.334	-3.428	-10.825	7.396	water
24. $[Fe(SEtOH)_4]^{2-}$	-193.297	-192.200	-192.917	-192.978	1.097	0.380	0.717	water
25. $[Mn(CN)_6]^{3-}$	-387.796	-378.415	-377.993	-390.114	9.381	9.803	-0.422	water
25. $[Mn(CN)_6]^{4-}$	-684.755	-644.859	-641.548	-692.939	39.896	43.207	-3.311	water
26. $[Mn(bpia)(Cl)_2]^{1+}$	-53.875	-51.765	-55.739	-51.090	2.110	-1.864	3.974	AN
26. $[Mn(bpia)(Cl)_2]^{0}$	-30.876	-32.401	-31.102	-34.131	-1.524	-0.226	-1.299	AN

27. [Mn(py$_2$(NMe)$_2$Cl$_2$)]$^{1+}$	-55.172	-51.597	-57.164	-50.241	3.575	-1.992	5.567	AN
27. [Mn(py$_2$(NMe)$_2$Cl$_2$)]^{0}	-28.761	-30.344	-29.150	-31.275	-1.582	-0.389	-1.194	AN
28. [Mn(bpteta)]$^{3+}$	-321.128	-318.342	-318.778	-320.812	2.786	2.350	0.436	AN
28. [Mn(bpteta)]$^{2+}$	-144.365	-143.191	-142.980	-144.619	1.174	1.385	-0.211	AN
29. [Ni(bpy)$_3$]$^{3+}$	-288.437	-283.266	-284.785	-287.054	5.171	3.652	1.519	water
30. [Ni(bpteta)]$^{3+}$	-325.745	-315.733	-320.925	-321.219	10.012	4.820	5.192	AN
prediction set (31-48)								
31. [Fe(cyclamAc)(N$_3$)]$^{2+}$	-167.315	-164.125	-166.330	-165.183	3.190	0.985	2.205	AN
31. [Fe(cyclamAc)(N$_3$)]$^{1+}$	-56.615	-55.866	-57.075	-55.836	0.749	-0.460	1.209	AN
31. [Fe(cyclamAc)(N$_3$)]^{0}	-19.810	-19.418	-21.043	-18.293	0.392	-1.234	1.625	AN
32. [Ni(tacn)]$^{3+}$	-369.019	-365.767	-365.349	-369.370	3.252	3.670	-0.418	water
33. [Fe(dtne)]$^{3+}$	-360.352	-355.153	-355.108	-360.396	5.199	5.244	-0.045	water
33. [Fe(dtne)]$^{2+}$	-161.444	-158.680	-158.688	-161.680	2.764	2.756	0.008	water
34. [Ni(dtne)]$^{3+}$	-360.034	-353.313	-354.533	-360.465	6.721	5.501	1.220	water
35. [Fe(terpy)$_2$]$^{3+}$	-270.353	-266.804	-266.961	-270.212	3.549	3.392	0.157	DMF
35. [Fe(terpy)$_2$]$^{2+}$	-121.525	-119.253	-119.565	-121.263	2.272	1.960	0.312	DMF
36. [Fe(SPh)$_4$]$^{1-}$	-43.712	-43.347	-43.128	-44.232	0.365	0.584	-0.218	AN
36. [Fe(SPh)$_4$]$^{2-}$	-137.387	-137.676	-136.458	-138.778	-0.289	0.929	-1.218	AN
37. [Fe(SCH$_2$CON(CH$_3$)$_2$)$_4$]$^{1-}$	-50.733	-48.978	-49.112	-50.427	1.755	1.620	0.134	AN
	-49.403	-47.594	-47.681	-49.125	1.809	1.722	0.087	DMF
37. [Fe(SCH$_2$CON(CH$_3$)$_2$)$_4$]$^{2-}$	-139.201	-138.455	-136.155	-140.698	0.746	3.046	-2.300	AN
	-137.523	-136.441	-134.274	-138.868	1.082	3.249	-2.167	DMF
38. [Fe(S-*i*-Pr)$_4$]$^{1-}$	-42.840	-42.818	-42.502	-42.818	0.022	0.338	-0.316	AN
38. [Fe(S-*i*-Pr)$_4$]$^{2-}$	-149.016	-149.880	-147.923	-152.081	-0.864	1.093	-1.957	AN
39. [Fe(bpy)(CN)$_4$]$^{3+}$	-82.749	-74.900	-84.096	-77.284	7.849	-1.347	9.196	water
39. [Fe(bpy)(CN)$_4$]$^{2+}$	-200.870	-199.798	-198.767	-211.271	1.072	2.103	-1.031	water
40. [Fe(tacnPy$_2$)(AN)]$^{3+}$	-305.298	-300.986	-301.119	-305.549	4.312	4.179	0.133	AN
40. [Fe(tacnPy$_2$)(AN)]$^{2+}$	-136.942	-134.462	-134.822	-136.940	2.480	2.120	0.360	AN
41. [Fe(N$_4$Py)(Cl)]$^{2+}$	-139.806	-136.901	-140.798	-137.254	2.905	-0.992	3.897	AN
41. [Fe(N$_4$Py)(Cl)]$^{1+}$	-47.711	-45.560	-51.275	-45.192	2.150	-3.564	5.715	AN
42. [Fe(N$_4$Py)(AN)]$^{3+}$	-290.331	-286.482	-286.389	-290.829	3.849	3.942	-0.093	AN
42. [Fe(N$_4$Py)(AN)]$^{2+}$	-131.806	-129.527	-129.867	-131.806	2.279	1.939	0.340	AN
43. [Fe(Py$_3$tacn)]$^{3+}$	-302.070	-296.883	-298.856	-300.764	5.187	3.214	1.973	AN
43. [Fe(Py$_3$tacn)]$^{2+}$	-138.213	-132.977	-134.163	-137.209	5.236	4.050	1.186	AN
44. [Ni(Py$_3$tacn)]$^{3+}$	-301.650	-301.902	-301.934	-302.001	-0.252	-0.284	0.032	AN
45. [Fe(TCTA)]^{0}	-39.970	-34.433	-38.337	-37.095	5.537	1.633	3.904	water
45. [Fe(TCTA)]$^{1-}$	-76.677	-72.485	-74.069	-76.294	4.191	2.608	1.584	water
46. [Ni(TCTA)]^{0}	-38.962	-36.759	-41.538	-39.054	2.204	-2.576	4.780	water
47. [Mn(TCTA)]^{0}	-39.943	-36.229	-38.271	-38.221	3.714	1.672	2.042	water
47. [Mn(TCTA)]$^{1-}$	-77.145	-71.304	-72.173	-77.856	5.841	4.972	0.868	water
48. [Fe(Pyr$_2$Py)$_2$]$^{3+}$	-286.304	-281.656	-281.651	-286.324	4.648	4.653	-0.005	AN
48. [Fe(Pyr$_2$Py)$_2$]$^{2+}$	-127.524	-125.153	-125.285	-127.415	2.371	2.239	0.132	AN