A Density Functional Theory and Quantum Theory of Atoms-in-Molecules Analysis of the Stability of Ni(II) Complexes of Some Amino-Alcohol Ligands

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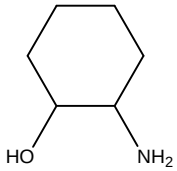
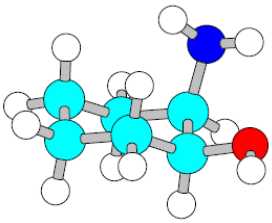
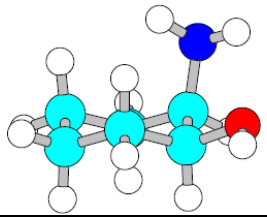
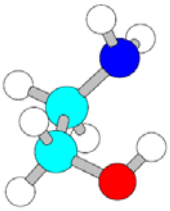
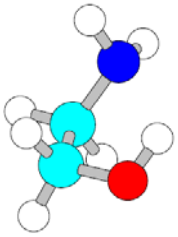
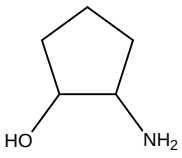
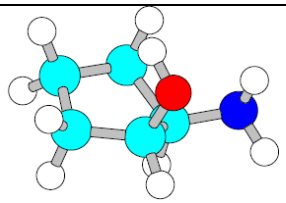
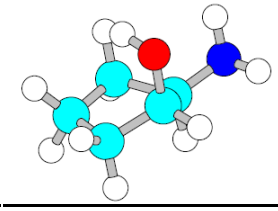
Supporting Information

Table S1. The Basis Set Superposition Error (BSSE), the Uncorrected Binding Energies (E_b), the ZPVE-Corrected Dissociation Energies (E_d^c), the Difference Between the E_d^c Values of the *cis* and *trans* Isomers, the Partial Charge on the Nickel Ion (Ni^Q) for Ni(II) Complexes of Three Amino Alcohol Ligands Obtained from a X3LYP/6-31+G(d,p) Study in the Gas Phase

Complex	BSSE /kcal mol^{-1}	E_b /kcal mol^{-1}	E_b(BSSE) /kcal mol^{-1}	E_d^c /kcal mol^{-1}	E_d^c(BSSE) /kcal mol^{-1}	ΔE_d^c(BSSE) (*cis* – *trans*) /kcal mol^{-1}	Ni^Q /e	ΔQ /e
$[Ni(BHEEN)(H_2O)_2]^{2+}$								
trans	10.7	-386.729	-376.003	377.570	366.843	2.595	1.421	0.579
cis	10.7	-389.151	-378.457	380.132	369.438		1.426	0.574
$[Ni(Cy_2EN)(H_2O)_2]^{2+}$								
trans	10.2	-394.452	-384.235	386.147	375.929	3.514	1.422	0.578
cis	10.0	-398.003	-387.981	389.465	379.443		1.429	0.571
$[Ni(Cyp_2EN)(H_2O)_2]^{2+}$								
trans	10.0	-386.099	-376.089	376.982	366.971	8.028	1.432	0.568
cis	10.6	-394.657	-384.022	385.634	375.000		1.429	0.571

Table S2. The Energy-Minimized Structures (X3LYP/6-311+G(d,p) of Three Amino-Alcohol Models for the Ligands Investigated in This Work.

Model ligand	**Energy-minimized structure**	**N–C–C–O torsion /deg**	**Electronic + thermal free energies, G_{calc} /au**	**$\Delta G = G_{constrained} - G_{free}$ /kcal mol^{-1}**
 (free)		-53.2	-366.21795	0.019
(constrained)		-51.9	-366.21792	
$NH_2CH_2CH_2OH$ (free)		-55.5	-210.29659	0.069
(constrained)		-52.5	-210.29648	
 (free)		44.5	-326.93588	2.014
(constrained)		59.4	-326.93267	

Tables S3. Strain Analysis in the Complexes of Ni(II) with BHEEN, Cy_2EN and Cyp_2EN Involving Bond Path Angles (BPA) and Geometric Bond Angles (GBA).
(BA = bite angle; CR = chelating ring)

Table S3A: *cis*-$[Ni(BHEEN)(H_2O)_2]^{2+}$

Atom A	Atom B	Atom C	BPA / °	GBA / °	BPA-GBA	Comment
O2	Ni1	N11	65.05	80.99	-15.95	BA 1
O3	Ni1	N11	102.13	92.21	9.93	
O3	Ni1	O2	83.08	84.29	-1.21	
H4	O3	Ni1	114.89	118.25	-3.36	
H5	O3	Ni1	112.59	114.46	-1.87	
H5	O3	H4	103.60	106.09	-2.49	
H6	O2	Ni1	119.99	121.32	-1.32	
O7	Ni1	N11	168.02	177.35	-9.33	
O7	Ni1	O2	102.97	96.44	6.54	
O7	Ni1	O3	75.54	86.86	-11.32	
O9	Ni1	N11	111.68	98.52	13.16	
O9	Ni1	O2	121.08	101.67	19.41	
O9	Ni1	O3	144.29	168.40	-24.10	
O9	Ni1	O7	73.60	82.61	-9.00	
H8	O7	Ni1	112.84	114.23	-1.39	
N10	Ni1	N11	76.50	84.08	-7.57	BA 2
N10	Ni1	O2	141.54	165.01	-23.46	
N10	Ni1	O3	105.41	94.99	10.42	
N10	Ni1	O7	115.48	98.47	17.01	
N10	Ni1	O9	72.94	81.82	-8.89	BA 3
H23	O3	Ni1	78.33	80.26	-1.93	H-bond
H23	O3	H4	144.73	150.22	-5.49	H-bond
H23	O3	H5	100.21	83.92	16.29	H-bond
H15	O3	Ni1	80.50	78.17	2.33	H-bond
H15	O3	H4	113.56	104.77	8.79	H-bond
H15	O3	H5	130.50	134.09	-3.59	H-bond
H15	O3	H23	33.22	53.60	-20.38	H-bond
C19	O2	Ni1	109.70	110.95	-1.25	CR 1
C19	O2	H6	105.24	110.03	-4.79	
C12	N10	Ni1	109.62	108.31	1.32	CR 2
C13	N11	Ni1	108.56	105.13	3.43	CR 2
C13	C12	N10	107.85	110.61	-2.75	CR 2
C12	C13	N11	105.45	107.68	-2.23	CR 2
H14	C13	N11	109.31	111.88	-2.57	
H14	C13	C12	111.56	110.08	1.48	
C13	H15	O3	28.98	112.96	-83.98	H-bond
H15	C13	N11	106.44	107.91	-1.47	
H15	C13	C12	111.70	110.96	0.74	
H15	C13	H14	112.01	108.33	3.68	
H16	C12	N10	106.06	108.43	-2.37	
H16	C12	C13	110.23	109.25	0.97	
H17	C12	N10	108.39	110.38	-2.00	
H17	C12	C13	112.50	110.55	1.95	

Atom A	Atom B	Atom C	BPA / °	GBA / °	BPA-GBA	Comment
H17	C12	H16	111.52	107.53	3.98	
C18	N11	Ni1	109.93	107.86	2.07	CR 1
C18	N11	C13	112.79	116.99	-4.21	
H22	C18	N11	109.46	112.05	-2.58	
C18	C19	O2	104.14	107.07	-2.92	CR 1
C19	C18	N11	105.74	108.11	-2.36	CR 1
C19	C18	H22	111.39	109.65	1.74	
H20	C19	O2	105.27	109.09	-3.82	
H20	C19	C18	112.00	110.94	1.06	
H21	C19	O2	107.69	109.35	-1.66	
H21	C19	C18	113.65	110.86	2.80	
H21	C19	H20	113.19	109.48	3.71	
C18	H23	O3	0.86	110.48	-109.61	H-bond
H23	C18	N11	106.82	108.24	-1.41	
H23	C18	H22	112.09	108.36	3.72	
H23	C18	C19	111.03	110.44	0.59	
C24	O9	Ni1	111.24	111.84	-0.61	CR 3
C25	C24	O9	104.30	107.48	-3.18	CR 3
C25	N10	Ni1	109.51	106.91	2.60	CR 3
C25	N10	C12	109.89	114.07	-4.18	
N10	C25	C24	106.78	110.43	-3.66	CR 3
H26	C24	O9	105.00	108.94	-3.94	
H26	C24	C25	113.08	111.91	1.17	
H27	C24	O9	107.80	109.36	-1.55	
H27	C24	C25	113.08	110.11	2.97	
H27	C24	H26	112.72	109.00	3.72	
H28	O9	Ni1	119.58	120.45	-0.87	
H28	O9	C24	105.01	109.91	-4.90	
H29	N10	Ni1	113.75	110.88	2.86	
H29	N10	C12	107.21	108.57	-1.36	
H29	N10	C25	106.78	108.11	-1.34	
H32	C25	C24	111.15	109.05	2.09	
H32	C25	N10	108.83	111.72	-2.89	
H31	C25	C24	110.99	110.04	0.95	
H31	C25	N10	106.97	107.37	-0.40	
H31	C25	H32	111.89	108.19	3.70	
H30	N11	Ni1	111.90	109.82	2.08	
H30	N11	C13	106.96	108.57	-1.62	
H30	N11	C18	106.72	108.31	-1.60	
H33	O7	Ni1	115.65	118.92	-3.27	
H33	O7	H8	103.76	106.25	-2.50	

Table S3B: *trans*-[Ni(BHEEN)(H_2O)$_2$]$^{2+}$

Atom A	Atom B	Atom C	BPA / °	GBA / °	BPA-GBA	Comment
N3	Ni1	N2	75.70	85.27	-9.58	BA 2
O4	Ni1	N2	76.65	80.81	-4.16	BA 3
O4	Ni1	N3	136.71	161.16	-24.45	
O5	Ni1	N2	136.52	161.10	-24.58	

Atom A	Atom B	Atom C	BPA / °	GBA / °	BPA-GBA	Comment
O5	Ni1	N3	76.64	80.85	-4.21	BA 1
O5	Ni1	O4	143.41	115.34	28.07	
H6	O5	Ni1	119.12	120.07	-0.94	
H7	O4	Ni1	119.10	120.05	-0.95	
C8	N2	Ni1	109.02	106.25	2.76	CR 2
C13	N2	Ni1	110.70	107.71	2.99	CR 3
N2	C13	C12	105.69	107.83	-2.13	CR 3
C13	N2	C8	113.00	117.83	-4.83	
C9	N3	Ni1	109.09	106.28	2.81	CR 2
C10	N3	Ni1	110.64	107.64	3.00	CR 1
N3	C10	C11	105.74	107.89	-2.15	CR 1
C10	N3	C9	112.93	117.80	-4.87	
C9	C8	N2	105.88	107.94	-2.06	CR 2
C8	C9	N3	105.89	107.94	-2.05	CR 2
H20	C10	C11	111.44	109.39	2.05	
H20	C10	N3	109.59	112.15	-2.56	
C11	O5	Ni1	108.59	110.31	-1.72	CR 1
O5	C11	C10	104.57	107.76	-3.19	CR 1
C11	O5	H6	104.66	109.34	-4.68	
C12	O4	Ni1	108.57	110.33	-1.76	CR 3
O4	C12	C13	104.59	107.79	-3.19	CR 3
C12	O4	H7	104.64	109.32	-4.68	
H31	C13	C12	111.45	109.40	2.05	
H31	C13	N2	109.61	112.19	-2.57	
O14	Ni1	N2	94.53	90.86	3.68	
O14	Ni1	N3	122.66	102.56	20.10	
O14	Ni1	O4	92.07	90.34	1.73	
O14	Ni1	O5	73.40	79.80	-6.39	
H15	O14	Ni1	115.96	115.39	0.57	
O16	Ni1	N2	123.11	102.76	20.35	
O16	Ni1	N3	95.57	91.30	4.26	
O16	Ni1	O4	72.95	79.54	-6.59	
O16	Ni1	O5	92.35	90.38	1.97	
O16	Ni1	O14	132.90	161.38	-28.48	
H17	O16	Ni1	123.49	125.11	-1.63	
H18	O16	Ni1	116.60	115.88	0.73	
H18	O16	H17	104.41	106.96	-2.55	
H19	O14	Ni1	122.77	124.57	-1.80	
H19	O14	H15	104.34	106.82	-2.48	
H22	C11	C10	113.30	110.71	2.59	
H22	C11	O5	107.79	109.28	-1.49	
H21	C10	C11	111.27	110.75	0.52	
H21	C10	N3	106.60	108.42	-1.82	
H21	C10	H20	111.89	108.25	3.64	
H23	C11	C10	111.79	110.65	1.14	
H23	C11	O5	105.41	109.03	-3.62	
H23	C11	H22	113.20	109.37	3.82	
H24	N3	Ni1	111.60	109.74	1.85	
H24	N3	C9	106.32	107.55	-1.23	

Atom A	Atom B	Atom C	BPA / °	GBA / °	BPA-GBA	Comment
H24	N3	C10	106.20	107.68	-1.48	
H21	C10	C11	111.27	110.75	0.52	
H25	C9	N3	106.40	108.26	-1.86	
H25	C9	C8	110.70	110.24	0.46	
H26	C9	N3	109.31	111.55	-2.24	
H26	C9	C8	112.38	110.68	1.70	
H26	C9	H25	111.81	108.15	3.66	
H27	C8	N2	109.38	111.65	-2.27	
H27	C8	C9	112.41	110.74	1.67	
H28	C8	N2	106.33	108.12	-1.79	
H28	C8	C9	110.66	110.19	0.46	
H28	C8	H27	111.82	108.16	3.66	
H29	N2	Ni1	111.38	109.49	1.89	
H29	N2	C8	106.32	107.48	-1.16	
H29	N2	C13	106.35	107.89	-1.55	
H30	C13	C12	111.27	110.77	0.50	
H30	C13	N2	106.65	108.41	-1.75	
H30	C13	H31	111.85	108.26	3.59	
H33	C12	C13	113.29	110.71	2.58	
H33	C12	O4	107.81	109.31	-1.50	
H32	C12	C13	111.76	110.60	1.16	
H32	C12	O4	105.41	109.02	-3.61	
H32	C12	H33	113.20	109.37	3.83	

Table S3C: *cis*-$[Ni(Cy_2EN)(H_2O)_2]^{2+}$

Atom A	Atom B	Atom C	BPA / °	GBA / °	BPA-GBA	Comment
O2	Ni1	N7	69.85	80.69	-10.84	BA 1
O3	Ni1	N7	116.63	97.70	18.93	
O3	Ni1	O2	78.59	80.76	-2.17	
O4	Ni1	N7	94.24	91.44	2.80	
O4	Ni1	O2	130.45	107.19	23.26	
O4	Ni1	O3	145.06	168.83	-23.77	
O5	Ni1	N7	159.38	174.10	-14.72	
O5	Ni1	O2	105.43	97.37	8.06	
O5	Ni1	O3	80.76	87.44	-6.68	
O5	Ni1	O4	73.29	83.79	-10.50	
H46	O5	Ni1	116.45	119.32	-2.86	
N6	Ni1	N7	78.00	85.01	-7.01	BA 2
N6	Ni1	O2	139.77	162.70	-22.93	
N6	Ni1	O3	95.56	91.63	3.93	
N6	Ni1	O4	74.19	82.74	-8.55	BA 3
N6	Ni1	O5	112.90	97.78	15.12	
H40	O3	Ni1	83.24	78.65	4.58	
C8	N6	Ni1	107.57	104.25	3.32	CR 2
C9	C8	N6	108.62	112.83	-4.21	CR 2
H43	C8	N6	109.73	112.15	-2.42	
H43	C8	C9	111.45	109.38	2.07	

Atom A	Atom B	Atom C	BPA / °	GBA / °	BPA-GBA	Comment
C9	N7	Ni1	111.48	109.76	1.72	CR 2
N7	C9	C8	108.11	111.82	-3.71	CR 2
C10	N7	Ni1	109.96	106.12	3.84	CR 1
C10	N7	C9	113.07	118.75	-5.68	
C18	C10	N7	111.14	114.88	-3.74	
C11	O2	Ni1	111.58	111.76	-0.18	CR 1
C11	C10	N7	104.62	106.61	-1.99	CR 1
C11	C10	C18	111.66	109.15	2.51	Cyclohexyl 1
C10	C11	O2	104.53	107.62	-3.09	CR 1
C12	O4	Ni1	110.06	109.90	0.16	CR 3
C13	C12	O4	103.70	106.40	-2.70	CR 3
C14	C13	C12	112.80	110.35	2.45	Cyclohexyl 2
C13	N6	Ni1	111.17	108.80	2.37	CR 3
C13	N6	C8	113.08	120.15	-7.08	
N6	C13	C12	108.44	111.81	-3.37	CR 3
N6	C13	C14	111.72	115.68	-3.96	
C15	C12	O4	108.75	111.12	-2.37	
C15	C12	C13	114.20	110.65	3.55	Cyclohexyl 2
C16	C14	C13	108.79	110.18	-1.39	Cyclohexyl 2
C14	C16	C17	108.57	111.29	-2.72	Cyclohexyl 2
C15	C17	C16	108.56	111.42	-2.87	Cyclohexyl 2
C17	C15	C12	109.50	110.64	-1.15	Cyclohexyl 2
H26	C17	C16	111.49	110.46	1.03	
H26	C17	C15	109.74	109.49	0.25	
H39	C18	C10	109.23	109.90	-0.67	
C19	C11	O2	108.93	111.25	-2.32	
C19	C11	C10	115.22	111.94	3.28	Cyclohexyl 1
C19	C20	C21	108.47	111.34	-2.86	Cyclohexyl 1
C20	C19	C11	109.32	110.36	-1.04	Cyclohexyl 1
C21	C18	C10	109.36	110.76	-1.40	Cyclohexyl 1
C21	C18	H39	111.31	110.11	1.20	
C18	C21	C20	108.81	111.60	-2.79	Cyclohexyl 1
H36	C21	C20	111.22	110.29	0.93	
H36	C21	C18	109.90	109.63	0.27	
H22	C14	C13	108.75	110.31	-1.57	
H22	C14	C16	109.66	109.18	0.48	
H23	C14	C13	108.42	109.32	-0.89	
H23	C14	C16	111.85	110.60	1.26	
H23	C14	H22	109.30	107.20	2.09	
H24	C16	C17	111.09	110.22	0.87	
H24	C16	C14	109.99	109.72	0.27	
H31	C13	C12	108.00	106.47	1.52	
H31	C13	C14	111.37	107.99	3.38	
H31	C13	N6	104.07	103.87	0.21	
H25	C16	C17	109.30	109.43	-0.13	
H25	C16	C14	108.38	109.27	-0.89	
H25	C16	H24	109.46	106.80	2.67	
H30	C12	O4	104.90	108.21	-3.31	
H30	C12	C13	112.12	111.21	0.91	

Atom A	Atom B	Atom C	BPA / °	GBA / °	BPA-GBA	Comment
H30	C12	C15	112.31	109.21	3.10	
H27	C17	C16	109.16	109.33	-0.17	
H27	C17	C15	108.68	109.38	-0.70	
H27	C17	H26	109.16	106.63	2.53	
H28	C15	C12	108.50	109.42	-0.92	
H28	C15	C17	111.29	110.62	0.68	
H29	C15	C12	106.64	108.52	-1.88	
H29	C15	C17	110.71	110.08	0.63	
H29	C15	H28	110.07	107.48	2.59	
H41	H30	C12	77.95	131.03	-53.09	CH-HC bond
H32	C19	C11	108.45	109.38	-0.93	
H32	C19	C20	111.45	110.66	0.79	
H33	C19	C11	106.98	108.79	-1.82	
H33	C19	C20	110.57	110.15	0.42	
H33	C19	H32	109.94	107.43	2.51	
H49	C11	O2	104.22	107.51	-3.29	
H49	C11	C10	109.94	108.40	1.54	
H49	C11	C19	113.05	109.96	3.09	
H34	C20	C21	109.21	109.34	-0.13	
H34	C20	C19	108.59	109.29	-0.70	
H35	C20	C21	111.41	110.43	0.98	
H35	C20	C19	109.83	109.64	0.19	
H35	C20	H34	109.27	106.68	2.59	
C10	H40	O3	43.16	115.80	-72.64	H-bond
H40	C10	N7	106.87	107.42	-0.55	
H40	C10	C18	112.07	109.03	3.04	
H40	C10	C11	110.12	109.65	0.47	
H37	C21	C20	109.23	109.37	-0.15	
H37	C21	C18	108.41	109.16	-0.75	
H37	C21	H36	109.22	106.65	2.57	
H50	N7	Ni1	110.01	107.71	2.30	
H50	N7	C9	106.83	107.60	-0.77	
H50	N7	C10	105.25	106.42	-1.17	
H38	C18	C10	107.05	108.98	-1.93	
H38	C18	H39	109.48	107.15	2.34	
H38	C18	C21	110.31	109.88	0.44	
H41	C9	C8	111.70	110.54	1.16	
H41	C9	N7	106.24	107.88	-1.64	
C9	H41	H30	0.77	108.34	-107.57	CH-HC bond
H42	C9	C8	110.94	108.40	2.53	
H42	C9	N7	108.67	111.12	-2.45	
H42	C9	H41	110.98	106.97	4.01	
H44	C8	N6	104.91	105.45	-0.54	
H44	C8	C9	110.96	109.61	1.35	
H44	C8	H43	110.93	107.19	3.74	
H51	N6	Ni1	112.38	110.74	1.64	
H51	N6	C8	106.67	106.76	-0.09	
H51	N6	C13	105.96	106.06	-0.09	
H45	O5	Ni1	113.02	114.65	-1.63	

Atom A	Atom B	Atom C	BPA / °	GBA / °	BPA-GBA	Comment
H45	O5	H46	103.76	106.35	-2.59	
H47	O3	Ni1	116.09	119.89	-3.80	
H47	O3	H40	137.15	143.54	-6.39	H-bond
H48	O3	Ni1	113.56	114.97	-1.41	
H48	O3	H40	101.52	90.16	11.36	H-bond
H48	O3	H47	104.04	106.52	-2.48	
H52	O4	Ni1	117.69	119.44	-1.75	
H52	O4	C12	104.41	109.61	-5.21	
H53	O2	Ni1	120.53	121.49	-0.96	
H53	O2	C11	104.37	109.40	-5.03	

Table S3D: *trans*-$[Ni(Cy_2EN)(H_2O)_2]^{2+}$

Atom A	Atom B	Atom C	BPA / °	GBA / °	BPA-GBA	Comment
H3	C2	O22	104.32	107.53	-3.21	
C4	C2	O22	108.95	111.43	-2.48	
C4	C2	H3	112.97	109.74	3.23	
C2	C4	C7	109.13	110.13	-1.00	Cyclohexyl 1
H5	C4	C7	110.52	110.10	0.42	
H5	C4	C2	107.18	108.97	-1.79	
H6	C4	C7	111.45	110.72	0.73	
H6	C4	C2	108.48	109.42	-0.94	
H6	C4	H5	109.96	107.45	2.50	
H8	C7	C4	108.62	109.34	-0.72	
H9	C7	C4	109.88	109.69	0.20	
H9	C7	H8	109.23	106.65	2.58	
C10	C7	C4	108.46	111.24	-2.78	Cyclohexyl 1
C10	C7	H8	109.27	109.44	-0.17	
C10	C7	H9	111.34	110.38	0.97	
H11	C10	C7	109.16	109.27	-0.10	
C16	C2	O22	104.61	107.82	-3.22	CR 1
C16	C2	H3	110.10	108.33	1.77	
C16	C2	C4	115.00	111.84	3.16	Cyclohexyl 1
C16	N21	Ni1	110.68	106.66	4.01	CR 1
N21	C16	C2	105.00	106.84	-1.85	CR 1
C13	C10	C7	108.88	111.72	-2.84	Cyclohexyl 1
C13	C10	H11	108.50	109.21	-0.71	
H12	C10	C7	111.22	110.31	0.91	
H12	C10	H11	109.20	106.63	2.58	
H12	C10	C13	109.83	109.57	0.25	
C13	C16	C2	111.94	109.72	2.22	Cyclohexyl 1
C13	C16	N21	111.22	114.63	-3.41	
C16	C13	C10	109.58	111.13	-1.54	Cyclohexyl 1
H14	C13	C10	110.29	109.84	0.45	
H14	C13	C16	107.02	108.90	-1.88	
H15	C13	C10	111.49	110.29	1.21	
H15	C13	C16	108.92	109.47	-0.56	
H15	C13	H14	109.43	107.12	2.31	

Atom A	Atom B	Atom C	BPA / °	GBA / °	BPA-GBA	Comment
H17	C16	C2	110.00	109.38	0.62	
H17	C16	N21	106.20	107.07	-0.87	
H17	C16	C13	112.10	109.07	3.03	
C18	N21	Ni1	108.87	106.62	2.25	CR 2
C18	N21	C16	113.44	118.79	-5.35	
H19	C18	N21	106.77	108.34	-1.57	
H20	C18	N21	109.63	111.84	-2.20	
H20	C18	H19	111.79	108.08	3.71	
O22	Ni1	N21	79.46	81.11	-1.65	BA 1
Ni1	O22	C2	110.61	110.84	-0.24	CR 1
H23	O22	C2	104.17	109.23	-5.06	
H23	O22	Ni1	120.49	121.78	-1.29	
H24	N21	Ni1	110.12	108.58	1.55	
H24	N21	C16	106.42	107.86	-1.44	
H24	N21	C18	107.22	108.00	-0.78	
N44	Ni1	N21	79.10	85.35	-6.25	BA 2
N44	Ni1	O22	134.27	159.31	-25.04	
H26	C25	O45	104.97	108.29	-3.32	
C27	C25	O45	109.35	112.02	-2.67	
C27	C25	H26	112.24	109.13	3.12	
C25	C27	C30	108.91	109.89	-0.98	Cyclohexyl 2
H28	C27	C30	110.83	110.22	0.62	
H28	C27	C25	107.09	108.91	-1.82	
H29	C27	C30	111.18	110.60	0.58	
H29	C27	C25	108.61	109.66	-1.04	
H29	C27	H28	110.10	107.53	2.57	
H31	C30	C27	108.49	109.24	-0.75	
H32	C30	C27	109.89	109.60	0.29	
H32	C30	H31	109.14	106.61	2.53	
C33	C30	C27	108.66	111.57	-2.91	Cyclohexyl 2
C33	C30	H31	109.23	109.40	-0.17	
C33	C30	H32	111.38	110.30	1.08	
H34	C33	C30	109.28	109.29	-0.01	
C36	C33	C30	108.93	111.95	-3.02	Cyclohexyl 2
C36	C33	H34	108.28	109.06	-0.78	
H35	C33	C30	111.08	110.17	0.91	
H35	C33	H34	109.38	106.69	2.69	
H35	C33	C36	109.84	109.53	0.31	
C39	C36	C33	108.75	110.16	-1.41	Cyclohexyl 2
H37	C36	C33	110.08	109.62	0.46	
H37	C36	C39	108.24	110.01	-1.78	
C39	N44	Ni1	113.14	111.45	1.69	CR 3
N44	C39	C36	112.20	116.30	-4.10	
H38	C36	C33	111.56	110.15	1.40	
H38	C36	C39	108.82	109.61	-0.79	
H38	C36	H37	109.33	107.25	2.08	
C41	N44	Ni1	108.22	104.29	3.93	CR 2
C41	N44	C39	114.57	121.86	-7.28	
C39	C25	O45	103.89	106.49	-2.60	CR 3

Atom A	Atom B	Atom C	BPA / °	GBA / °	BPA-GBA	Comment
C39	C25	H26	111.17	110.13	1.04	
C39	C25	C27	114.43	110.74	3.69	Cyclohexyl 2
C25	C39	C36	112.65	110.01	2.64	Cyclohexyl 2
C25	C39	N44	107.71	110.53	-2.81	CR 3
H40	C39	C36	111.75	108.28	3.47	
H40	C39	N44	104.06	104.56	-0.50	
H40	C39	C25	108.01	106.57	1.43	
C41	C18	N21	106.31	108.56	-2.24	CR 2
C41	C18	H19	110.28	109.88	0.40	
C41	C18	H20	111.79	110.12	1.66	
C18	C41	N44	104.87	106.97	-2.10	CR 2
H42	C41	N44	109.60	111.97	-2.37	
H42	C41	C18	112.01	109.82	2.20	
H43	C41	N44	108.18	109.68	-1.50	
H43	C41	C18	110.66	110.44	0.22	
H43	C41	H42	111.25	107.98	3.27	
O48	Ni1	N21	124.55	101.56	22.99	
O48	Ni1	O22	70.37	79.81	-9.45	
O48	Ni1	N44	89.90	87.70	2.20	
O45	Ni1	N21	132.88	159.67	-26.79	
O45	Ni1	O22	142.11	116.29	25.82	
O45	Ni1	N44	78.78	80.38	-1.60	BA 3
Ni1	O45	C25	104.03	105.14	-1.11	CR 3
O45	Ni1	O48	96.42	92.31	4.11	
H46	O45	C25	104.05	109.47	-5.42	
H46	O45	Ni1	113.89	116.91	-3.02	
H47	N44	Ni1	109.41	107.31	2.10	
H47	N44	C39	106.07	105.90	0.17	
H47	N44	C41	105.01	105.17	-0.16	
O48	H17	C16	22.74	114.23	-91.49	H-bond
H17	O48	Ni1	73.17	75.96	-2.80	H-bond
H49	O48	Ni1	110.06	112.19	-2.13	
H49	O48	H17	104.67	90.47	14.20	H-bond
H50	O48	Ni1	113.77	117.67	-3.89	
H50	O48	H17	146.25	150.57	-4.33	H-bond
H50	O48	H49	103.38	106.01	-2.63	
O51	Ni1	N21	81.63	84.37	-2.74	
O51	Ni1	O22	84.93	87.66	-2.73	
O51	Ni1	N44	130.60	106.58	24.02	
O51	Ni1	O48	137.50	165.04	-27.54	
O51	Ni1	O45	82.11	85.89	-3.78	
O51	H26	C25	43.41	115.21	-71.80	H-bond
H26	O51	Ni1	62.40	77.91	-15.51	H-bond
H52	O51	Ni1	115.70	116.87	-1.16	
H52	O51	H26	88.19	85.78	2.41	H-bond
H53	O51	Ni1	122.47	123.50	-1.03	
H53	O51	H26	78.84	69.56	9.28	H-bond
H53	O51	H52	102.87	105.58	-2.71	

Table S3E: *cis*-[Ni(Cyp$_2$EN)(H$_2$O)$_2$]$^{2+}$

Atom A	Atom B	Atom C	BPA / °	GBA / °	BPA-GBA	Comment
O2	Ni1	N11	68.94	82.82	-13.87	BA 1
O3	Ni1	N11	104.36	92.79	11.57	
O3	Ni1	O2	80.51	83.56	-3.05	
H4	O3	Ni1	115.19	118.35	-3.16	
H5	O3	Ni1	113.30	115.35	-2.05	
H5	O3	H4	103.82	106.34	-2.52	
H6	O2	Ni1	116.74	119.02	-2.28	:
O9	Ni1	N11	110.85	98.29	12.56	
O9	Ni1	O2	119.72	101.00	18.72	
O9	Ni1	O3	143.78	168.44	-24.66	
O7	Ni1	N11	167.46	177.03	-9.56	
O7	Ni1	O2	98.89	94.21	4.68	
O7	Ni1	O3	75.59	86.85	-11.26	
O7	Ni1	O9	71.91	82.25	-10.34	
H29	O7	Ni1	117.32	120.05	-2.72	
H8	O7	Ni1	115.48	117.02	-1.54	
H8	O7	H29	104.23	106.76	-2.53	
N10	Ni1	N11	74.58	83.46	-8.88	BA 2
N10	Ni1	O2	143.07	165.96	-22.89	
N10	Ni1	O3	103.57	94.14	9.43	
N10	Ni1	O9	78.50	83.94	-5.44	BA 3
N10	Ni1	O7	117.80	99.50	18.30	
H21	O3	Ni1	81.25	78.82	2.43	H-bond
H21	O3	H4	140.64	147.90	-7.26	H-bond
H21	O3	H5	100.99	87.21	13.78	H-bond
H15	O3	Ni1	81.27	78.43	2.83	H-bond
H15	O3	H4	110.06	100.24	9.82	H-bond
H15	O3	H5	132.34	137.13	-4.79	H-bond
H15	O3	H21	33.84	54.32	-20.48	H-bond
C19	O2	Ni1	106.49	106.67	-0.17	CR 1
C19	O2	H6	104.94	110.07	-5.13	
C12	N10	Ni1	109.39	108.48	0.91	CR 2
C13	N11	Ni1	107.95	104.93	3.01	CR 2
C13	C12	N10	107.68	110.72	-3.05	CR 2
C12	C13	N11	105.59	107.94	-2.36	CR 2
H14	C13	N11	109.54	111.98	-2.44	
H14	C13	C12	111.29	109.71	1.59	
C13	H15	O3	32.36	113.47	-81.10	H-bond
H15	C13	N11	106.44	107.88	-1.44	
H15	C13	C12	111.77	111.07	0.70	
H15	C13	H14	111.88	108.27	3.60	
H16	C12	N10	106.82	108.86	-2.04	
H16	C12	C13	109.95	108.79	1.15	
H17	C12	N10	108.73	110.65	-1.91	
H17	C12	C13	112.20	110.25	1.95	
H17	C12	H16	111.24	107.48	3.76	
C18	N11	Ni1	108.76	105.31	3.46	CR 1

Atom A	Atom B	Atom C	BPA / °	GBA / °	BPA-GBA	Comment
C18	N11	C13	113.69	117.94	-4.26	
C30	C18	N11	114.78	120.13	-5.35	
C18	C19	O2	104.44	107.63	-3.18	CR 1
C19	C18	N11	106.36	108.41	-2.05	CR 1
C19	C18	C30	106.51	102.46	4.05	Cyclopentyl 1
H20	C19	O2	104.39	108.10	-3.71	
H20	C19	C18	110.90	109.41	1.48	
C18	H21	O3	34.26	114.59	-80.32	H-bond
H21	C18	N11	106.13	107.21	-1.07	
H21	C18	C30	113.24	109.52	3.72	
H21	C18	C19	109.57	108.65	0.92	
C22	O9	Ni1	108.17	107.85	0.32	CR 3
C23	C22	O9	104.41	107.78	-3.38	CR 3
C35	C23	C22	107.11	102.79	4.32	Cyclopentyl 2
C23	N10	Ni1	107.29	103.13	4.16	CR 3
C23	N10	C12	111.66	116.32	-4.66	
N10	C23	C22	107.67	111.09	-3.42	CR 3
N10	C23	C35	115.30	121.18	-5.89	
H24	H16	C12	25.58	119.05	-93.46	CH-HC bond
H24	C22	O9	104.15	108.00	-3.85	
H24	C22	C23	112.42	110.87	1.54	
C22	H24	H16	10.36	112.11	-101.75	CH-HC bond
H25	O9	Ni1	116.34	118.27	-1.93	
H25	O9	C22	104.75	109.92	-5.17	
H26	N10	Ni1	112.96	110.70	2.25	
H26	N10	C12	107.97	109.28	-1.32	
H26	N10	C23	107.62	108.77	-1.14	
H27	N11	Ni1	112.42	111.11	1.31	
H27	N11	C13	107.40	108.88	-1.48	
H27	N11	C18	106.71	108.55	-1.84	
H28	C23	C22	108.97	107.58	1.39	
H28	C23	C35	112.39	108.29	4.09	
H28	C23	N10	105.21	105.32	-0.10	
H45	C30	C18	111.90	112.56	-0.66	
H42	C34	C31	111.00	111.09	-0.09	
H33	C31	C34	112.77	112.87	-0.09	
C31	C19	O2	113.21	117.21	-4.00	
C31	C19	C18	109.04	103.47	5.56	Cyclopentyl 1
C31	C19	H20	114.38	110.72	3.66	
C19	C31	C34	102.55	102.93	-0.38	Cyclopentyl 1
C19	C31	H33	111.95	112.94	-0.99	
H32	C31	C34	111.21	110.95	0.26	
H32	C31	H33	110.80	108.07	2.73	
H32	C31	C19	107.16	109.01	-1.86	
C34	C30	C18	102.97	103.60	-0.63	Cyclopentyl 1
C30	C34	C31	103.26	106.68	-3.42	Cyclopentyl 1
C34	C30	H45	112.87	112.68	0.19	
C30	C34	H42	110.48	110.43	0.05	
C36	C22	O9	112.92	116.70	-3.78	

Atom A	Atom B	Atom C	BPA / °	GBA / °	BPA-GBA	Comment
C36	C22	C23	108.57	102.98	5.59	Cyclopentyl 2
C36	C22	H24	113.97	110.38	3.59	
C41	C35	C23	102.66	103.31	-0.65	Cyclopentyl 2
H37	C36	C22	106.97	108.90	-1.93	
H38	C36	C22	111.88	112.85	-0.97	
H38	C36	H37	110.83	108.06	2.77	
C41	C36	C22	102.76	103.17	-0.41	Cyclopentyl 2
C36	C41	C35	103.37	106.70	-3.33	Cyclopentyl 2
C41	C36	H37	111.22	110.93	0.29	
C41	C36	H38	112.78	112.87	-0.09	
H39	C35	C23	109.00	110.79	-1.79	
H39	C35	C41	110.67	110.41	0.26	
H40	C35	C23	111.07	111.95	-0.88	
H40	C35	C41	112.93	112.78	0.15	
H40	C35	H39	110.25	107.62	2.63	
H43	C34	C31	110.59	110.47	0.12	
H43	C34	H42	110.16	106.94	3.22	
H43	C34	C30	111.19	111.29	-0.09	
H44	C30	C18	107.61	109.59	-1.98	
H44	C30	H45	110.26	107.71	2.56	
H44	C30	C34	110.92	110.69	0.23	
H46	C41	C35	110.50	110.47	0.03	
H46	C41	C36	110.95	111.07	-0.12	
H47	C41	C35	111.17	111.28	-0.10	
H47	C41	C36	110.51	110.44	0.07	
H47	C41	H46	110.18	106.94	3.24	

Table S4. Topological Properties of Ni(II) Complexes with Amino-alcohol Ligands as Determined at the X3LYP/6-311++G(d,p) Level of Theory of the X3LYP/6-31+G(d,p) Energy-Minimized Structures. Atom Numbering is Given in Figure S2

***cis*-[Ni(BHEEN)(H_2O)$_2$]$^{2+}$**		**(Gas phase)**					
Coordination sphere			***ρ***	$\nabla^2\rho$	***V***	***G***	***H***
	Ligand	**BCP**					
	BHEEN	Ni1 N11	0.078351	0.332142	-0.107438	0.095237	-0.012201
		Ni1 N10	0.073175	0.303350	-0.097094	0.086466	-0.010628
		Average:	***0.075763***	***0.317746***	***-0.102266***	***0.090851***	***-0.011415***
		Ni1 O2	0.051654	0.276383	-0.070963	0.070029	-0.000934
		Ni1 O9	0.052219	0.292695	-0.073251	0.073212	-0.000038
		Average:	***0.051936***	***0.284539***	***-0.072107***	***0.071621***	***-0.000486***
	H_2O	Ni1 O3	0.045356	0.256447	-0.062939	0.063525	0.000586
		Ni1 O7	0.052259	0.309869	-0.075851	0.076659	0.000808
		Average:	***0.048807***	***0.283158***	***-0.069395***	***0.070092***	***0.000697***
		Average (Ni-O):	***0.050372***	***0.283848***	***-0.070751***	***0.070857***	***0.000106***
		Average all bonds to Ni:	***0.058836***	***0.295147***	***-0.081256***	***0.077521***	***-0.003735***
		RCP					
	(N,N) chelate	Ni1 N10 C12 C13 N11	0.022327	0.111710	-0.020318	0.024123	0.003805
	(N,O) chelate	Ni1 O9 C24 C25 N10	0.019787	0.103860	-0.018480	0.022222	0.003743
		Ni1 O2 C19 C18 N11	0.020663	0.108889	-0.019331	0.023277	0.003945
		Average(N,O chelate):	0.020225	0.106374	-0.018906	0.022750	0.003844
		Average all RCPs:	***0.020925***	***0.108153***	***-0.019376***	***0.023207***	***0.003831***
Weak interactions							
		BCP					
	O···H-C (2.745 Å)	O3···H23C18	0.006883	0.026058	-0.004632	0.005573	0.000941
		RCP					

		Ni1 O3 H23 C18 N11	0.006866	0.027769	-0.004821	0.005882	0.001061

***cis*-[Ni(BHEEN)(H_2O)$_2$]$^{2+}$**		**(Solvent)**					
Coordination sphere			***ρ***	$\nabla^2\rho$	***V***	***G***	***H***
	Ligand	**BCP**					
	BHEEN	Ni1 N11	0.079486	0.338848	-0.110136	0.097424	-0.012712
		Ni1 N10	0.075138	0.314181	-0.101214	0.089879	-0.011334
		Average:	***0.077312***	***0.326514***	***-0.105675***	***0.093652***	***-0.012023***
		Ni1 O2	0.053962	0.292294	-0.075227	0.074150	-0.001077
		Ni1 O9	0.055296	0.311028	-0.078538	0.078148	-0.000391
		Average:	***0.054629***	***0.301661***	***-0.076882***	***0.076149***	***-0.000734***
	H_2O	Ni1 O3	0.052716	0.304613	-0.075406	0.075780	0.000374
		Ni1 O7	0.054768	0.318197	-0.079274	0.079411	0.000138
		Average:	***0.053742***	***0.311405***	***-0.077340***	***0.077596***	***0.000256***
		Average (Ni-O):	***0.054185***	***0.306533***	***-0.077111***	***0.076872***	***-0.000239***
		Average all bonds to Ni:	**0.061894**	**0.313193**	**-0.086632**	**0.082465**	**-0.004167**
		RCP					
	(N,N) chelate	Ni1 N10 C12 C13 N11	0.022858	0.115330	-0.021046	0.024939	0.003893
	(N,O) chelate	Ni1 O9 C24 C25 N10	0.020364	0.105032	-0.019198	0.022728	0.003530
		Ni1 O2 C19 C18 N11	0.021130	0.109371	-0.019916	0.023629	0.003714
		Average(N,O chelate):	***0.020747***	***0.107201***	***-0.019557***	***0.023179***	***0.003622***
		Average all RCPs:	***0.021450***	***0.109911***	***-0.020053***	***0.023766***	***0.003712***
Weak interactions		**BCP**					
	O···H-C (2.654 Å)	O3···H23C18	0.007524	0.030083	-0.005242	0.006381	0.001140

O···H-C (2.631 Å)	O3···H15C13		0.008210	0.030784	-0.005520	0.006608	0.001088
		Average:	***0.007867***	***0.030433***	***-0.005381***	***0.006495***	***0.001114***
	RCP						
	Ni1 O3 H23 C18 N11		0.008092	0.034151	-0.005884	0.007211	0.001327
	Ni1 O3 H15 C13 N11		0.007517	0.031366	-0.005400	0.006621	0.001221
		Average:	***0.007804***	***0.032759***	***-0.005642***	***0.006916***	***0.001274***

***trans*-[Ni(BHEEN)(H_2O)$_2$]$^{2+}$**	**(Gas Phase)**						
Coordination Sphere			***ρ***	$\nabla^2\rho$	***V***	***G***	***H***
Ligand	**BCP**						
BHEEN	Ni1 N4		0.078614	0.329208	-0.107046	0.094674	-0.012372
	Ni1 N5		0.078614	0.329208	-0.107046	0.094674	-0.012372
		Average:	***0.078614***	***0.329208***	***-0.107046***	***0.094674***	***-0.012372***
	Ni O2		0.046849	0.230644	-0.061268	0.059465	-0.001804
	Ni1 O3		0.046849	0.230644	-0.061268	0.059465	-0.001804
		Average:	***0.046849***	***0.230644***	***-0.061268***	***0.059465***	***-0.001804***
H2O	Ni1 O28		0.049679	0.282189	-0.069579	0.070063	0.000484
	Ni1 O29		0.049679	0.282188	-0.069579	0.070063	0.000484
		Average:	***0.049679***	***0.282189***	***-0.069579***	***0.070063***	***0.000484***
		Average (Ni-O):	***0.048264***	***0.256416***	***-0.065424***	***0.064764***	***-0.000660***
		Average all bonds to Ni:	***0.058381***	***0.280680***	***-0.079298***	***0.074734***	***-0.004564***
	RCP						
(N,N) chelate	Ni1 N4 C7 C6 N5		0.022703	0.113302	-0.020825	0.024575	0.003750
(N,O) chelate	Ni1 O2 C21 C20 N4		0.020093	0.103671	-0.018466	0.022192	0.003726
	Ni1 O3 C11 C8 N5		0.020093	0.103671	-0.018466	0.022192	0.003726
		Average(N,O chelate):	***0.020093***	***0.103671***	***-0.018466***	***0.022192***	***0.003726***
		Average all RCPs:	***0.020963***	***0.106881***	***-0.019252***	***0.022986***	***0.003734***

***trans*-[Ni(BHEEN)(H_2O)$_2$]$^{2+}$**		**(Solvent)**					
Coordination sphere			***ρ***	$\nabla^2\rho$	***V***	***G***	***H***
	Ligand	**BCP**					
	BHEEN	Ni1 N4	0.078664	0.336382	-0.108713	0.096404	-0.012309
		Ni1 N5	0.078699	0.337026	-0.108871	0.096563	-0.012307
		Average:	***0.078682***	***0.336704***	***-0.108792***	***0.096484***	***-0.012308***
		Ni O2	0.052148	0.265678	-0.069895	0.068157	-0.001738
		Ni1 O3	0.052117	0.265498	-0.069855	0.068115	-0.001740
		Average:	***0.052132***	***0.265588***	***-0.069875***	***0.068136***	***-0.001739***
	H_2O	Ni1 O28	0.053595	0.310313	-0.076508	0.077043	0.000535
		Ni1 O29	0.053829	0.312740	-0.077027	0.077606	0.000579
		Average:	***0.053712***	***0.311527***	***-0.076767***	***0.077325***	***0.000557***
		Average (Ni-O):	***0.052922***	***0.288557***	***-0.073321***	***0.072730***	***-0.000591***
		Average all bonds to Ni:	***0.061509***	***0.304606***	***-0.085145***	***0.080648***	***-0.004497***
		RCP					
	(N,N) chelate	Ni1 N4 C7 C6 N5	0.023131	0.115845	-0.021506	0.025234	0.003728
	(N,O) chelate	Ni1 O2 C21 C20 N4	0.020806	0.105901	-0.019389	0.022932	0.003543
		Ni1 O3 C11 C8 N5	0.020802	0.105837	-0.019393	0.022926	0.003533
		Average(N,O chelate):	***0.020804***	***0.105869***	***-0.019391***	***0.022929***	***0.003538***
		Average all RCPs:	***0.021580***	***0.109195***	***-0.020096***	***0.023697***	***0.003601***

***cis*-[Ni(Cy_2EN)(H_2O)$_2$]$^{2+}$**		**(Gas Phase)**					
Coordination Sphere			***ρ***	$\nabla^2\rho$	***V***	***G***	***H***
	Ligand	**BCP**					
	Cy_2EN	Ni1 N7	0.076067	0.318788	-0.102920	0.091308	-0.011611
		Ni1 N6	0.077123	0.324317	-0.104899	0.092989	-0.011910
		Average:	***0.076595***	***0.321552***	***-0.103910***	***0.092149***	***-0.011761***

		Ni1 O2	0.053685	0.286127	-0.074004	0.072768	-0.001236
		Ni1 O5	0.051352	0.302907	-0.074285	0.075006	0.000721
		Average:	***0.052519***	***0.294517***	***-0.074145***	***0.073887***	***-0.000258***
	H_2O	Ni1 O3	0.043928	0.242294	-0.060231	0.060402	0.000171
		Ni1 O4	0.054673	0.311543	-0.078068	0.077977	-0.000091
		Average:	***0.049300***	***0.276918***	***-0.069150***	***0.069190***	***0.000040***
		Average (Ni-O):	***0.050910***	***0.285718***	***-0.071647***	***0.071538***	***-0.000109***
		Average all bonds to Ni:	***0.059471***	***0.297663***	***-0.082401***	***0.078408***	***-0.003993***
		RCP					
	(N,N) chelate	Ni1 N6 C8 C9 N7	0.021855	0.108799	-0.019927	0.023563	0.003636
	(N,O) chelate	Ni1 O2 C11 C10 N7	0.020729	0.107136	-0.019298	0.023041	0.003743
		Ni1 O4 C12 C13 N6	0.019901	0.103373	-0.018593	0.022218	0.003625
		Average(N,O chelate):	***0.020315***	***0.105254***	***-0.018946***	***0.022630***	***0.003684***
		Average all RCPs:	***0.020828***	***0.106436***	***-0.019273***	***0.022941***	***0.003668***
		RCP					
	Cyclohexyl moieties	C10 C11 C19 C20 C21 C18	0.018483	0.111074	-0.016589	0.022179	0.005590
		C12 C13 C14 C16 C17 C15	0.018525	0.111090	-0.016622	0.022197	0.005575
		Average:	***0.018504***	***0.111082***	***-0.016606***	***0.022188***	***0.005583***
Weak interactions		BCP					
	O···HC (2.686 Å)	O3···H40C10	0.007327	0.025494	-0.004737	0.005555	0.000818
	CH···HC (2.343 Å)	H41···H30	0.007060	0.022974	-0.003867	0.004805	0.000938
		RCP					
		Ni1 O3 H40 C10 N7	0.006998	0.028702	-0.004908	0.006042	0.001134
		Ni1 O4 C12 H30 H41 C9 N7	0.005994	0.024795	-0.004009	0.005104	0.001095
		N6 C8 C9 H41 H30 C12 C13	0.005684	0.026266	-0.003955	0.005261	0.001306
		CCP					

	Ni1 O4 N6 N7 C8 C9 C12 C13 C14 C15 C16 H30 H41	0.005512	0.026129	-0.004145	0.005338	0.001194

***cis*-[Ni(Cy$_2$EN)(H$_2$O)$_2$]$^{2+}$**	**Solvent**					
Coordination Sphere		***ρ***	***∇^{2}ρ***	***V***	***G***	***H***
Ligand	**BCP**					
Cy$_2$EN	Ni1 N7	0.075376	0.315839	-0.101936	0.090448	-0.011488
	Ni1 N6	0.078096	0.328832	-0.106920	0.094564	-0.012356
	Average:	***0.076736***	***0.322336***	***-0.104428***	***0.092506***	***-0.011922***
	O2 Ni1	0.056047	0.303361	-0.078505	0.077173	-0.001332
	O5 Ni1	0.055118	0.323254	-0.080386	0.080600	0.000214
	Average:	***0.055582***	***0.313308***	***-0.079445***	***0.078886***	***-0.000559***
H$_2$O	Ni1 O3	0.056207	0.315925	-0.080131	0.079556	-0.000575
	Ni1 O4	0.051538	0.295442	-0.073321	0.094564	0.021243
	Average:	***0.053872***	***0.305683***	***-0.076726***	***0.087060***	***0.010334***
	Average (Ni-O):	***0.054727***	***0.309496***	***-0.078086***	***0.082973***	***0.004887***
	Average all bonds to Ni:	***0.062063***	***0.313776***	***-0.086866***	***0.086151***	***-0.000716***
	RCP					
(N,N) chelate	Ni1 N6 C8 C9 N7	0.021964	0.109540	-0.020071	0.023728	0.003657
(N,O) chelate	Ni1 O4 C12 C13 N6	0.020293	0.103440	-0.019057	0.022459	0.003401
	Ni1 O2 C11 C10 N7	0.021130	0.107326	-0.019802	0.023317	0.003514
	Average(N,O chelate):	***0.020711***	***0.105383***	***-0.019430***	***0.022888***	***0.003458***
	Average all RCPs:	***0.021129***	***0.106769***	***-0.019644***	***0.023168***	***0.003524***
	RCP					

	Cyclohexyl moieties	C10 C11 C19 C20 C21 C18		0.018484	0.111104	-0.016617	0.022196	0.005580
		C12 C13 C14 C16 C17 C15		0.018540	0.111222	-0.016672	0.022239	0.005567
			Average:	***0.018512***	***0.111163***	***-0.016644***	***0.022217***	***0.005573***
Weak interactions								
		BCP						
	O···HC (2.587 Å)	O3···H40C10		0.008500	0.030262	-0.005562	0.006564	0.001002
	CH···HC (2.303 Å)	H41···H30		0.007606	0.024683	-0.004179	0.005175	0.000996
		RCP						
		Ni1 O3 H40 C10 N7		0.007975	0.033628	-0.005736	0.007071	0.001336
		Ni1 O4 C12 H30 H41 C9 N7		0.006467	0.026792	-0.004347	0.005523	0.001175
		N6 C8 C9 H41 H30 C12 C13		0.005956	0.027896	-0.004209	0.005591	0.001383
		CCP						
		Ni1 O4 N6 N7 C8 C9 C12 C13 C14 C15 C16 H30 H41		0.005832	0.027938	-0.004434	0.005709	0.001275

***trans*-[Ni(Cy$_2$EN)(H$_2$O)$_2$]$^{2+}$**	**(Gas phase)**						
Coordination Sphere			***ρ***	***$\nabla^2\rho$***	***V***	***G***	***H***
Ligand	**BCP**						
Cy$_2$EN	Ni1 N21		0.079018	0.323065	-0.106524	0.093645	-0.012879
	Ni N44		0.077204	0.314338	-0.103113	0.090849	-0.012264
		Average:	***0.078111***	***0.318702***	***-0.104819***	***0.092247***	***-0.012572***
	Ni1 O22		0.048997	0.249918	-0.065613	0.064046	-0.001567
	Ni1 O45		0.054554	0.287209	-0.074847	0.073325	-0.001523
		Average:	***0.051775***	***0.268564***	***-0.070230***	***0.068685***	***-0.001545***
H$_2$O	Ni1 O48		0.051479	0.289061	-0.071773	0.072019	0.000246
	Ni1 O51		0.039568	0.198939	-0.051434	0.050584	-0.000850

		Average:	***0.045524***	***0.244000***	***-0.061604***	***0.061302***	***-0.000302***
		Average (Ni-O):	***0.048649***	***0.256282***	***-0.065917***	***0.064994***	***-0.000923***
		Average all bonds to Ni:	***0.058470***	***0.277088***	***-0.078884***	***0.074078***	***-0.004806***
		RCP					
	(N,N) chelate	Ni1 N21 C18 C41 N44	0.022945	0.113600	-0.020997	0.024699	0.003701
	(N,O) chelate	Ni1 N21 C16 C2 O22	0.020452	0.106660	-0.018967	0.022816	0.003849
		Ni1 N44 C39 C25 O45	0.020966	0.109927	-0.019642	0.023562	0.003920
		Average(N,O chelate):	***0.020709***	***0.108293***	***-0.019305***	***0.023189***	***0.003884***
		Average all RCPs:	***0.021454***	***0.110062***	***-0.019869***	***0.023692***	***0.003823***
	Cyclohexyl moieties	C2 C4 C7 C10 C13 C16	0.018454	0.111075	-0.016571	0.022170	0.005599
		C25 C27 C30 C33 C36 C39	0.018512	0.111102	-0.016618	0.022197	0.005579
		Average:	***0.018483***	***0.111088***	***-0.016594***	***0.022183***	***0.005589***
Weak interactions	bond length	**BCP**					
	O···HC (2.701Å)	O51···H26C25	0.007106	0.026895	-0.004783	0.005753	0.000970
		RCP					
		Ni1 O45 C25 H26 O51	0.007089	0.028081	-0.004986	0.006003	0.001017

***trans*-[Ni(Cy_2EN)(H_2O)$_2$]$^{2+}$**	**(Solvent)**					
Coordination Sphere		***ρ***	$\nabla^2\rho$	***V***	***G***	***H***
Ligand	**BCP**					
Cy_2EN	N21 Ni1	0.078964	0.324317	-0.106870	0.093975	-0.012895
	N44 Ni1	0.076971	0.321123	-0.104349	0.092315	-0.012034
	Average:	***0.077967***	***0.322720***	***-0.105610***	***0.093145***	***-0.012465***

		Ni1 O22	0.054986	0.289163	-0.075519	0.073905	-0.001614
		Ni1 O45	0.054960	0.281406	-0.074519	0.072435	-0.002084
		Average:	***0.054973***	***0.285284***	***-0.075019***	***0.073170***	***-0.001849***
	H_2O	Ni1 O48	0.053676	0.306021	-0.076079	0.076292	0.000213
		Ni1 O51	0.046276	0.251629	-0.063238	0.063073	-0.000165
		Average:	***0.049976***	***0.278825***	***-0.069659***	***0.069682***	***0.000024***
		Average (Ni-O):	***0.052475***	***0.282055***	***-0.072339***	***0.071426***	***-0.000913***
		Average all bonds to Ni:	***0.060972***	***0.295610***	***-0.083429***	***0.078666***	***-0.004763***
		RCP					
	(N,N) chelate	Ni1 N21 C18 C41 N44	0.023023	0.113483	-0.021327	0.024849	0.003522
	(N,O) chelate	Ni1 N21 C16 C2 O22	0.021019	0.106331	-0.019605	0.023094	0.003489
		Ni1 N44 C39 C25 O45	0.020842	0.106905	-0.019482	0.023104	0.003622
		Average(N,O chelate):	***0.020931***	***0.106618***	***-0.019543***	***0.023099***	***0.003556***
		Average all RCPs:	***0.021628***	***0.108907***	***-0.020138***	***0.023682***	***0.003544***
	Cyclohexyl moieties	C2 C4 C7 C10 C13 C16	0.018404	0.110748	-0.016527	0.022107	0.005580
		C25 C27 C30 C33 C36 C39	0.018554	0.111269	-0.016683	0.022250	0.005567
		Average:	***0.018479***	***0.111009***	***-0.016605***	***0.022179***	***0.005574***
Weak interactions		**BCP**					
	O···HC (2.587 Å)	C25-H26···O51	0.009969	0.032762	-0.006677	0.007434	0.000757
	O···HC (2.714 Å)	C16-H17···O48	0.006708	0.025600	-0.004463	0.005431	0.000969
		RCP					
		Ni1 O45 C25 H26 O51	0.009731	0.036786	-0.007310	0.008253	0.000943
		Ni1 N21 C16 H17 O48	0.006706	0.026399	-0.004526	0.005563	0.001037

***cis*-[Ni(Cyp$_2$EN)(H$_2$O)$_2$]$^{2+}$**		**Gas Phase**					
Coordination Sphere			***ρ***	**$\nabla^2\rho$**	***V***	***G***	***H***
	Ligand	**BCP**					
	Cyp$_2$EN	Ni1 N10	0.070815	0.286590	-0.092014	0.081831	-0.010184
		Ni1 N11	0.076848	0.319938	-0.103836	0.091910	-0.011926
		Average:	***0.073831***	***0.303264***	***-0.097925***	***0.086871***	***-0.011055***
		Ni1 O2	0.048445	0.246370	-0.064598	0.063095	-0.001503
		Ni1 O9	0.050051	0.269710	-0.068555	0.067991	-0.000564
		Average:	***0.049248***	***0.258040***	***-0.066577***	***0.065543***	***-0.001033***
	H$_2$O	Ni1 O3	0.045770	0.257150	-0.063392	0.063840	0.000448
		Ni1 O7	0.052074	0.307476	-0.075451	0.076160	0.000709
		Average:	***0.048922***	***0.282313***	***-0.069421***	***0.070000***	***0.000579***
		Average (Ni-O):	***0.049085***	***0.270176***	***-0.067999***	***0.067772***	***-0.000227***
		Average all bonds to Ni:	***0.057334***	***0.281205***	***-0.077974***	***0.074138***	***-0.003836***
		RCP					
	(N,N) chelate	Ni1 N10 C12 C13 N11	0.022221	0.110504	-0.020128	0.023877	0.003749
	(N,O) chelate	Ni1 O2 C19 C18 N11	0.019554	0.098218	-0.018266	0.021410	0.003144
		Ni1 O9 C22 C23 N10	0.018790	0.093875	-0.017615	0.020542	0.002927
		Average(N,O chelate):	***0.019172***	***0.096046***	***-0.017941***	***0.020976***	***0.003035***
		Average all RCPs:	***0.020188***	***0.100866***	***-0.018670***	***0.021943***	***0.003273***
	Cyclopropyl moieties	C18 C19 C31 C34 C30	0.039753	0.225491	-0.045810	0.051091	0.005281
		C22 C23 C35 C41 C36	0.039800	0.224458	-0.045747	0.050931	0.005184
		Average:	***0.039776***	***0.224975***	***-0.045779***	***0.051011***	***0.005232***
Weak interactions							
		BCP					
	O···HC (2.658 Å)	O3···H21C18	0.007825	0.027614	-0.005165	0.006034	0.000869

	O···HC (2.749 Å)	O3···H15C13	0.006900	0.026231	-0.004610	0.005584	0.000974
		RCP					
		Ni1 O3 H21 C18 N11	0.007562	0.031035	-0.005446	0.006602	0.001157
		Ni1 O3 H15 C13 N11	0.006899	0.026897	-0.004674	0.005699	0.001025

***cis*-[Ni(Cyp$_2$EN)(H$_2$O)$_2$]$^{2+}$**	**(Solvent)**					
Coordination Sphere		***ρ***	***$\nabla^2\rho$***	***V***	***G***	***H***
Ligand	**BCP**					
Cyp$_2$EN	Ni1 N11	0.077176	0.321567	-0.104743	0.092567	-0.012176
	Ni1 N10	0.072234	0.294188	-0.094967	0.084257	-0.010710
	Average:	***0.074705***	***0.307878***	***-0.099855***	***0.088412***	***-0.011443***
	Ni1 O2	0.050399	0.258345	-0.067909	0.066248	-0.001662
	Ni1 O9	0.052454	0.282330	-0.072397	0.071490	-0.000907
	Average:	***0.051426***	***0.270337***	***-0.070153***	***0.068869***	***-0.001284***
H$_2$O	O3 Ni1	0.052695	0.304908	-0.075513	0.075870	0.000357
	O7 Ni1	0.055440	0.326713	-0.081079	0.081379	0.000299
	Average:	***0.054067***	***0.315811***	***-0.078296***	***0.078625***	***0.000328***
	Average (Ni-O):	***0.052747***	***0.293074***	***-0.074225***	***0.073747***	***-0.000478***
	Average all bonds to Ni:	***0.060066***	***0.298009***	***-0.082768***	***0.078635***	***-0.004133***
	RCP					
(N,N) chelate	Ni1 N10 C12 C13 N11	0.022581	0.112710	-0.020612	0.024395	0.003783
(N,O) chelate	Ni1 O9 C22 C23 N10	0.019575	0.096331	-0.018642	0.021363	0.002720
	Ni1 O2 C19 C18 N11	0.020183	0.099871	-0.019124	0.022046	0.002922
	Average(N,O chelate):	***0.019879***	***0.098101***	***-0.018883***	***0.021704***	***0.002821***
	Average all RCPs:	***0.020780***	***0.102971***	***-0.019460***	***0.022601***	***0.003142***
Cyclopropyl	C22 C23 C35 C41 C36	0.039744	0.224422	-0.045751	0.050928	0.005177

	moieties						
		C18 C19 C31 C34 C30		0.039682	0.225625	-0.045819	0.051113	0.005294
			Average:	***0.039713***	***0.225023***	***-0.045785***	***0.051021***	***0.005235***
Weak interactions								
		BCP						
	O···HC (2.587 Å)	O3···H21C18		0.008515	0.031459	-0.005735	0.006800	0.001065
	O···HC (2.623 Å)	O3···H15C13		0.008223	0.030688	-0.005502	0.006587	0.001085
	CH···HC (2.140 Å)	C12H16···H24C22		0.010933	0.041787	-0.006730	0.008589	0.001858
		RCP						
		Ni1 O3 H21 C18 N11		0.008273	0.034899	-0.006065	0.007395	0.001330
		Ni1 O3 H15 C13 N11		0.008035	0.034330	-0.005865	0.007224	0.001359
		N10 C12 H16 H24 C22 C23		0.010933	0.042484	-0.006818	0.008720	0.001901

***trans*-[Ni(Cyp$_2$EN)(H$_2$O)$_2$]$^{2+}$**	**Gas Phase**					
Coordination Sphere			***ρ***	***∇²ρ***	***V***	***G***	***H***
	Ligand	**BCP**					
	Cyp2EN	Ni40 N3		0.075216	0.300504	-0.098352	0.086739	-0.011613
		Ni40 N15		0.076587	0.309562	-0.101351	0.089371	-0.011980
			Average:	***0.075902***	***0.305033***	***-0.099851***	***0.088055***	***-0.011797***
		Ni40 O29		0.038405	0.159354	-0.046237	0.043038	-0.003199
		Ni40 O31		0.041494	0.177511	-0.050738	0.047558	-0.003180
			Average:	***0.039950***	***0.168433***	***-0.048487***	***0.045298***	***-0.003189***
	H$_2$O	Ni40 O42		0.053217	0.303184	-0.075117	0.075456	0.000340
		Ni40 O44		0.049499	0.264204	-0.066829	0.066440	-0.000389
			Average:	***0.051358***	***0.283694***	***-0.070973***	***0.070948***	***-0.000025***
			Average (Ni-O):	***0.045654***	***0.226063***	***-0.059730***	***0.058123***	***-0.001607***
			Average all bonds to Ni:	***0.055736***	***0.252387***	***-0.073104***	***0.068100***	***-0.005004***

	RCP					
(N,N) chelate	N3 C34 C33 N15 Ni40	0.022576	0.112268	-0.020559	0.024313	0.003754
(N,O) chelate	C1 C4 N3 Ni40 O31	0.018457	0.087435	-0.016604	0.019231	0.002628
	N15 C16 C18 O29 Ni40	0.017973	0.086865	-0.016065	0.018890	0.002826
	Average(N,O chelate):	***0.018215***	***0.087150***	***-0.016334***	***0.019061***	***0.002727***
	Average all RCPs:	***0.019669***	***0.095523***	***-0.017743***	***0.020812***	***0.003069***
Cyclopropyl moieties	C1 C4 C6 C9 C12	0.039823	0.224907	-0.045810	0.051018	0.005208
	C16 C18 C20 C23 C26	0.039679	0.225328	-0.045707	0.051020	0.005313
	Average:	***0.039751***	***0.225118***	***-0.045759***	***0.051019***	***0.005260***

Table S5. Comparison of the Electron Density at the Bond Critical Points of the Metal-Ligand Bonds in the Gas Phase Structures of *Cis* and *Trans* Complexes of Ni(II) and Three Amino-Alcohol Ligands

	cis **/au**	***trans*** **/au**	***cis – trans*** **/au**	ΔE_d^c**(BSSE)** **(*cis – trans*)** **/kcal mol^{-1}**
ρ_{bcp}**(Ni–N)**				
$[Ni(BHEEN)(H_2O)_2]^{2+}$	0.075763	0.078614	–0.002851	2.595
$[Ni(Cy_2EN)(H_2O)_2]^{2+}$	0.076595	0.078111	–0.001516	3.514
$[Ni(Cyp_2EN)(H_2O)_2]^{2+}$	0.073831	0.075902	–0.002071	8.028
ρ_{bcp}**(Ni–OH)**				
$[Ni(BHEEN)(H_2O)_2]^{2+}$	0.051936	0.046849	0.005087	2.595
$[Ni(Cy_2EN)(H_2O)_2]^{2+}$	0.052519	0.051775	0.000744	3.514
$[Ni(Cyp_2EN)(H_2O)_2]^{2+}$	0.049248	0.039950	0.009298	8.028
ρ_{bcp}**(Ni–OH$_2$)**				
$[Ni(BHEEN)(H_2O)_2]^{2+}$	0.050685	0.047981	0.002704	2.595
$[Ni(Cy_2EN)(H_2O)_2]^{2+}$	0.051231	0.049275	0.001956	3.514
$[Ni(Cyp_2EN)(H_2O)_2]^{2+}$	0.049117	0.044513	0.004604	8.028

Table S6. Differential Stabilization From Weak Energy-Lowering Interactions in *Cis* and *Trans* Ni(II) Complexes of Amino-Alcohol Ligands.*

	δE(*cis – trans*) /kcal mol^{-1}	
	Gas Phase	Solvent
$[Ni(BHEEN)(H_2O)_2]^{2+}$	–1.45	–3.38
$[Ni(Cy_2EN)(H_2O)_2]^{2+}$	–1.20	0.44
$[Ni(Cyp_2EN)(H_2O)_2]^{2+}$	–3.07	

*See Table S3 for full details.

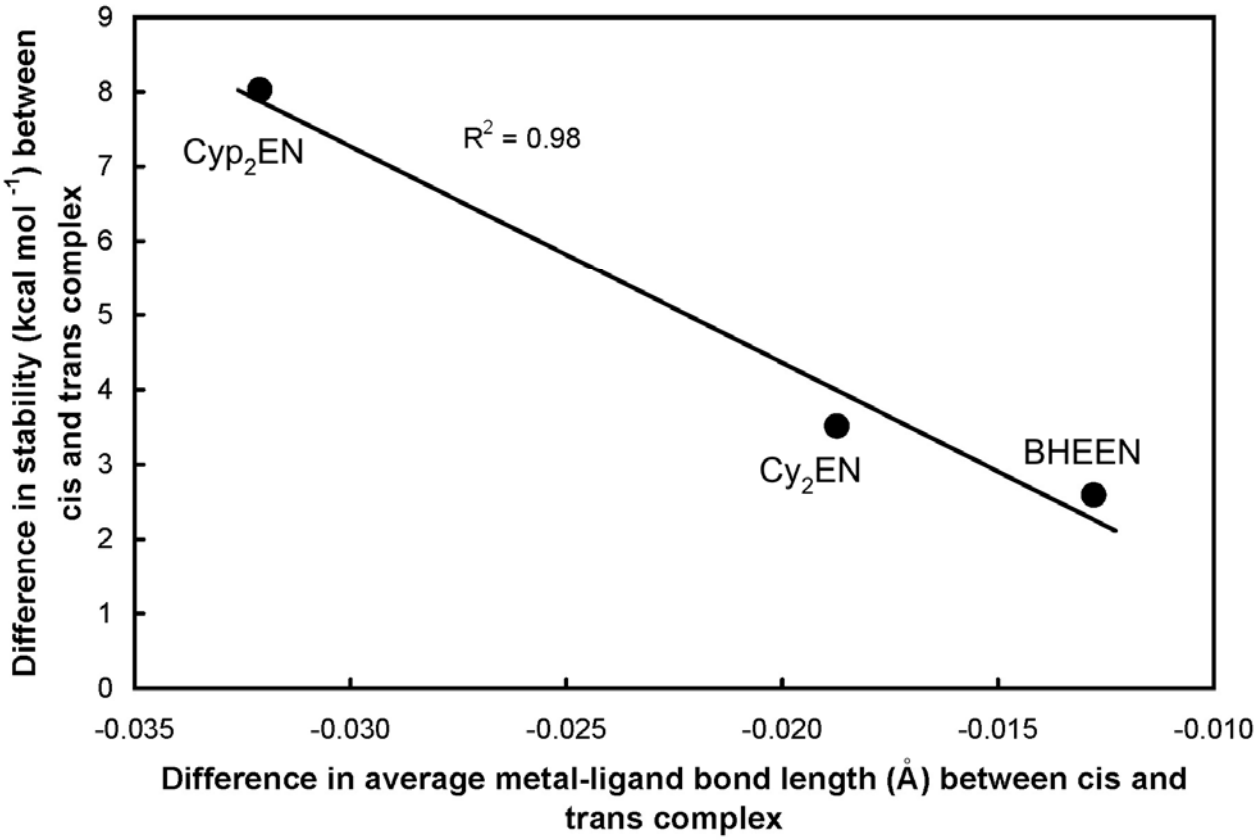
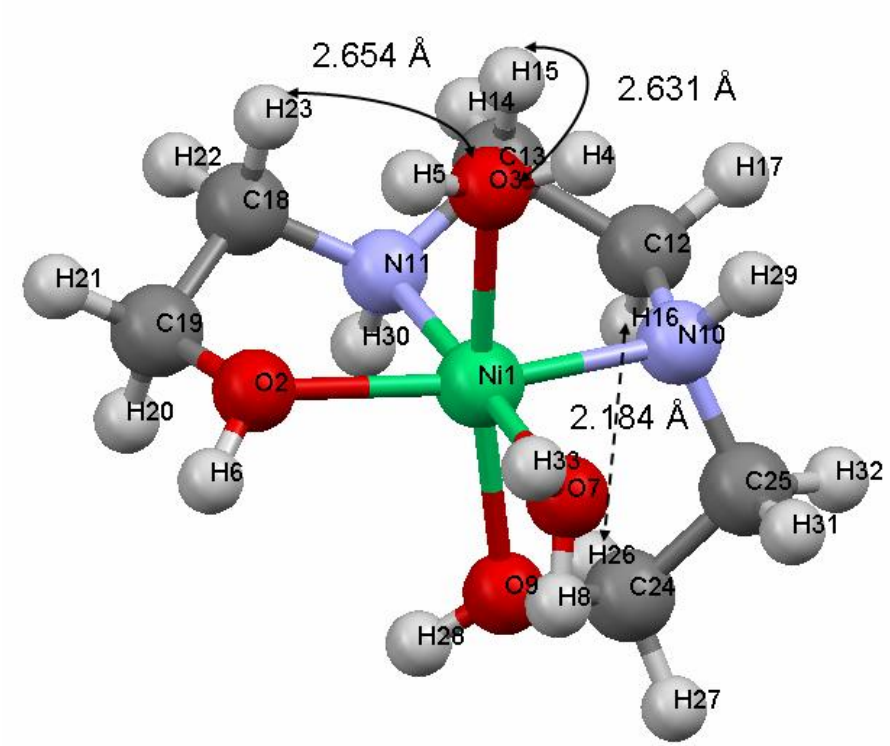
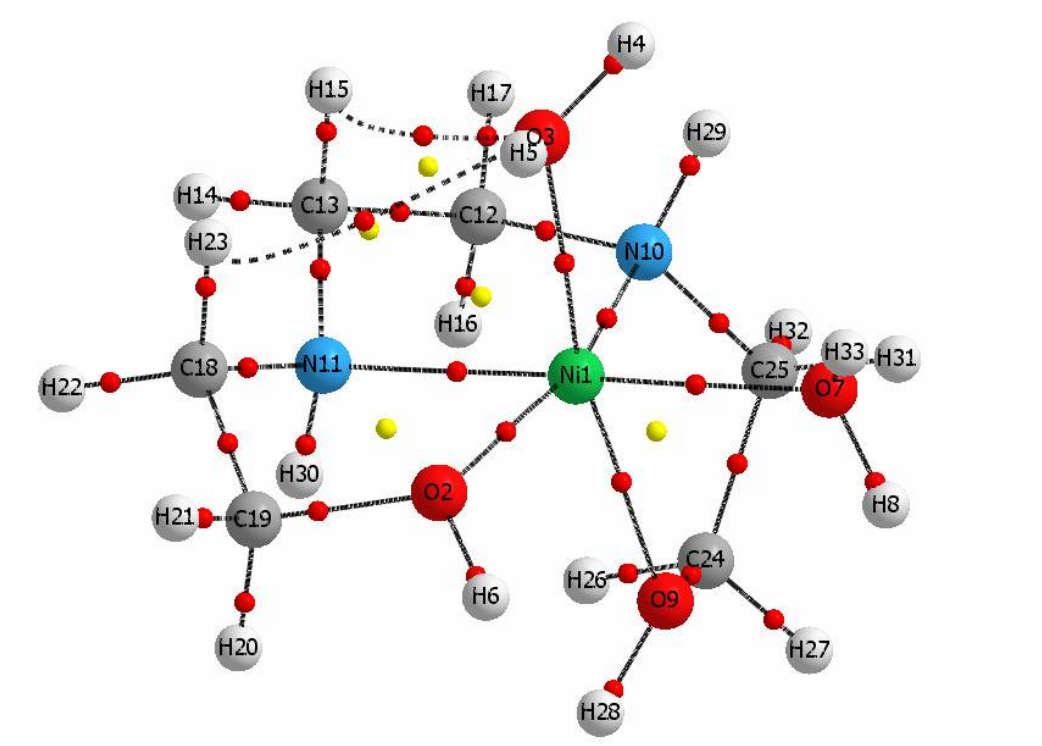


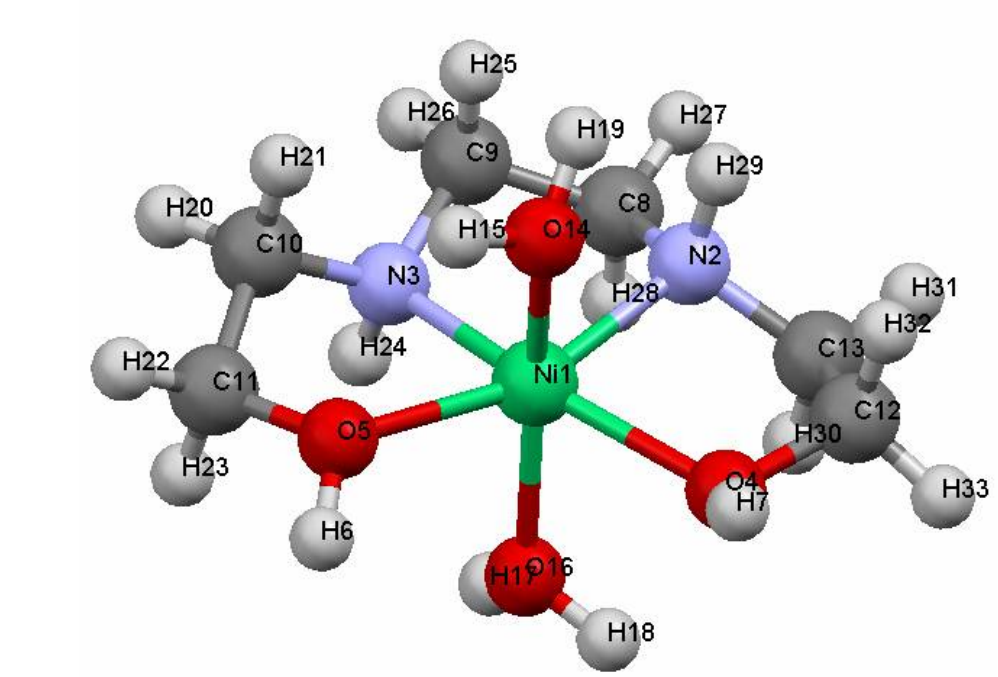
Figure S1. The difference in the stability of the Ni(II) complexes of three amino-alcohol ligands ($E_d{}^c(\mathrm{BSSE})_{cis}$ - $E_d{}^c(\mathrm{BSSE})_{trans}$) correlates inversely with the difference of the *average* metal-ligand bond length (to the four donor atoms of the amino alcohol ligand and the two H_2O ligands) ($\overline{d}_{cis} - \overline{d}_{trans}$). Thus, the greatest difference in the average metal-ligand bond length between the *cis* and *trans* isomers is found when the amino ligand is Cyp_2EN (–0.032 Å) which results in the greatest difference in stability between the *cis* and *trans* complexes (8.1 kcal mol^{-1}).

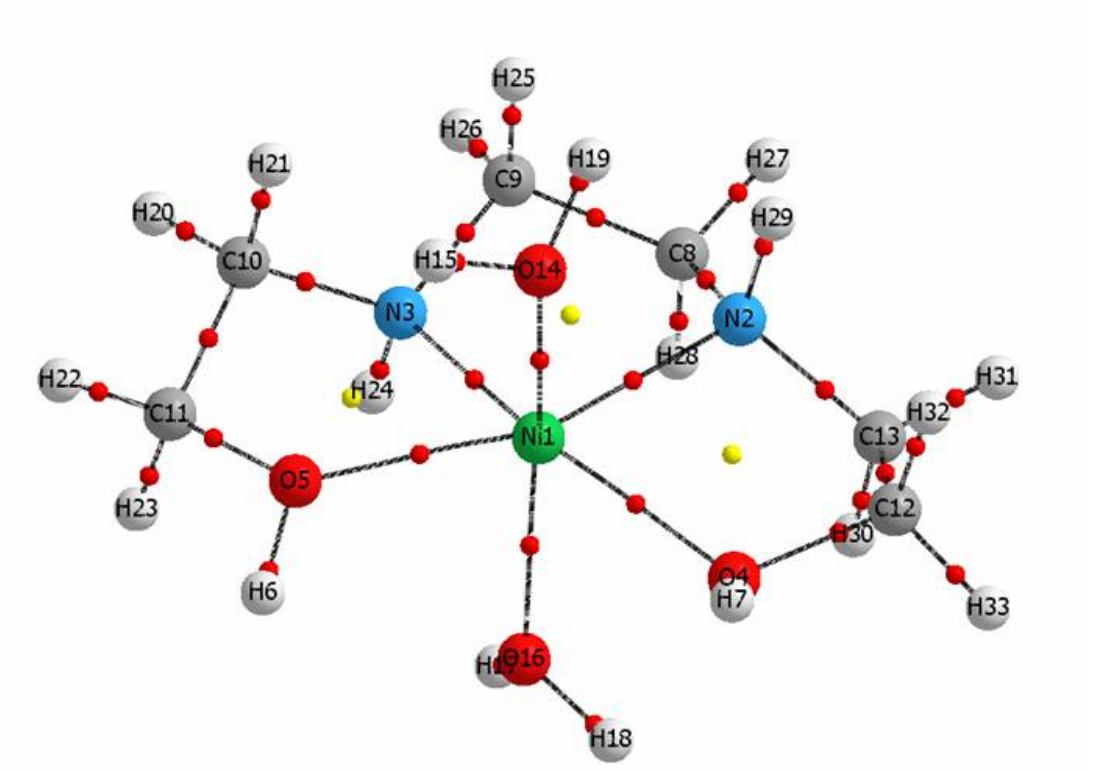
S2A: *cis*-$[Ni(BHEEN)(H_2O)_2]^{2+}$



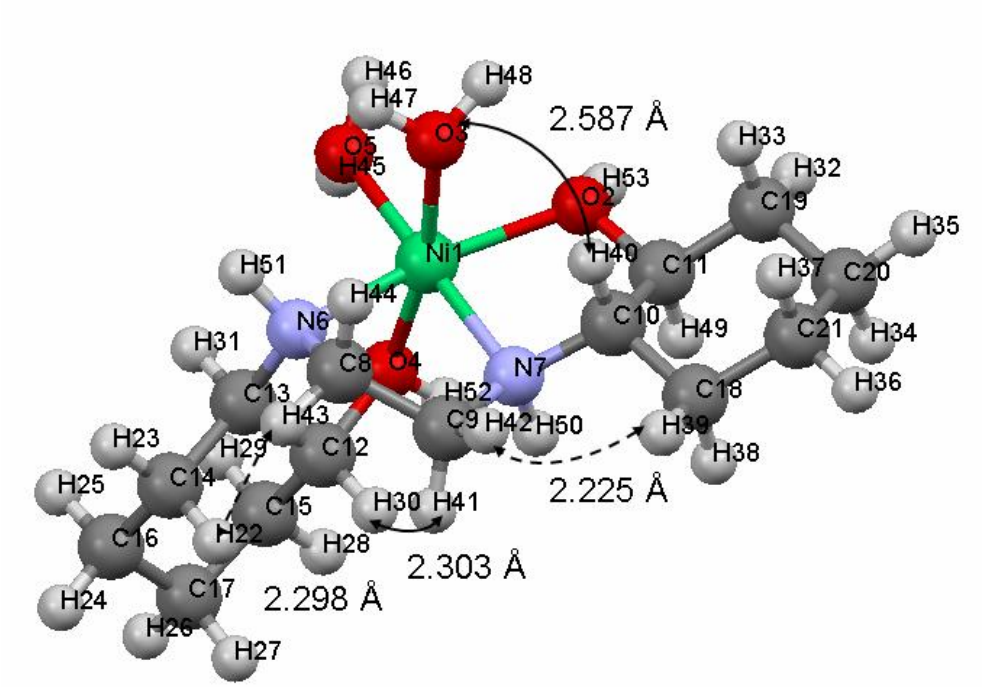


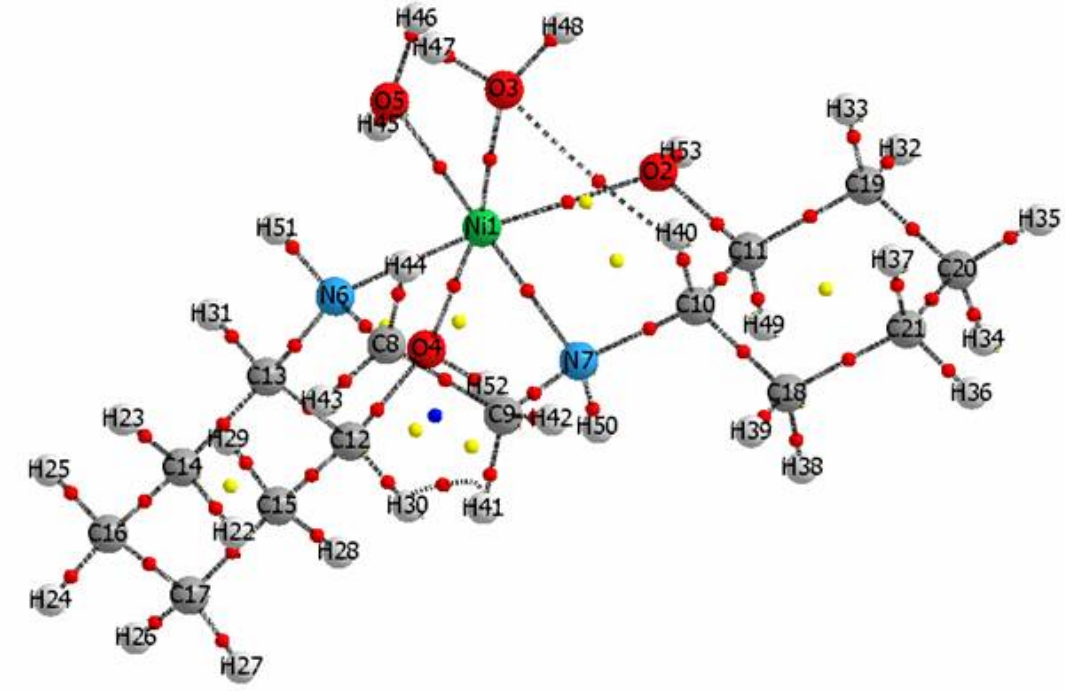
S2B: *trans*-$[Ni(BHEEN)(H_2O)_2]^{2+}$



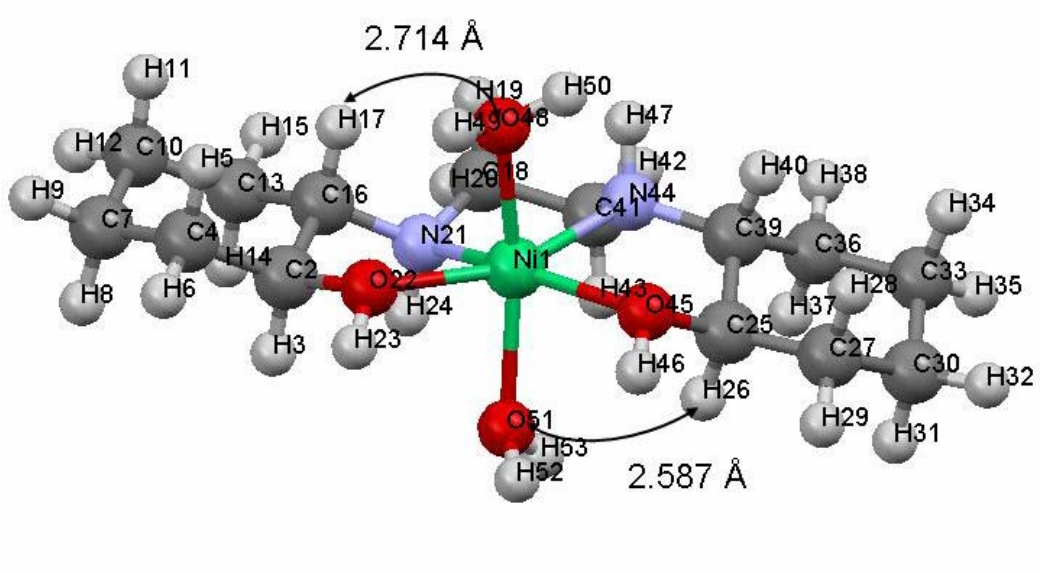


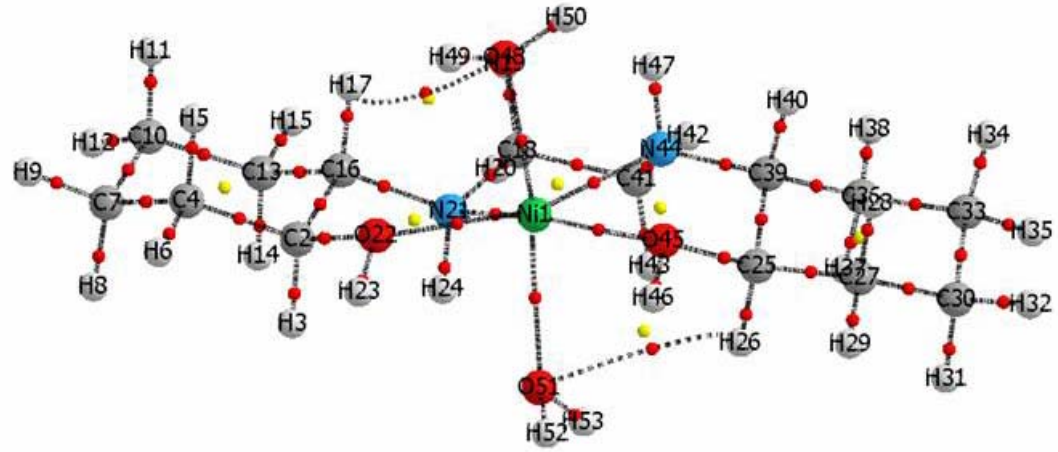
S2C: *cis*-$[Ni(Cy_2EN)(H_2O)_2]^{2+}$



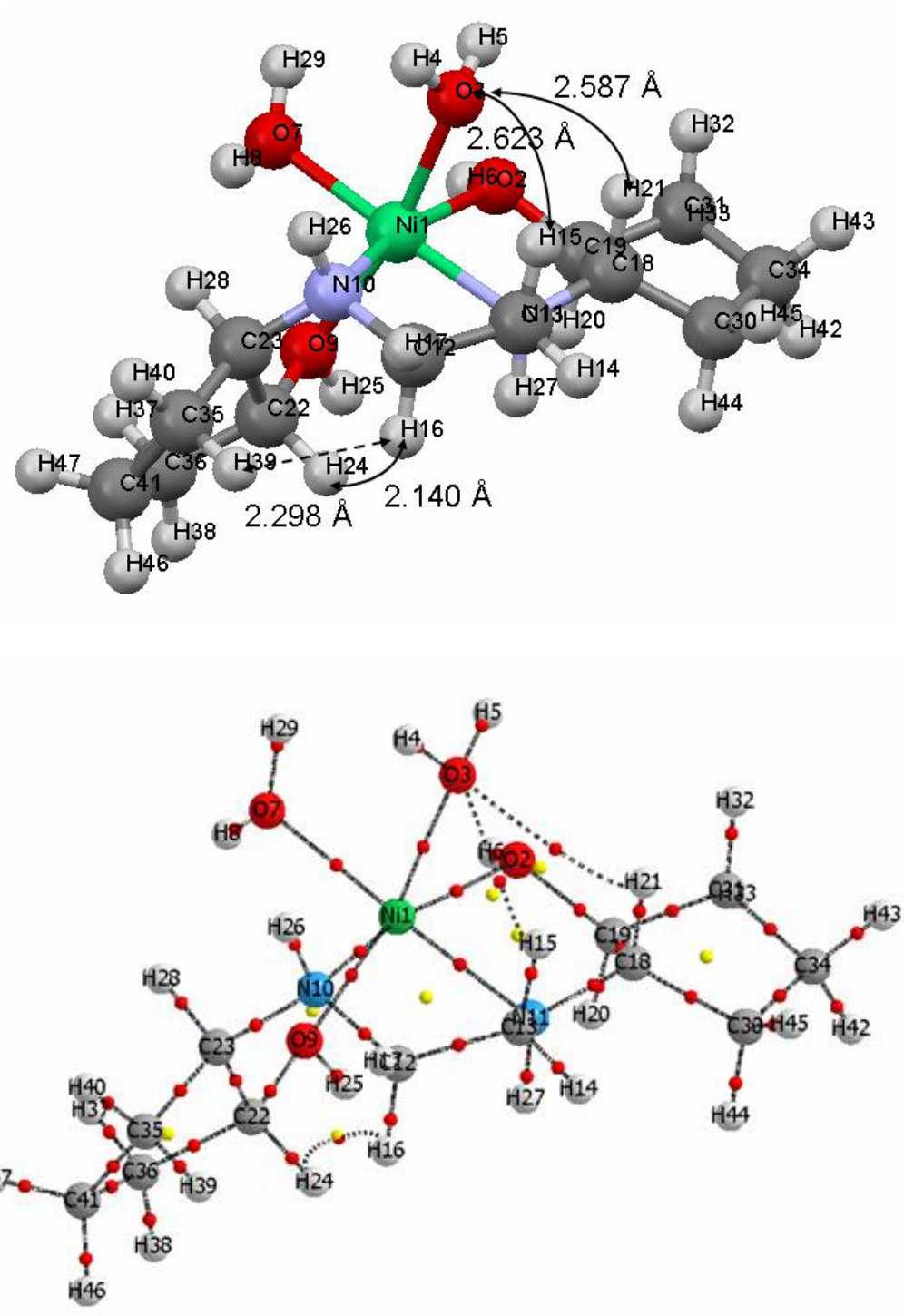


S2D: *trans*-$[Ni(Cy_2EN)(H_2O)_2]^{2+}$





S2E: *cis*-$[Ni(Cyp_2EN)(H_2O)_2]^{2+}$



Figures S2. Structures, showing also atom numbering, and relevant molecular graphs of complexes energy-optimized in solvent.

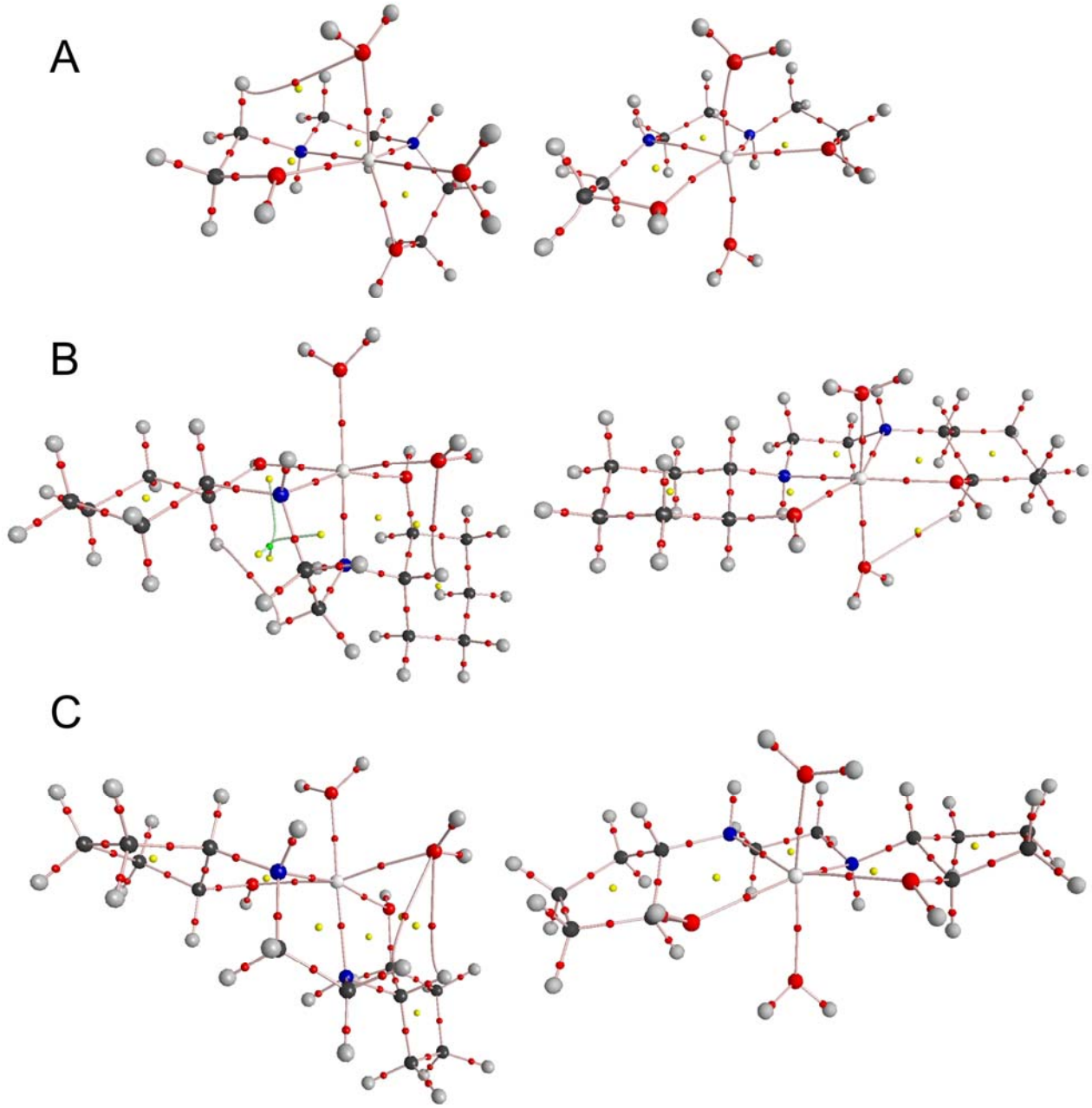


Figure S3. Molecular graphs of (left) the cis and (right) the trans complexes of Ni(II) with amino-alcohol ligands obtained in the gas phase. **A:** $[Ni(BHEEN)(H_2O)_2]^{2+}$; **B:** $[Ni(Cy_2EN)(H_2O)_2]^{2+}$; **C:** $[Ni(Cyp_2EN)(H_2O)_2]^{2+}$. Critical points are shown as small spheres (bcp = red; rcp = yellow; ccp = green). Bond paths are shown as grey tubes connecting the atoms (H = white; C = black; Ni = white; N = blue; O = red). The atom numbering is given in Figure S3.

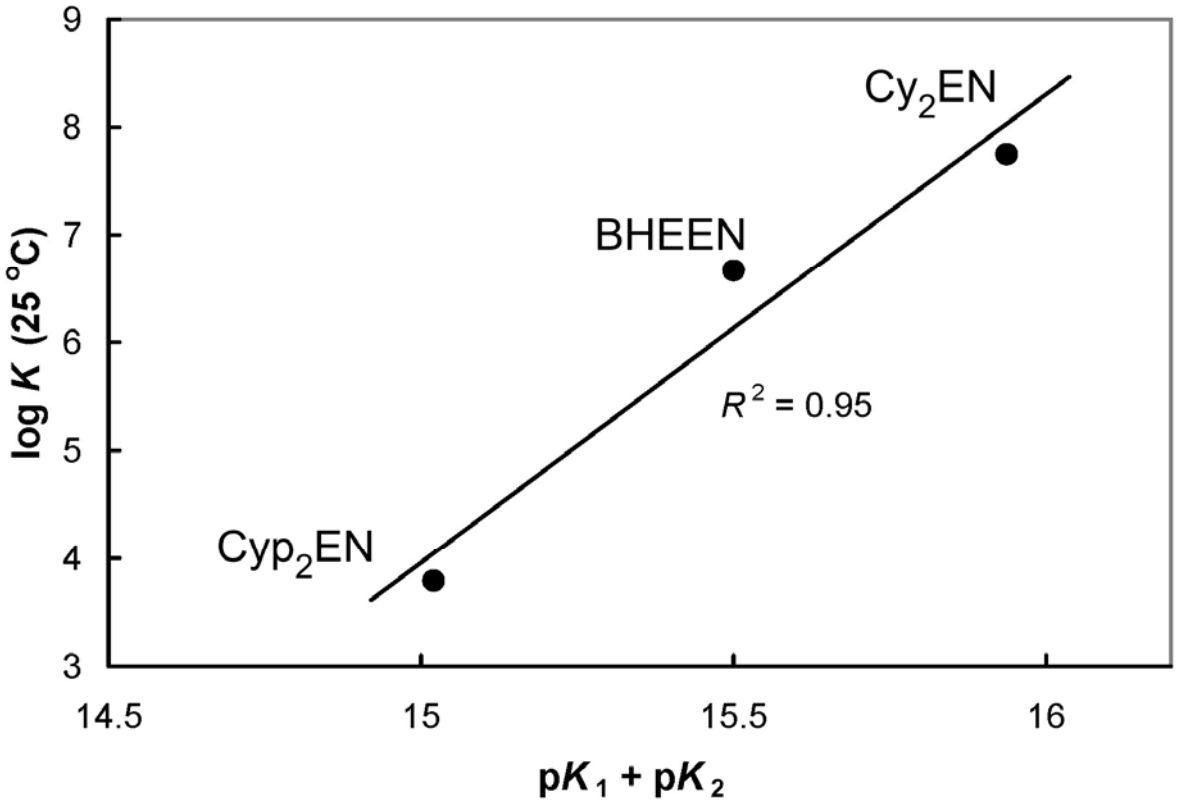


Figure S4. Correlation between log K_1 of the Ni(II) complex with the amino-alcohol ligands Cyp_2EN, BHEEN and Cy_2EN, and the sum of the two protonation constants of the two amino groups of the ligand, a measure of the donor power of the ligand towards a Lewis acid.