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Supporting Information

Title Expanding the Potential of Redox Carriers for Flow Battery Applications

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General Remarks

All syntheses were conducted under atmospheric conditions. The ligand H_2bpb was prepared according to literature procedure.[1] All remaining reagents were purchased from commercial sources and used as received. DMSO-d_6 was dried over activated molecular sieves before use. NMR spectra were obtained on a Bruker Neo 500 spectrometer at room temperature, then processed and analyzed with MestReNova software (v11.0.4-18998). ^{1}H and ^{13}C chemical shifts are reported in parts per million (ppm) relative to TMS, with the residual solvent peak used as an internal reference. NMR multiplicities are reported as follows: singlet (s), doublet (d), triplet (t), quartet (q), multiplet (m), broad signal (br). Coupling constants (*J*) are reported in hertz (Hz). Infrared spectroscopy was performed on a Thermo Scientific Nicolet iS 50 spectrometer using an ATR attachment (diamond crystal).

All electrochemical analyses were carried out in an argon-filled glovebox. The supporting electrolyte was tetrabutylammonium hexafluorophosphate (Sigma, ≥98%), crystallized three times from absolute ethanol. Acetonitrile was used as the solvent and was dried with purification columns from Glasscontour (Laguna Beach, CA) and verified with Karl Fisher titration (Mettler-Toledo C20, using NIST traceable standards). Cyclic voltammetry was performed with a Metrohm PGSTAT204 potentiostat in a three electrode electrochemical cell consisting of a glassy carbon disk working electrode (0.07 cm^2, BASi), a Ag/Ag^+ reference electrode (BASi) containing 0.01 M $AgNO_3$ or $AgBF_4$ in acetonitrile, and a platinum wire counter electrode (23 cm, ALS). All experiments were run at scan rates ranging from 20 to 300 mV/s in an acetonitrile electrolyte containing 1 or 2 mM of analyte and 0.1 M $TBAPF_6$.

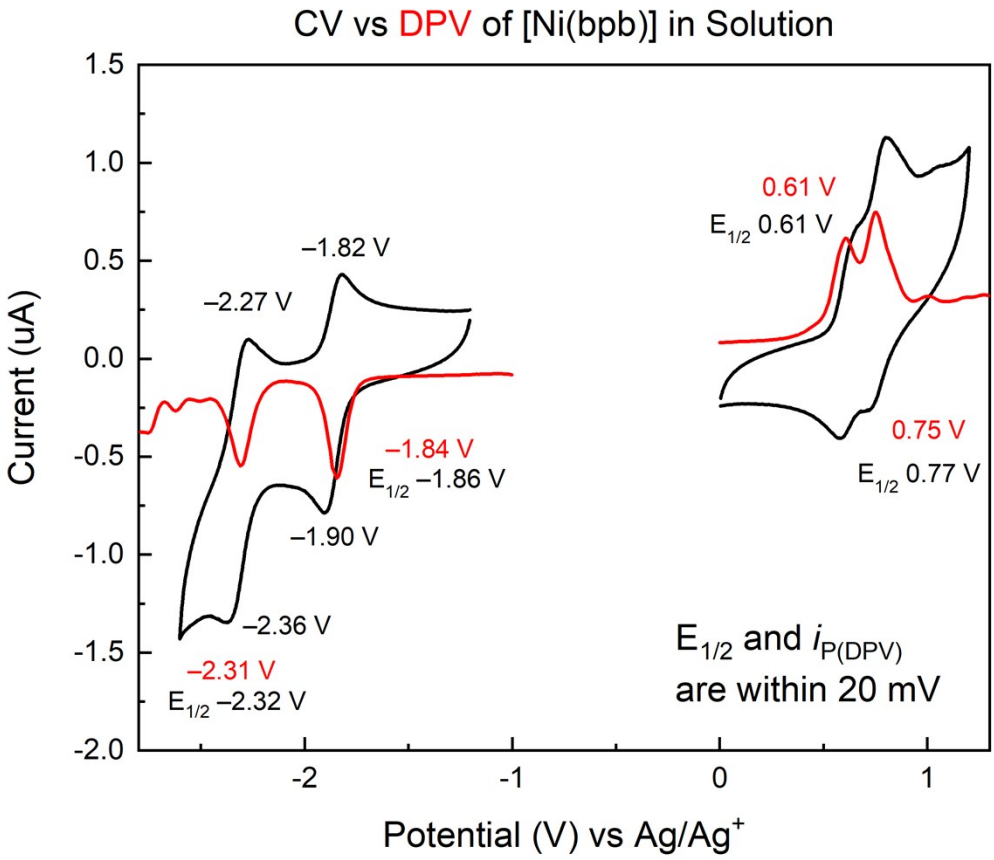


Figure S1. Solution state CV and DPV measurements of [Ni(bpb)] showing that the peak current potential $i_{P(DPV)}$ values obtained from DPV experiments correlate well with the $E_{1/2}$ values from CV measurements. CV experiments were performed on an MeCN solution

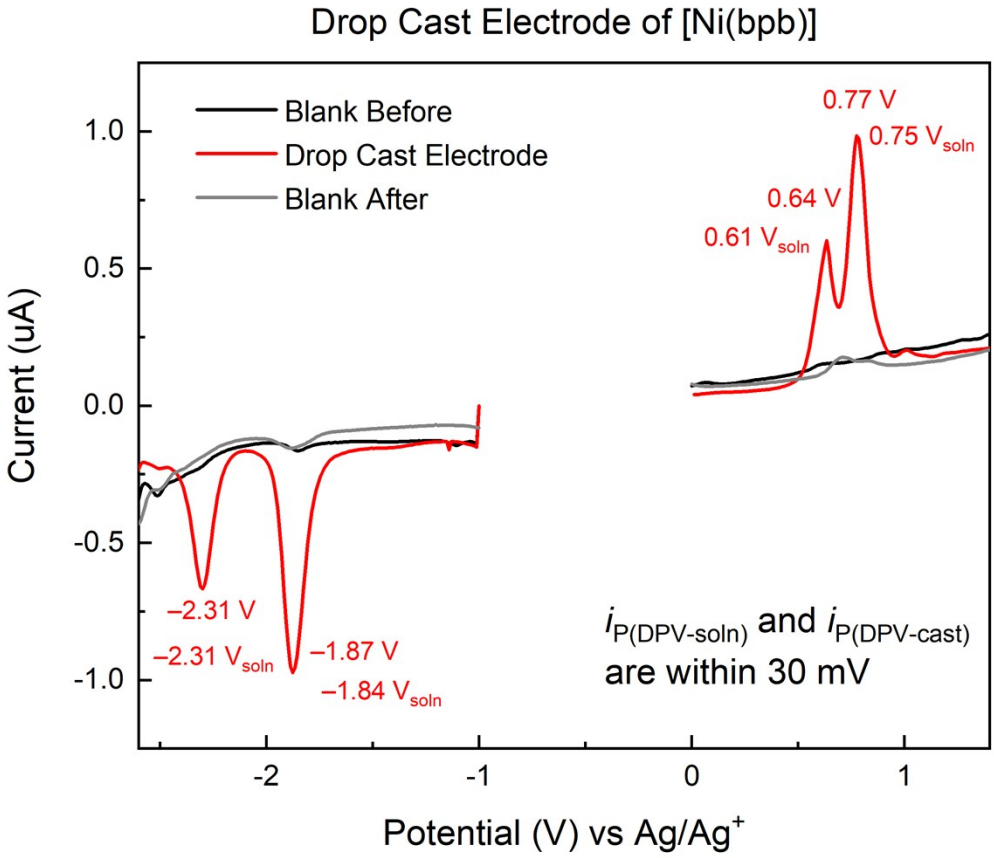


Figure S2. DPV experiments of [Ni(bpb)] drop cast onto a glassy carbon disk electrode. Blank DPVs were obtained prior to and after the drop cast voltammogram, (black and grey trace, respectively) to ensure the measurement was due to the material on the surface. The obtained potentials are within 30 mV of the solution state measurements (V_{soln}) indicating drop cast measurements correlate well with solution state experimental values.

Table S1. Differences in wave spacing (percentage diff from complex **1**).

Differences/Complex	*+ Waves Diff*	*+/- Wave Diff*	*- Wave Diff*	*Max Wave Diff*
[Ni(bpb)], **1**	0.13	2.51	0.39	3.03
[Ni(bpb-NMe_2)], **2**	0.31 (138)	2.81 (12)	0.53 (36)	3.65 (20)
[Ni(bpb-F)], **3**	0.2 (54)	2.68 (7)	0.43 (10)	3.11 (3)
[Ni(bpb-CF_3)], **4**	0.22 (69)	2.93 (20)	0.18 (-54)	3.42 (13)
[Ni(bpb-NO_2)], **5**	0.22 (69)	2.93 (17)	0.34 (-13)	3.49 (15)

Computational Details

Table S2. Selected geometrical parameters of the computed $[Ni(bpb)(MeCN)_2]^0$, $[Ni(bpb)(DMF)_2]^0$, and experimental $[Ni(bpb)(H_2O)]^0$ complexes. Distances are in Å, angles are in °, here and elsewhere.

Parameters	Exp. $[Ni(bpb)(H_2O)]^0$	Theor. $[Ni(bpb)(MeCN)_2]^0$	Theor. $[Ni(bpb)(DMF)_2]^0$
Avg. $Ni\text{-}N_{amide}$	1.85	1.84	1.85
Avg. $Ni\text{-}N_{pyridine}$	1.95	1.93	1.94
Avg. $N_{amide}\text{-}C_{amide}$	1.33	1.35	1.34
Avg. $C_{amide}\text{-}O_{amide}$	1.25	1.22	1.23
Avg. $N_{amide}\text{-}C_{linker}$	1.41	1.40	1.39
$C_{linker}\text{-}C_{linker}$	1.41	1.41	1.41
Avg. $N_{MeCN}\text{-}H_{pyridine}/O_{DMF}\text{-}H_{pyridine}$	-	2.98	2.36
Avg. $H_{MeCN}\text{-}O_{amide}/H_{DMF}\text{-}O_{amide}$	-	2.98	3.03
Avg. $H_{MeCN}\text{-}N_{amide}/H_{DMF}\text{-}N_{amide}$	-	2.91	2.78
Avg. $N_{amide}\text{-}Ni\text{-}N_{pyridine}$	83.8	84.4	84.3
$N_{pyridine}\text{-}Ni\text{-}N_{pyridine}$	108.2	107.0	107.2
$N_{pyridine}\text{-}N_{amide}\text{-}N_{pyridine}\text{-}N_{amide}$	4.2	10.4	9.57

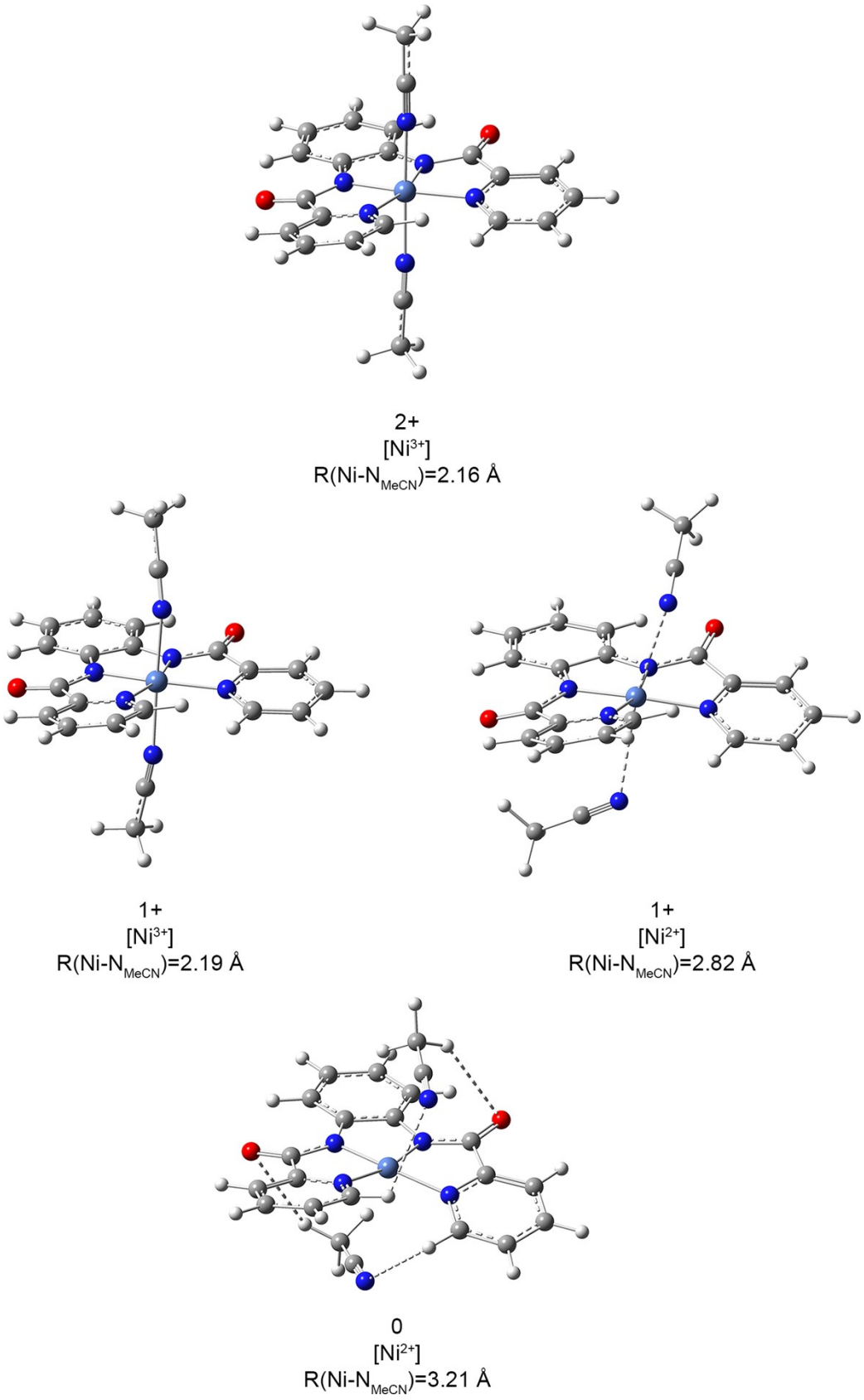
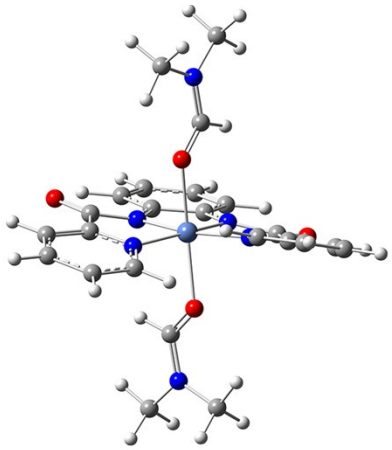


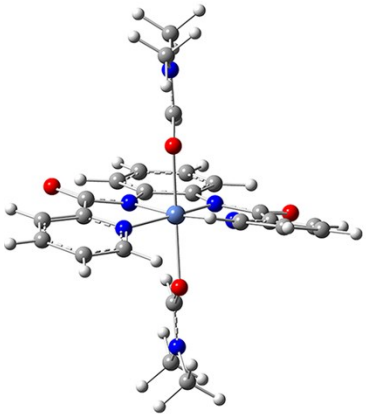
Figure S3. Geometrical structures of the neutral and oxidized $[Ni(bpb)(MeCN)_2]^n$, n=2+, 1+, 0.



2+

[Ni^{3+}]

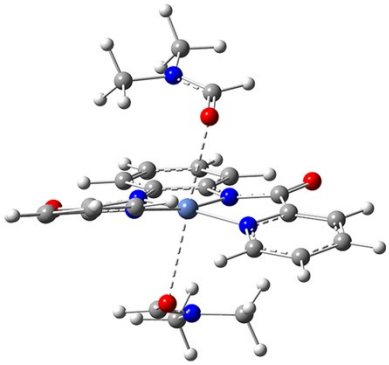
R(Ni-O_{DMF})=2.11 Å



1+

[Ni^{3+}]

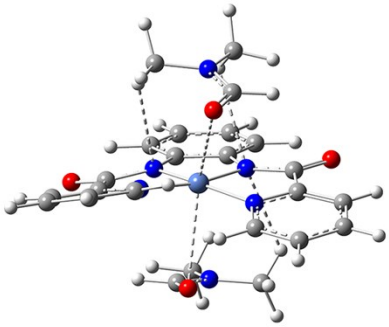
R(Ni-O_{DMF})=2.14 Å



1+

[Ni^{2+}]

R(Ni-O_{DMF})=2.64 Å



0

[Ni^{2+}]

R(Ni-O_{DMF})=2.87-3.53 Å

Figure S4. Geometrical structures of the neutral and oxidized $[Ni(bpb)(DMF)_2]^n$, n=2+, 1+, 0.

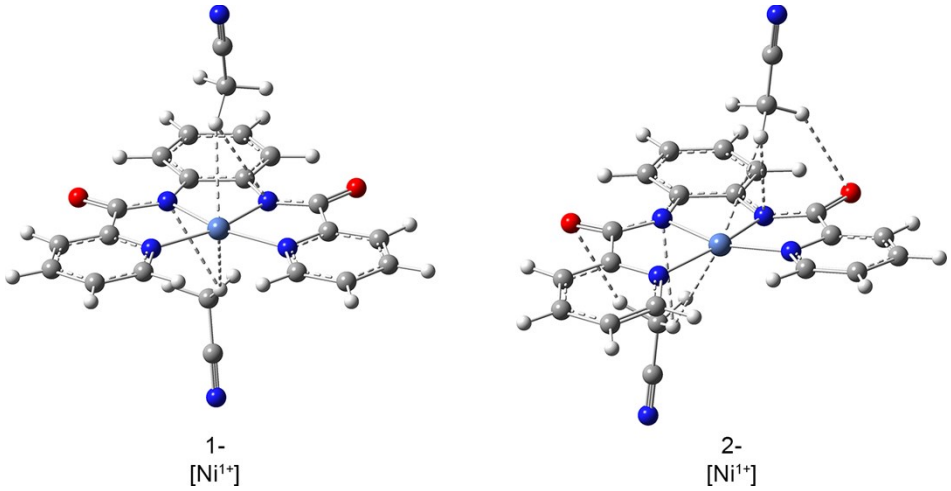


Figure S5. Geometrical structures of the reduced $[Ni(bpb)(MeCN)_2]^n$, n=1-, 2-.

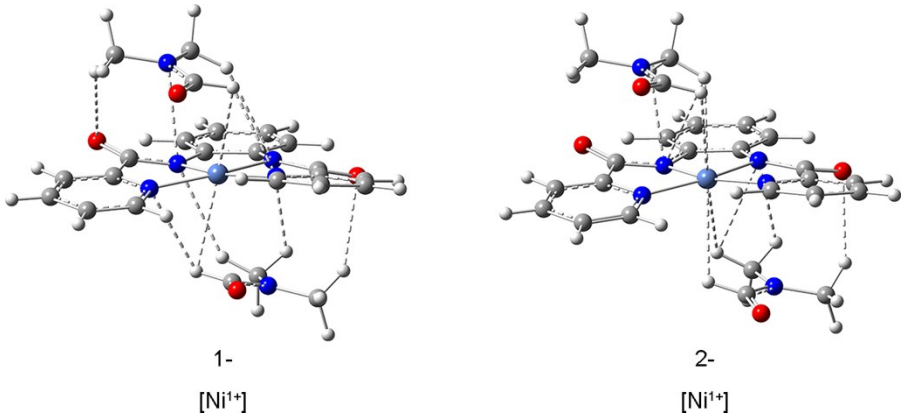


Figure S6. Geometrical structures of the reduced $[Ni(bpb)(DMF)_2]^n$, n=1-, 2-.

Table S3. Computed reduction potentials of complexes [**1-5**]^n (n=2+, 1+, 0, 1-, 2-) in MeCN.

Complex/ reduction step*	**1**	**2**	**3**	**4**	**5**
2+ → 1+	1.02 [L] **1.08 [M]**	**0.98 [M]**	1.04 [L] **1.11 [M]**	**1.16 [M]**	**1.20 [M]**
1+ → 0	0.74 [M] **0.69 [L]**	**0.53 [L]**	0.77 [M] **0.71 [L]**	**0.88 [L]**	**1.01 [L]**
0 → 1−	**−1.73 [M]**	**−2.03 [M]**	**−1.68 [M]**	**−1.64 [M]**	**−1.56 [M]**
1− → 2−	**−2.53 [L]**	**−3.03 [L]**	**−2.51 [L]**	**−2.45 [L]**	**−2.02 [L]**

* [L] or [M] indicates ligand- or metal-centered events, respectively. Values shown in bold correspond to the redox potentials based on the most thermodynamically stable electronic configurations. For the 1+ → 0 and 2+ → 1+ steps, two values of reduction potentials are shown based on the two possible localization sites in the $[Ni(bpb\text{-}R)]^{1+}$ complexes for comparison.

Table S4. Free energies (eV) in acetonitrile of the $[Ni(bpb\text{-}R)(MeCN)_2]^{1+}$ complexes (R=$(NMe_2)_2$, F, CF_3, NO_2) wherein an unpaired electron is localized on the ligand (L) relative to those with the electron localized on the metal (M). Zero-point energies are taken from the gas-phase optimized complexes.

Complexes/ |e| localization site	L [Ni^{2+}]	M [Ni^{3+}]
$[Ni(bpb)(MeCN)_2]^{1+}$	0.00	+0.055
$[Ni(bpb\text{-}(NMe_2)_2)(MeCN)_2]^{1+}$	0.00	N/A†
$[Ni(bpb\text{-}F)(MeCN)_2]^{1+}$	0.00	+0.066
$[Ni(bpb\text{-}CF_3)(MeCN)_2]^{1+}$	0.00	+0.172‡
$[Ni(bpb\text{-}NO_2)(MeCN)_2]^{1+}$	0.00	+0.164‡

† $[Ni(bpb\text{-}(NMe_2)_2)(MeCN)_2]^{1+}$ structure with the Ni^{3+} center could not be obtained in PCM.
‡ These gas-phase optimized $[Ni(bpb\text{-}R)(MeCN)_2]^{1+}$ structures are not local minima at the Ni^{3+} oxidation state, exhibiting 1 imaginary frequency. Following a normal-mode vibration corresponding to the imaginary frequency upon geometry optimization, structures with the Ni^{3+} center rearrange into those with the Ni^{2+} configurations.

Table S5. Free energies (in eV) in acetonitrile of the $[Ni(bpb\text{-}R)(DMF)_2]^{1+}$ complexes (R=$(NMe_2)_2$, F, CF_3, NO_2) wherein an unpaired electron is localized on the ligand (L) relative to those with the electron localized on the metal (M). Zero-point energies are taken from the gas-phase optimized complexes.

Complexes/ |e| localization site	L [Ni^{2+}]	M [Ni^{3+}]
$[Ni(bpb)(DMF)_2]^{1+}$	+0.139	0.00
$[Ni(bpb\text{-}(NMe_2)_2)(DMF)_2]^{1+}$	+0.206	0.00
$[Ni(bpb\text{-}F)(DMF)_2]^{1+}$	+0.137	0.00
$[Ni(bpb\text{-}CF_3)(DMF)_2]^{1+}$	+0.133	0.00
$[Ni(bpb\text{-}NO_2)(DMF)_2]^{1+}$	+0.358	0.00

Table S6. Selected geometrical parameters of the computed $[Ni(bpb)(MeCN)_2]^n$ complexes by charge state *n*.

Parameters/*n*	2+	1+ [Ni^{3+}]	1+ [Ni^{2+}]	0	1-	2-
Avg. Ni-N_{amide}	1.88	1.87	1.85	1.84	2.00	2.00
Avg. Ni-$N_{pyridine}$	1.99	1.99	1.93	1.93	2.06	2.00
Avg. N_{amide}-C_{amide}	1.39	1.35	1.38	1.35	1.33	1.34
Avg. C_{amide}-O_{amide}	1.20	1.22	1.21	1.22	1.24	1.25
Avg. N_{amide}-C_{linker}	1.36	1.40	1.35	1.40	1.38	1.38
C_{linker}-C_{linker}	1.45	1.40	1.45	1.41	1.43	1.44
Avg. direct Ni-N_{MeCN}	2.16	2.19	2.82	3.21	-	-
Avg. H_{MeCN}-O_{amide}	-	-	2.72	2.98	-	2.65
Avg. Ni-H_{MeCN}	-	-	-	3.24	2.70	2.95
Avg. H_{MeCN}-N_{amide}	-	-	3.60	2.91	2.76	2.90
Avg. N_{amide}-Ni-$N_{pyridine}$	83.7	83.6	84.6	84.4	80.9	81.3
$N_{pyridine}$-Ni-$N_{pyridine}$	110.2	109.0	108.0	107.0	117.1	117.5
$N_{pyridine}$-N_{amide}-$N_{pyridine}$-N_{amide}	6.5	5.5	13.1	10.4	0.5	13.5

Table S7. Selected geometrical parameters of the computed $[Ni(bpb)(DMF)_2]^n$ complexes by charge state *n*.

Parameters/*n*	2+	1+ [Ni^{3+}]	1+ [Ni^{2+}]	0	1-	2-
Avg. Ni-N_{amide}	1.87	1.87	1.85	1.85	2.00	2.01
Avg. Ni-$N_{pyridine}$	1.96	1.98	1.93	1.94	2.05	2.01
Avg. N_{amide}-C_{amide}	1.39	1.35	1.38	1.34	1.33	1.34
Avg. C_{amide}-O_{amide}	1.20	1.22	1.21	1.23	1.24	1.25
Avg. N_{amide}-C_{linker}	1.36	1.40	1.35	1.39	1.38	1.38
C_{linker}-C_{linker}	1.45	1.41	1.45	1.41	1.43	1.44
Avg. direct Ni-O_{DMF}	2.11	2.14	2.64	3.20	-	-
Avg. H_{DMF}-O_{amide}	-	-	-	3.03	2.80	2.69
Avg. Ni-H_{DMF}	-	-	-	-	2.71	2.83
Avg. H_{DMF}-N_{amide}	2.73	2.74	2.88	2.78	2.70	2.62
Avg. N_{amide}-Ni-$N_{pyridine}$	83.89	83.76	84.59	84.25	80.92	80.96
$N_{pyridine}$-Ni-$N_{pyridine}$	109.23	108.78	107.88	107.24	116.07	117.68
$N_{pyridine}$-N_{amide}-$N_{pyridine}$-N_{amide}	-7.89	-6.44	11.65	9.57	-4.21	-3.60

Table S8. Mulliken spin densities on selected fragments, oxidation states of Ni, and spin state of the $[Ni(bpb)(MeCN)_2]^n$ complexes by charge state *n*.

Fragment/*n*	2+ (open-shell singlet)	1+ (doublet)	1+ (doublet)	0 (singlet)	1- (doublet)	2- (open-shell singlet)
Oxidation state of Ni	Ni^{3+}	Ni^{3+}	Ni^{2+}	Ni^{2+}	Ni^{1+}	Ni^{1+}
Ni center	1.02	1.05	0.05	0.00	0.89	1.05
Pyridyls	0.00; 0.00	0.01; 0.01	0.00; 0.00	0.00	0.00; 0.00	-0.52; -0.52
$-C_6H_4-$ linker	-0.67	0.00	0.52	0.00	0.00	0.01
Amides	-0.55	-0.17	0.42	0.00	0.10	0.00
MeCNs	0.12	0.11	0.00	0.00	-0.01	-0.01

Table S9. Mulliken spin densities on selected fragments, oxidation states of Ni, and spin state of the $[Ni(bpb)(DMF)_2]^n$ complexes by charge state *n*.

Fragment/*n*	2+ (open-shell singlet)	1+ (doublet)	1+ (doublet)	0 (singlet)	1- (doublet)	2- (open-shell singlet)
Oxidation state of Ni	Ni^{3+}	Ni^{3+}	Ni^{2+}	Ni^{2+}	Ni^{1+}	Ni^{1+}
Ni center	0.91	0.97	0.06	0.00	0.90	0.99
Pyridyls	-0.01; -0.01	0.00; 0.00	0.01; 0.01	0.00	0.01; 0.01	-0.48; -0.47
$-C_6H_4-$ linker	-0.55	0.00	0.43	0.00	0.01	0.00
Amides	-0.53	-0.13	0.47	0.00	0.10	-0.01
DMFs	0.18	0.16	0.02	0.00	-0.02	-0.03

Table S10. Selected geometrical parameters of the computed $[Ni(bpb\text{-}(NMe_2)_2)]^n$ complexes in MeCN by charge state *n*. For the 1- and 2- charge states, MeCN molecules are omitted.

Parameters/*n*	2+	1+ [Ni^{2+}]	0	1-	2-
Avg. Ni-N_{amide}	1.88	1.85	1.85	2.00	2.00
Avg. Ni-$N_{pyridine}$	1.95	1.91	1.92	2.06	2.01
Avg. N_{amide}-C_{amide}	1.39	1.38	1.35	1.32	1.34
Avg. C_{amide}-O_{amide}	1.20	1.21	1.22	1.24	1.25
Avg. N_{amide}-C_{linker}	1.36	1.35	1.40	1.38	1.38
C_{linker}-C_{linker}	1.45	1.45	1.42	1.44	1.44
Avg. direct Ni-N_{MeCN}	2.18	2.95	3.35	-	-
Avg. H_{MeCN}-O_{amide}	-	2.62	3.19	-	-
Avg. Ni-H_{MeCN}	-	-	3.07	-	-
H_{MeCN}-N_{amide}	-	3.33	2.86	-	-
Avg. N_{amide}-Ni-$N_{pyridine}$	83.9	84.7	84.4	80.8	81.0
$N_{pyridine}$-Ni-$N_{pyridine}$	109.3	107.4	106.8	116.9	116.7
$N_{pyridine}$-N_{amide}-$N_{pyridine}$-N_{amide}	6.1	11.0	9.4	0.05	3.7

Table S11. Selected geometrical parameters of the computed $[Ni(bpb\text{-}(NMe_2)_2)]^n$ complexes in DMF by charge state *n*. For the 1- and 2- charge states, DMF molecules are omitted.

Parameters/*n*	2+	1+ [Ni^{3+}]	1+ [Ni^{2+}]	0	1-	2-
Avg. Ni-N_{amide}	1.87	1.87	1.84	1.85	2.00	2.00
Avg. Ni-$N_{pyridine}$	1.94	1.95	1.91	1.93	2.06	2.01
Avg. N_{amide}-C_{amide}	1.39	1.35	1.38	1.34	1.32	1.34
Avg. C_{amide}-O_{amide}	1.21	1.22	1.21	1.23	1.24	1.25
Avg. N_{amide}-C_{linker}	1.36	1.40	1.35	1.39	1.38	1.38
C_{linker}-C_{linker}	1.45	1.41	1.44	1.41	1.44	1.44
Avg. direct Ni-O_{DMF}	2.12	2.16	2.76	3.36	-	-
Avg. H_{DMF}-O_{amide}	-	-	3.19	3.31	-	-
Avg. Ni-H_{DMF}	3.13	3.01	-	-	-	-
H_{DMF}-N_{amide}	2.80	2.73	2.85	2.79	-	-
Avg. N_{amide}-Ni-$N_{pyridine}$	84.0	83.9	84.7	84.2	80.8	81.0
$N_{pyridine}$-Ni-$N_{pyridine}$	108.7	108.3	107.3	107.1	116.9	116.7
$N_{pyridine}$-N_{amide}-$N_{pyridine}$-N_{amide}	-7.0	-5.8	10.4	8.1	0.05	3.7

Table S12. Mulliken spin densities on selected fragments, oxidation states of Ni, and spin state of the $[Ni(bpb\text{-}(NMe_2)_2)]^n$ complexes in MeCN by charge state *n*. For the 1- and 2- charge states, MeCN molecules are omitted.

Fragment/*n*	2+ (open-shell singlet)	1+ (doublet)	0 (singlet)	1- (doublet)	2- (open-shell singlet)
Oxidation state of Ni	Ni^{3+}	Ni^{2+}	Ni^{2+}	Ni^{1+}	Ni^{1+}
Ni center	0.99	0.08	0.00	0.88	1.08
Pyridyls	-0.01; -0.01	0.01; 0.01	0.00	0.01; 0.01	-0.52; -0.53
$-C_6H_4-$ linker	-0.55	0.42	0.00	0.01	0.00
Amides	-0.53	0.48	0.00	0.10	0.00
MeCNs	0.11	0.00	0.00	-	-
$(NMe_2)_2$	0.00	0.00	0.00	0.00	-0.02

Table S13. Mulliken spin densities on selected fragments, oxidation states of Ni, and spin state of the $[Ni(bpb\text{-}(NMe_2)_2)]^n$ complexes in DMF by charge state *n*. For the 1- and 2- charge states, DMF molecules are omitted.

Fragment/*n*	2+ (open-shell singlet)	1+ (doublet)	1+ (doublet)	0 (singlet)	1- (doublet)	2- (open-shell singlet)
Oxidation state of Ni	Ni^{3+}	Ni^{3+}	Ni^{2+}	Ni^{2+}	Ni^{1+}	Ni^{1+}
Ni center	-0.91	0.96	0.09	0.00	0.86	0.84
Pyridyls	0.01; 0.01	0.00; 0.00	0.02	0.00	0.04	-0.38; -0.37
$-C_6H_4-$ linker	0.54	0.00	0.40	0.00	0.01	-0.02
Amides	0.52	-0.11	0.47	0.00	0.10	-0.05
DMFs	-0.17	0.15	0.02	0.00	-	-
$(NMe_2)_2$	0.00	0.00	0.00	0.00	0.00	-0.02

Table S14. Selected geometrical parameters of the computed [Ni(bpb-F)]^n complexes in MeCN by charge state *n*. For the 1- and 2- charge states, MeCN molecules are omitted.

Parameters/*n*	2+	1+ [Ni^{3+}]	1+ [Ni^{2+}]	0	1-	2-
Avg. Ni-N_{amide}	1.88	1.87	1.85	1.84	2.00	2.00
Avg. Ni-$N_{pyridine}$	1.99	1.99	1.93	1.93	2.06	2.00
Avg. N_{amide}-C_{amide}	1.39	1.35	1.37	1.35	1.32	1.34
Avg. C_{amide}-O_{amide}	1.20	1.22	1.21	1.22	1.24	1.25
Avg. N_{amide}-C_{linker}	1.36	1.40	1.35	1.39	1.38	1.38
C_{linker}-C_{linker}	1.45	1.40	1.45	1.41	1.44	1.44
Avg. direct Ni-N_{MeCN}	2.16	2.19	2.82	3.21	-	-
Avg. H_{MeCN}-O_{amide}	-	-	2.75	2.94	-	-
Avg. Ni-H_{MeCN}	-	-	-	3.24	-	-
H_{MeCN}-N_{amide}	-	-	3.66	2.92	-	-
Avg. N_{amide}-Ni-$N_{pyridine}$	83.6	83.5	84.5	84.3	80.8	81.1
$N_{pyridine}$-Ni-$N_{pyridine}$	110.3	109.2	108.2	107.1	116.9	117.0
$N_{pyridine}$-N_{amide}-$N_{pyridine}$-N_{amide}	6.3	5.5	13.0	10.4	0.6	7.6

Table S15. Selected geometrical parameters of the computed [Ni(bpb-F)]^n complexes in DMF by charge state *n*. For the 1- and 2- charge states, DMF molecules are omitted.

Parameters/*n*	2+	1+ [Ni^{3+}]	1+ [Ni^{2+}]	0	1-	2-
Avg. Ni-N_{amide}	1.87	1.87	1.85	1.85	2.00	2.00
Avg. Ni-$N_{pyridine}$	1.96	1.98	1.93	1.94	2.06	2.00
Avg. N_{amide}-C_{amide}	1.39	1.35	1.37	1.34	1.32	1.34
Avg. C_{amide}-O_{amide}	1.20	1.22	1.21	1.23	1.24	1.25
Avg. N_{amide}-C_{linker}	1.35	1.40	1.35	1.39	1.38	1.38
C_{linker}-C_{linker}	1.45	1.41	1.45	1.41	1.44	1.44
Avg. direct Ni-O_{DMF}	2.11	2.14	2.64	3.21	-	-
Avg. H_{DMF}-O_{amide}	-	-	-	3.19	-	-
Avg. Ni-H_{DMF}	3.16	3.00	-	-	-	-
H_{DMF}-N_{amide}	2.74	2.75	3.29	2.83	-	-
Avg. N_{amide}-Ni-$N_{pyridine}$	83.7	83.7	84.4	84.2	80.8	81.1
$N_{pyridine}$-Ni-$N_{pyridine}$	109.4	108.9	108.1	107.4	116.9	117.0
$N_{pyridine}$-N_{amide}-$N_{pyridine}$-N_{amide}	-7.5	-6.4	11.5	9.5	0.6	7.6

Table S16. Mulliken spin densities on selected fragments, oxidation states of Ni, and spin state of the $[Ni(bpb\text{-}F)]^n$ complexes in MeCN by charge state *n*. For the 1- and 2- charge states, MeCN molecules are omitted.

Fragment/*n*	2+ (open-shell singlet)	1+ (doublet)	1+ (doublet)	0 (singlet)	1- (doublet)	2- (open-shell singlet)
Oxidation state of Ni	Ni^{3+}	Ni^{3+}	Ni^{2+}	Ni^{2+}	Ni^{1+}	Ni^{1+}
Ni center	1.02	1.05	0.05	0.00	0.90	1.13
Pyridyls	0.00; 0.00	0.00; 0.00	0.00; 0.00	0.00	-0.01; -0.01	-0.54; -0.61
$-C_6H_4-$ linker	-0.57	0.00	0.49	0.00	0.01	0.01
Amides	-0.52	-0.17	0.45	0.00	0.11	0.01
MeCNs	0.12	0.11	0.00	0.00	-	-
F	-0.04	0.00	0.00	0.00	0.00	0.00

Table S17. Mulliken spin densities on selected fragments, oxidation states of Ni, and spin state of the $[Ni(bpb\text{-}F)]^n$ complexes in DMF by charge state *n*. For the 1- and 2- charge states, DMF molecules are omitted.

Fragment/*n*	2+ (open-shell singlet)	1+ (doublet)	1+ (doublet)	0 (singlet)	1- (doublet)	2- (open-shell singlet)
Oxidation state of Ni	Ni^{3+}	Ni^{3+}	Ni^{2+}	Ni^{2+}	Ni^{1+}	Ni^{1+}
Ni center	0.91	0.96	0.06	0.00	0.87	0.89
Pyridyls	0.00; 0.00	0.00; 0.00	0.01; 0.01	0.00	0.01; 0.01	-0.43; -0.42
$-C_6H_4-$ linker	-0.55	-0.01	0.44	0.00	0.01	-0.01
Amides	-0.50	-0.13	0.46	0.00	0.10	-0.03
DMFs	0.18	0.17	0.02	0.00	-	-
F	-0.03	0.00	0.02	0.00	0.00	0.00

Table S18. Selected geometrical parameters of the computed $[Ni(bpb\text{-}CF_3)]^n$ complexes in MeCN by charge state *n*. For the 1- and 2- charge states, MeCN molecules are omitted.

Parameters/*n*	2+	1+ [Ni^{2+}]	0	1-	2-
Avg. Ni-N_{amide}	1.88	1.85	1.84	2.00	2.00
Avg. Ni-$N_{pyridine}$	1.99	1.93	1.93	2.06	2.00
Avg. N_{amide}-C_{amide}	1.39	1.38	1.35	1.33	1.34
Avg. C_{amide}-O_{amide}	1.20	1.21	1.22	1.24	1.25
Avg. N_{amide}-C_{linker}	1.36	1.35	1.39	1.38	1.38
C_{linker}-C_{linker}	1.44	1.45	1.42	1.44	1.45
Avg. direct Ni-N_{MeCN}	2.15	2.80	3.20	-	-
Avg. H_{MeCN}-O_{amide}	-	2.79	3.00	-	-
Avg. Ni-H_{MeCN}	-	-	3.28	-	-
H_{MeCN}-N_{amide}	-	3.69	2.93	-	-
Avg. N_{amide}-Ni-$N_{pyridine}$	83.6	84.6	84.3	80.9	81.2
$N_{pyridine}$-Ni-$N_{pyridine}$	110.3	108.1	107.2	117.0	117.0
$N_{pyridine}$-N_{amide}-$N_{pyridine}$-N_{amide}	6.5	13.0	10.5	0.01	7.4

Table S19. Selected geometrical parameters of the computed $[Ni(bpb\text{-}CF_3)]^n$ complexes in DMF by charge state *n*. For the 1- and 2- charge states, DMF molecules are omitted.

Parameters/*n*	2+	1+ [Ni^{3+}]	1+ [Ni^{2+}]	0	1-	2-
Avg. Ni-N_{amide}	1.87	1.87	1.96	1.90	2.00	2.00
Avg. Ni-$N_{pyridine}$	1.96	1.97	2.06	2.00	2.06	2.00
Avg. N_{amide}-C_{amide}	1.39	1.36	1.37	1.34	1.33	1.34
Avg. C_{amide}-O_{amide}	1.20	1.22	1.21	1.23	1.24	1.25
Avg. N_{amide}-C_{linker}	1.35	1.40	1.34	1.38	1.38	1.38
C_{linker}-C_{linker}	1.45	1.41	1.46	1.42	1.44	1.45
Avg. direct Ni-O_{DMF}	2.10	2.14	2.24	2.60	-	-
Avg. H_{DMF}-O_{amide}	-	-	-	2.89	-	-
Avg. Ni-H_{DMF}	3.16	3.01	3.14	-	-	-
H_{DMF}-N_{amide}	2.74	2.75	2.91	2.83	-	-
Avg. N_{amide}-Ni-$N_{pyridine}$	83.8	83.7	81.7	83.1	80.9	81.2
$N_{pyridine}$-Ni-$N_{pyridine}$	109.3	108.9	115.5	110.8	117.0	117.0
$N_{pyridine}$-N_{amide}-$N_{pyridine}$-N_{amide}	-7.9	-6.2	0.9	7.8	0.01	7.4

Table S20. Mulliken spin densities on selected fragments, oxidation states of Ni, and spin state of the $[Ni(bpb\text{-}CF_3)]^n$ complexes in MeCN by charge state *n*. For the 1- and 2- charge states, MeCN molecules are omitted.

Fragment/*n*	2+ (open-shell singlet)	1+ (doublet)	0 (singlet)	1- (doublet)	2- (open-shell singlet)
Oxidation state of Ni	Ni^{3+}	Ni^{2+}	Ni^{2+}	Ni^{1+}	Ni^{1+}
Ni center	1.01	0.06	0.00	0.90	1.13
Pyridyls	0.00; 0.00	0.00; 0.00	0.00	0.00; 0.00	-0.51; -0.61
$-C_6H_4-$ linker	-0.56	0.44	0.00	0.01	0.01
Amides	-0.57	0.49	0.00	0.11	0.00
MeCNs	0.12	0.01	0.00	-	-
CF_3	0.00	0.00	0.00	0.00	0.00

Table S21. Mulliken spin densities on selected fragments, oxidation states of Ni, and spin state of the $[Ni(bpb\text{-}CF_3)]^n$ complexes in DMF by charge state *n*. For the 1- and 2- charge states, DMF molecules are omitted.

Fragment/*n*	2+ (open-shell singlet)	1+ (doublet)	1+ (doublet)	0 (singlet)	1- (doublet)	2- (open-shell singlet)
Oxidation state of Ni	Ni^{3+}	Ni^{3+}	Ni^{2+}	Ni^{2+}	Ni^{1+}	Ni^{1+}
Ni center	0.91	0.96	0.01	0.00	0.87	0.89
Pyridyls	0.01; 0.00	0.00; 0.00	0.01; 0.01	0.00	0.01; 0.01	-0.41; -0.41
$-C_6H_4-$ linker	-0.54	0.00	0.44	0.00	0.01	-0.03
Amides	-0.54	-0.13	0.53	0.00	0.10	-0.05
DMFs	0.18	0.17	0.01	0.00	-	-
CF_3	0.00	0.00	0.00	0.00	0.00	0.00

Table S22. Selected geometrical parameters of the computed $[Ni(bpb\text{-}NO_2)]^n$ complexes by charge state *n*. For the 1- and 2- charge states, MeCN molecules are omitted.

Parameters/*n*	2+	1+ [Ni^{2+}]	0	1-	2-
Avg. Ni-N_{amide}	1.88	1.85	1.84	2.00	2.00
Avg. Ni-$N_{pyridine}$	1.99	1.93	1.93	2.06	2.03
Avg. N_{amide}-C_{amide}	1.39	1.38	1.35	1.33	1.36
Avg. C_{amide}-O_{amide}	1.20	1.21	1.22	1.24	1.24
Avg. N_{amide}-C_{linker}	1.35	1.35	1.39	1.38	1.35
C_{linker}-C_{linker}	1.45	1.45	1.42	1.44	1.46
Avg. direct Ni-N_{MeCN}	2.15	2.79	3.19	-	-
Avg. H_{MeCN}-O_{amide}	-	2.96	3.00	-	-
Avg. Ni-H_{MeCN}	-	-	3.31	-	-
H_{MeCN}-N_{amide}	-	-	2.95	-	-
Avg. N_{amide}-Ni-$N_{pyridine}$	83.6	84.6	84.3	81.0	81.4
$N_{pyridine}$-Ni-$N_{pyridine}$	110.4	108.1	107.2	117.1	116.2
$N_{pyridine}$-N_{amide}-$N_{pyridine}$-N_{amide}	6.5	13.0	10.7	0.00	0.4

Table S23. Selected geometrical parameters of the computed $[Ni(bpb\text{-}NO_2)]^n$ complexes in DMF by charge state *n*. For the 1- and 2- charge states, DMF molecules are omitted.

Parameters/*n*	2+	1+ [Ni^{3+}]	1+ [Ni^{2+}]	0	1-	2-
Avg. Ni-N_{amide}	1.87	1.87	1.85	1.84	2.00	2.00
Avg. Ni-$N_{pyridine}$	1.96	1.97	1.93	1.94	2.06	2.03
Avg. N_{amide}-C_{amide}	1.39	1.36	1.39	1.35	1.33	1.36
Avg. C_{amide}-O_{amide}	1.20	1.21	1.21	1.22	1.24	1.24
Avg. N_{amide}-C_{linker}	1.35	1.40	1.35	1.38	1.38	1.35
C_{linker}-C_{linker}	1.45	1.41	1.45	1.42	1.44	1.46
Avg. direct Ni-O_{DMF}	2.10	2.13	2.68	3.20	-	-
Avg. H_{DMF}-O_{amide}	-	-	2.94	3.09	-	-
Avg. Ni-H_{DMF}	3.16	3.01	3.22	2.97	-	-
H_{DMF}-N_{amide}	2.76	2.77	3.11	2.78	-	-
Avg. N_{amide}-Ni-$N_{pyridine}$	83.8	83.7	84.4	84.3	81.0	81.4
$N_{pyridine}$-Ni-$N_{pyridine}$	109.4	108.9	108.1	107.5	117.1	116.2
$N_{pyridine}$-N_{amide}-$N_{pyridine}$-N_{amide}	-7.9	-6.1	12.0	9.7	0.00	0.4

Table S24. Mulliken spin densities on selected fragments, oxidation states of Ni, and spin state of the $[Ni(bpb\text{-}NO_2)]^n$ complexes by charge state *n*. For the 1- and 2- charge states, MeCN molecules are omitted.

Fragment/*n*	2+ (open-shell singlet)	1+ (doublet)	0 (singlet)	1- (doublet)	2- (open-shell singlet)
Oxidation state of Ni	Ni^{3+}	Ni^{2+}	Ni^{2+}	Ni^{1+}	Ni^{1+}
Ni center	1.01	0.07	0.00	0.90	1.04
Pyridyls	0.00; 0.00	0.00; 0.00	0.00	0.00; 0.00	-0.23; -0.57
$-C_6H_4-$ linker	-0.53	0.40	0.00	0.01	-0.05
Amides	-0.58	0.50	0.00	0.10	-0.03
MeCNs	0.12	0.00	0.00	-	-
NO_2	-0.01	0.02	0.00	0.00	-0.15

Table S25. Mulliken spin densities on selected fragments, oxidation states of Ni, and spin state of the $[Ni(bpb\text{-}NO_2)]^n$ complexes in DMF by charge state *n*. For the 1- and 2- charge states, DMF molecules are omitted.

Fragment/*n*	2+ (open-shell singlet)	1+ (doublet)	1+ (doublet)	0 (singlet)	1- (doublet)	2- (open-shell singlet)
Oxidation state of Ni	Ni^{3+}	Ni^{3+}	Ni^{2+}	Ni^{2+}	Ni^{1+}	Ni^{1+}
Ni center	-0.91	0.96	0.09	0.00	0.88	0.92
Pyridyls	0.01; 0.01	0.00; 0.00	0.01; 0.01	0.00	0.01; 0.01	-0.25; -0.25
$-C_6H_4-$ linker	0.51	0.00	0.38	0.00	0.01	-0.13
Amides	0.55	-0.13	0.48	0.00	0.10	-0.04
DMFs	-0.18	0.17	0.03	0.00	-	-
NO_2	0.01	0.00	0.01	0.00	0.00	-0.24

Table S26. Free energies (eV) in MeCN of the LS (singlet, Ni^{2+}) state configurations of the $[Ni(bpb\text{-}R)(MeCN)_2]^0$ complexes relative to their HS (triplet, Ni^{2+}) states. Zero-point energies are taken from the corresponding gas-phase optimized complexes.

Complex/Spin State	LS	HS
$[Ni(bpb)(MeCN)_2]^0$	+0.13	0.00
$[Ni(bpb\text{-}(NMe_2)_2)(MeCN)_2]^0$	+0.01	0.00
$[Ni(bpb\text{-}F)(MeCN)_2]^0$	0.00	+0.29
$[Ni(bpb\text{-}CF_3)(MeCN)_2]^0$	+0.12	0.00
$[Ni(bpb\text{-}NO_2)(MeCN)_2]^0$	0.00	+0.37

Table S27. Free energies (eV) in DMF of the LS (singlet, Ni^{2+}) state configurations of the $[Ni(bpb\text{-}R)(DMF)_2]^0$ complexes relative to their HS (triplet, Ni^{2+}) states. Zero-point energies are taken from the corresponding gas-phase optimized complexes.

Complex/Spin State	LS	HS
$[Ni(bpb)(DMF)_2]^0$	+0.16	0.00
$[Ni(bpb\text{-}(NMe_2)_2)(DMF)_2]^0$	+0.08	0.00
$[Ni(bpb\text{-}F)(DMF)_2]^0$	+0.18	0.00
$[Ni(bpb\text{-}CF_3)(DMF)_2]^0$	+0.37	0.00
$[Ni(bpb\text{-}NO_2)(DMF)_2]^0$	0.00	+0.23

Table S28. Cartesian coordinates of the $[Ni(bpb)(MeCN)_2]^n$ complexes, n=2+, 1+, 0, 1-, 2-, and $[Ni(bpb)]^n$ n=1-, 2- complexes

Complexes	*Cartesian coordinates*			
$[Ni(bpb)(MeCN)_2]^{2+}$	28	-0.000012	-0.312175	0.000010
	7	1.618165	-1.448954	-0.182896
	7	1.243620	1.099843	-0.029027
	7	-1.243579	1.099901	0.028938
	7	-1.618242	-1.448865	0.182999
	8	3.498399	1.517261	0.181288
	8	-3.498334	1.517407	-0.181449
	6	1.705302	-2.755116	-0.429783

	1	0.778268	-3.290568	-0.598117
	6	2.928758	-3.406580	-0.505979
	1	2.953862	-4.473228	-0.707773
	6	4.096372	-2.675892	-0.329670
	1	5.066592	-3.163108	-0.377968
	6	4.002893	-1.305902	-0.117667
	1	4.875048	-0.666137	-0.010417
	6	2.747469	-0.730202	-0.062059
	6	2.582781	0.746807	0.057671
	6	-2.582754	0.746919	-0.057761
	6	-2.747512	-0.730073	0.062076
	6	-4.002963	-1.305713	0.117698
	1	-4.875087	-0.665919	0.010372
	6	-4.096507	-2.675681	0.329814
	1	-5.066749	-3.162851	0.378127
	6	-2.928930	-3.406405	0.506218
	1	-2.954087	-4.473035	0.708104
	6	-1.705443	-2.755002	0.429997
	1	-0.778436	-3.290480	0.598402
	6	0.723504	2.354242	-0.005671
	6	-0.723406	2.354275	0.005537
	6	-1.433749	3.569321	0.003694
	6	-0.710865	4.737082	0.001019
	6	0.711067	4.737050	-0.001215
	6	1.433900	3.569257	-0.003863
	1	-2.516292	3.570481	0.004527
	1	-1.237593	5.687838	0.001755
	1	1.237838	5.687783	-0.001972
	1	2.516443	3.570369	-0.004698
	7	-0.188360	-0.377436	-2.147540
	7	0.188344	-0.377255	2.147564
	6	0.281873	-0.300500	3.290676
	6	-0.281849	-0.300804	-3.290663
	6	-0.400110	-0.203327	-4.726530
	1	-0.614596	-1.189520	-5.151430
	1	-1.213288	0.482826	-4.986053
	1	0.535997	0.174136	-5.151456
	6	0.400186	-0.202861	4.726528
	1	0.607467	-1.190257	5.152218
	1	1.218095	0.477689	4.985946

	1	-0.533417	0.181535	5.150757
$[Ni(bpb)(MeCN)_2]^{1+}$	28	-0.013626	-0.405235	-0.002984
	7	1.561156	-1.607193	-0.182628
$[Ni^{3+}]$	7	1.277508	0.950865	-0.062450
	7	-1.224298	1.023099	0.042984
	7	-1.653642	-1.513262	0.199108
	8	3.539499	1.298795	0.176844
	8	-3.465683	1.498620	-0.167596
	6	1.617570	-2.916153	-0.426220
	1	0.675607	-3.425111	-0.597416
	6	2.821472	-3.598779	-0.495185
	1	2.822706	-4.665663	-0.695741
	6	4.005092	-2.892897	-0.312819
	1	4.963372	-3.403892	-0.354475
	6	3.943893	-1.523204	-0.103901
	1	4.825194	-0.897818	0.008783
	6	2.702510	-0.913452	-0.059136
	6	2.569702	0.573860	0.051330
	6	-2.537445	0.720018	-0.048325
	6	-2.754133	-0.755982	0.078708
	6	-4.028242	-1.292392	0.141644
	1	-4.872946	-0.617942	0.031133
	6	-4.166494	-2.654123	0.364826
	1	-5.152289	-3.108320	0.420869
	6	-3.023978	-3.425648	0.542596
	1	-3.084716	-4.488838	0.753358
	6	-1.783167	-2.814565	0.455730
	1	-0.870892	-3.375844	0.622788
	6	0.764893	2.255566	-0.029591
	6	-0.637965	2.295900	-0.000458
	6	-1.310469	3.515641	0.003046
	6	-0.562233	4.688450	-0.014596
	6	0.826523	4.648644	-0.034886
	6	1.506221	3.434564	-0.042816
	1	-2.393318	3.535779	0.019063
	1	-1.079097	5.644678	-0.011184
	1	1.397307	5.573483	-0.045826
	1	2.588453	3.392510	-0.058373
	7	-0.217342	-0.426744	-2.182216
	7	0.191825	-0.403502	2.181083

	6	0.317087	0.064011	3.223201
	6	-0.303386	0.011492	-3.240694
	6	-0.411983	0.577373	-4.565352
	1	-0.243708	-0.195853	-5.321321
	1	-1.410185	1.005155	-4.702955
	1	0.334527	1.368888	-4.686625
	6	0.476471	0.666466	4.526293
	1	0.341753	-0.085559	5.309867
	1	1.478080	1.100291	4.610474
	1	-0.266957	1.459723	4.655584
$[Ni(bpb)(MeCN)_2]^{1+}$	28	-0.000151	0.388873	0.000104
	7	-1.419715	1.522599	-0.650092
$[Ni^{2+}]$	7	-1.198736	-0.986673	-0.298717
	7	1.199174	-0.985957	0.299171
	7	1.418789	1.523473	0.650077
	8	-3.489133	-1.334741	-0.421892
	8	3.489795	-1.332698	0.421904
	6	-1.376340	2.796035	-1.033485
	1	-0.402770	3.274232	-1.011909
	6	-2.507581	3.471327	-1.471473
	1	-2.427077	4.510251	-1.775750
	6	-3.719659	2.797090	-1.518418
	1	-4.621532	3.304321	-1.850255
	6	-3.755038	1.454441	-1.162885
	1	-4.659905	0.856135	-1.216124
	6	-2.582647	0.850592	-0.750645
	6	-2.513024	-0.618385	-0.471469
	6	2.513299	-0.616883	0.471560
	6	2.582144	0.852194	0.750422
	6	3.754300	1.456836	1.162175
	1	4.659536	0.859078	1.215287
	6	3.718218	2.799548	1.517386
	1	4.619883	3.307412	1.848819
	6	2.505714	3.473050	1.470659
	1	2.424685	4.511994	1.774724
	6	1.374747	2.796977	1.033189
	1	0.400846	3.274515	1.011865
	6	-0.705796	-2.237867	-0.159624
	6	0.706897	-2.237443	0.160400
	6	1.396582	-3.456857	0.309695

	6	0.693901	-4.626328	0.153229
	6	-0.691439	-4.626744	-0.151960
	6	-1.394783	-3.457700	-0.308672
	1	2.455228	-3.451373	0.541049
	1	1.208549	-5.576944	0.265924
	1	-2.453431	-3.452870	-0.540035
	7	1.410770	0.767165	-2.406810
	7	-1.410588	0.768109	2.407297
	6	-2.197058	0.005417	2.765836
	6	2.197342	0.004836	-2.765903
	6	3.188478	-0.967909	-3.179649
	1	3.961703	-0.485418	-3.785458
	1	3.654709	-1.415494	-2.294678
	1	2.715419	-1.756733	-3.772682
	6	-3.188037	-0.967836	3.178747
	1	-3.961626	-0.485873	3.784527
	1	-3.653861	-1.415027	2.293367
	1	-2.715027	-1.756817	3.771610
	1	-1.205561	-5.577671	-0.264457
$[Ni(bpb)(MeCN)_2]^0$	28	-0.319014	-0.000051	0.000022
	7	-1.467554	-1.508330	-0.363225
	7	1.039247	-1.239223	-0.110157
	7	1.039037	1.239359	0.110134
	7	-1.467801	1.508034	0.363301
	8	1.405725	-3.521013	0.119970
	8	1.405105	3.521207	-0.120082
	6	-2.749084	-1.541506	-0.721461
	1	-3.224071	-0.582369	-0.897431
	6	-3.431060	-2.735817	-0.905509
	1	-4.476335	-2.716404	-1.198078
	6	-2.752689	-3.932186	-0.717388
	1	-3.265203	-4.882658	-0.843307
	6	-1.402304	-3.895400	-0.397280
	1	-0.794016	-4.787147	-0.277868
	6	-0.789779	-2.665178	-0.247802
	6	0.695212	-2.538231	-0.042930
	6	0.694772	2.538305	0.042887
	6	-0.790232	2.664998	0.247818
	6	-1.402967	3.895118	0.397273
	1	-0.794840	4.786968	0.277808

	6	-2.753350	3.931678	0.717417
	1	-3.266027	4.882066	0.843314
	6	-3.431506	2.735196	0.905600
	1	-4.476769	2.715611	1.198198
	6	-2.749325	1.540998	0.721575
	1	-3.224139	0.581783	0.897589
	6	2.327327	-0.705456	-0.045470
	6	2.327207	0.705813	0.045389
	6	3.530642	1.406182	0.079898
	6	4.727079	0.694531	0.037816
	6	4.727196	-0.693764	-0.037954
	6	3.530881	-1.405619	-0.080009
	1	3.514857	2.489002	0.137103
	1	5.669198	1.237091	0.065460
	1	5.669408	-1.236165	-0.065617
	1	3.515281	-2.488443	-0.137210
	7	-1.515143	0.866487	-2.855102
	7	-1.514746	-0.866654	2.855237
	6	-0.371973	-0.952336	2.983426
	6	-0.372410	0.952430	-2.983370
	6	1.066013	1.054517	-3.134868
	1	1.334764	1.047201	-4.195722
	1	1.429140	1.979889	-2.675429
	1	1.550524	0.211067	-2.631760
	6	1.066487	-1.054071	3.134818
	1	1.335323	-1.046649	4.195650
	1	1.429806	-1.979368	2.675377
	1	1.550747	-0.210517	2.631644
$[Ni(bpb)(MeCN)_2]^{1-}$	28	-0.000645	-0.722867	0.000612
	7	-1.753006	-1.796725	0.153930
	7	-1.293133	0.796704	0.120367
	7	1.293858	0.794805	-0.121505
	7	1.750261	-1.799277	-0.151246
	8	-3.582825	1.271887	0.194378
	8	3.584182	1.266827	-0.196092
	6	-1.974177	-3.111988	0.193164
	1	-1.088753	-3.741718	0.147449
	6	-3.238433	-3.675778	0.287196
	1	-3.350660	-4.756459	0.314666
	6	-4.336245	-2.826093	0.343217

	1	-5.345363	-3.226669	0.416123
	6	-4.118411	-1.457419	0.305092
	1	-4.920635	-0.726299	0.343958
	6	-2.819342	-0.972043	0.212623
	6	-2.591834	0.530606	0.175095
	6	2.592206	0.526895	-0.175788
	6	2.817700	-0.976110	-0.211202
	6	4.116102	-1.463361	-0.303152
	1	4.919304	-0.733375	-0.343120
	6	4.332088	-2.832376	-0.339442
	1	5.340654	-3.234410	-0.411940
	6	3.233141	-3.680507	-0.282126
	1	3.343908	-4.761374	-0.308147
	6	1.969657	-3.114890	-0.188697
	1	1.083392	-3.743362	-0.142006
	6	-0.713523	2.051976	0.062472
	6	0.715919	2.050933	-0.065454
	6	1.395363	3.272175	-0.125729
	6	0.694951	4.474499	-0.064723
	6	-0.689312	4.475510	0.058398
	6	-1.391332	3.274210	0.121054
	1	2.476403	3.251916	-0.218967
	1	1.241472	5.415222	-0.113630
	1	-1.234572	5.417031	0.106022
	1	-2.472396	3.255526	0.214338
	7	0.517100	0.549548	5.553032
	7	-0.513848	0.539594	-5.553724
	6	-0.501781	0.876367	-4.450601
	6	0.504662	0.884403	4.449328
	6	0.490929	1.303426	3.062906
	1	1.493403	1.596581	2.736213
	1	0.149580	0.492431	2.406808
	1	-0.181588	2.154940	2.919338
	6	-0.488507	1.297763	-3.064899
	1	0.183473	2.149919	-2.922679
	1	-1.491203	1.590838	-2.738843
	1	-0.146757	0.488094	-2.407372
$[Ni(bpb)(MeCN)_2]^{2-}$	28	-0.000591	-0.732247	0.000359
	7	-1.590610	-1.767720	0.629081
	7	-1.105288	0.792500	0.684189

	7	1.106025	0.790616	-0.684583
	7	1.588057	-1.770189	-0.627710
	8	-3.201633	1.300439	1.595212
	8	3.203103	1.295234	-1.595759
	6	-1.868835	-3.089400	0.587882
	1	-1.111521	-3.702716	0.100290
	6	-3.009958	-3.665643	1.096910
	1	-3.148590	-4.742718	1.015847
	6	-3.982450	-2.838705	1.706876
	1	-4.900320	-3.252770	2.119672
	6	-3.721658	-1.473099	1.743005
	1	-4.418074	-0.755061	2.171076
	6	-2.548146	-0.956736	1.215776
	6	-2.307798	0.517635	1.207615
	6	2.308198	0.513842	-1.207757
	6	2.546628	-0.960843	-1.214959
	6	3.719454	-1.479075	-1.741877
	1	4.416797	-0.762229	-2.170441
	6	3.978456	-2.844997	-1.704864
	1	4.895779	-3.260527	-2.117400
	6	3.004912	-3.670262	-1.094319
	1	3.142143	-4.747462	-1.012550
	6	1.864556	-3.092197	-0.585637
	1	1.106455	-3.704203	-0.097617
	6	-0.610445	2.045776	0.381097
	6	0.612792	2.044738	-0.382329
	6	1.192771	3.269976	-0.732135
	6	0.594678	4.476830	-0.363741
	6	-0.589112	4.477830	0.361012
	6	-1.188809	3.271997	0.730136
	1	2.119738	3.244204	-1.296451
	1	1.065285	5.417849	-0.651426
	1	-1.058467	5.419646	0.648133
	1	-2.115799	3.247793	1.294484
	7	4.232542	0.925039	4.047402
	7	-4.229596	0.924677	-4.049249
	6	-3.555284	0.941842	-3.112152
	6	3.557791	0.942356	3.110590
	6	2.715576	0.968254	1.932513
	1	3.287248	1.282000	1.051324

	1	2.284493	-0.017884	1.722112
	1	1.883704	1.668899	2.061105
	6	-2.713635	0.967701	-1.933657
	1	-1.880195	1.666381	-2.062836
	1	-3.285142	1.283983	-1.053256
	1	-2.284753	-0.019040	-1.721549
$[Ni(bpb)]^{1-}$	28	0.00000000	-0.49578300	0.00001400
	7	1.75043900	-1.57651700	0.00010500
	7	1.30304800	1.01792000	-0.00006300
	7	-1.30305100	1.01791700	0.00007300
	7	-1.75043500	-1.57652000	-0.00008300
	8	3.60041200	1.48356500	-0.00028900
	8	-3.60041700	1.48355800	0.00023700
	6	1.97282200	-2.89287800	0.00028200
	1	1.08469700	-3.52103600	0.00044100
	6	3.23871000	-3.46134800	0.00028700
	1	3.34902000	-4.54293300	0.00044100
	6	4.34063300	-2.61401800	0.00009400
	1	5.35176500	-3.01715700	0.00008300
	6	4.12281500	-1.24476100	-0.00007200
	1	4.92585300	-0.51321100	-0.00021000
	6	2.82158400	-0.75333500	-0.00005100
	6	2.59963800	0.75064800	-0.00015800
	6	-2.59964100	0.75064300	0.00013800
	6	-2.82158300	-0.75334100	0.00003700
	6	-4.12281200	-1.24477100	0.00002800
	1	-4.92585200	-0.51322300	0.00013500
	6	-4.34062600	-2.61402800	-0.00012800
	1	-5.35175700	-3.01717100	-0.00014000
	6	-3.23870000	-3.46135500	-0.00027900
	1	-3.34900600	-4.54294100	-0.00042100
	6	-1.97281400	-2.89288200	-0.00024900
	1	-1.08468700	-3.52103700	-0.00037600
	6	0.71737600	2.27138900	-0.00004300
	6	-0.71738100	2.27138700	0.00005300
	6	-1.39803900	3.49347100	0.00010700
	6	-0.69506500	4.69535800	0.00005900
	6	0.69505500	4.69535900	-0.00004200
	6	1.39803100	3.49347400	-0.00009400
	1	-2.48319600	3.47299700	0.00018500
	1	-1.24345600	5.63698000	0.00010100
	1	1.24344500	5.63698300	-0.00008200
	1	2.48318800	3.47300200	-0.00017500
$[Ni(bpb)]^{2-}$	28	-0.00000900	-0.50278700	0.00003000
	7	1.70085200	-1.55238200	-0.09365800

	7	1.29956300	1.01618900	0.07498500
	7	-1.29952500	1.01623000	-0.07495700
	7	-1.70091100	-1.55232500	0.09370400
	8	3.58861000	1.50608000	0.20293700
	8	-3.58855200	1.50620100	-0.20298000
	6	1.93450800	-2.87786700	-0.23139200
	1	1.04096300	-3.49017300	-0.35441600
	6	3.18034000	-3.46092300	-0.23272700
	1	3.26581700	-4.54091700	-0.35046900
	6	4.32526100	-2.63840000	-0.08359000
	1	5.33015500	-3.05737200	-0.07607500
	6	4.10426500	-1.26777500	0.04189100
	1	4.91667000	-0.55055800	0.14460800
	6	2.82297600	-0.74329600	0.03199100
	6	2.60983300	0.73532300	0.11970800
	6	-2.60980500	0.73540900	-0.11971400
	6	-2.82300200	-0.74320100	-0.03199500
	6	-4.10430500	-1.26764400	-0.04195300
	1	-4.91668000	-0.55040100	-0.14470900
	6	-4.32535000	-2.63826400	0.08351600
	1	-5.33025300	-3.05720700	0.07594800
	6	-3.18046600	-3.46082500	0.23270600
	1	-3.26598300	-4.54081900	0.35044200
	6	-1.93461800	-2.87780400	0.23142900
	1	-1.04110200	-3.49014000	0.35449300
	6	0.71975200	2.26837200	0.04681000
	6	-0.71967200	2.26839400	-0.04679600
	6	-1.39387500	3.49422900	-0.09130900
	6	-0.69271700	4.70183500	-0.04518700
	6	0.69288300	4.70181400	0.04517100
	6	1.39399800	3.49418400	0.09130700
	1	-2.47704000	3.46709500	-0.16361000
	1	-1.24307800	5.64396300	-0.08186900
	1	1.24327700	5.64392300	0.08184100
	1	2.47716300	3.46701600	0.16360500

Table S29. Cartesian coordinates of $[Ni(bpb\text{-}(NMe_2)_2)(MeCN)_2]^n$, n=2+, 1+, 0, and $[Ni(bpb\text{-}(NMe_2)_2]^n$, n=1-, 2-.

Complexes	*Cartesian coordinates*			
$[Ni(bpb\text{-}(NMe_2)_2)(MeCN)_2]^{2+}$	28	-0.000014	0.287798	0.000018
	7	1.590822	-0.843505	-0.095740
	7	1.246254	1.693515	0.030598
	7	-1.246279	1.693520	-0.030477

	7	-1.590852	-0.843507	0.095697
	8	3.492411	2.107698	0.335494
	8	-3.492430	2.107724	-0.335383
	6	1.710357	-2.152488	-0.351796
	1	0.798080	-2.692760	-0.578136
	6	2.915513	-2.809674	-0.372397
	1	2.914931	-3.869574	-0.598135
	6	4.123357	-2.105458	-0.119003
	6	3.979516	-0.705948	0.076020
	1	4.824492	-0.037924	0.207457
	6	2.733918	-0.142596	0.071544
	6	2.579938	1.337385	0.175605
	6	-2.579962	1.337400	-0.175523
	6	-2.733945	-0.142586	-0.071548
	6	-3.979545	-0.705934	-0.076058
	1	-4.824519	-0.037900	-0.207458
	6	-4.123391	-2.105454	0.118889
	6	-2.915550	-2.809686	0.372250
	1	-2.914971	-3.869599	0.597934
	6	-1.710391	-2.152504	0.351684
	1	-0.798116	-2.692790	0.577997
	6	0.723290	2.944645	0.028864
	6	-0.723310	2.944648	-0.028707
	6	-1.430879	4.162027	-0.064876
	6	-0.709751	5.329782	-0.032990
	6	0.709740	5.329778	0.033185
	6	1.430863	4.162021	0.065055
	1	-2.512282	4.160497	-0.115930
	1	-1.236031	6.280388	-0.057616
	1	1.236023	6.280383	0.057823
	1	2.512266	4.160486	0.116109
	7	-0.076111	0.265676	-2.178165
	7	0.076124	0.265474	2.178195
	6	0.107095	0.354218	3.323311
	6	-0.106972	0.354615	-3.323269
	6	-0.146399	0.469133	-4.763068
	1	-0.308179	-0.516177	-5.211993
	1	-0.962327	1.136102	-5.060222
	1	0.800818	0.878352	-5.129293
	6	0.146666	0.468485	4.763126

	1	0.308598	-0.516893	5.211856
	1	0.962562	1.135486	5.060310
	1	-0.800556	0.877541	5.129530
	7	-5.316326	-2.707878	0.091706
	7	5.316291	-2.707886	-0.091857
	6	-6.524926	-1.929786	-0.143465
	1	-6.472780	-1.402552	-1.103208
	1	-7.382186	-2.601428	-0.176624
	1	-6.693626	-1.202460	0.660187
	6	-5.419628	-4.139035	0.333252
	1	-6.460332	-4.445767	0.233626
	1	-4.830461	-4.705191	-0.398250
	1	-5.085604	-4.397607	1.345928
	6	5.419587	-4.139032	-0.333474
	1	4.830423	-4.705222	0.398006
	1	5.085554	-4.397555	-1.346159
	1	6.460290	-4.445772	-0.233870
	6	6.524895	-1.929809	0.143344
	1	6.472761	-1.402630	1.103118
	1	7.382154	-2.601454	0.176453
	1	6.693585	-1.202437	-0.660269
$[Ni(bpb\text{-}(NMe_2)_2)(MeCN)_2]^{1+}$	28	-0.000001	0.176040	0.000004
	7	-1.522342	-0.955936	0.241170
$[Ni^{2+}]$	7	-1.234986	1.548882	-0.000668
	7	1.234972	1.548893	0.000717
	7	1.522348	-0.955918	-0.241171
	8	-3.488841	1.915506	-0.401074
	8	3.488835	1.915534	0.401058
	6	-1.607913	-2.240277	0.597385
	1	-0.671911	-2.738434	0.827407
	6	-2.800365	-2.917110	0.712518
	1	-2.772481	-3.956148	1.018731
	6	-4.023461	-2.254722	0.450743
	6	-3.920589	-0.876815	0.143628
	1	-4.784071	-0.239071	-0.010676
	6	-2.685423	-0.290860	0.064852
	6	-2.555020	1.184277	-0.146178
	6	2.555014	1.184299	0.146185
	6	2.685425	-0.290835	-0.064860
	6	3.920594	-0.876781	-0.143650

	1	4.784073	-0.239033	0.010650
	6	4.023472	-2.254689	-0.450761
	6	2.800379	-2.917084	-0.712532
	1	2.772499	-3.956120	-1.018745
	6	1.607924	-2.240257	-0.597391
	1	0.671924	-2.738420	-0.827408
	6	-0.723023	2.801279	-0.012928
	6	0.722997	2.801285	0.012993
	6	1.426927	4.022230	0.030539
	6	0.707999	5.193219	0.015435
	6	-0.708047	5.193213	-0.015321
	6	-1.426964	4.022217	-0.030451
	1	2.510249	4.015970	0.054335
	1	1.235950	6.143263	0.026143
	1	-1.236007	6.143252	-0.026008
	1	-2.510286	4.015948	-0.054248
	7	0.761830	-0.055776	2.836814
	7	-0.761525	-0.055496	-2.836667
	6	-1.260699	0.928765	-3.169572
	6	1.260758	0.928667	3.169539
	6	1.894118	2.177190	3.545407
	1	2.287278	2.112292	4.564598
	1	2.717247	2.395219	2.855816
	1	1.166706	2.993897	3.497489
	6	-1.894341	2.177089	-3.545636
	1	-2.287959	2.111772	-4.564623
	1	-2.717177	2.395298	-2.855760
	1	-1.166980	2.993877	-3.498360
	7	5.212253	-2.883482	-0.515040
	7	-5.212239	-2.883522	0.515020
	6	6.437872	-2.146444	-0.267793
	1	6.431839	-1.691648	0.730525
	1	7.284500	-2.831501	-0.318335
	1	6.592766	-1.358076	-1.016079
	6	5.273809	-4.285870	-0.879243
	1	6.311098	-4.620008	-0.848395
	1	4.700412	-4.903851	-0.176494
	1	4.892987	-4.455822	-1.895141
	6	-6.437860	-2.146495	0.267755
	1	-6.431814	-1.691695	-0.730562

	1	-7.284483	-2.831560	0.318277
	1	-6.592777	-1.358131	1.016040
	6	-5.273791	-4.285901	0.879256
	1	-6.311082	-4.620035	0.848447
	1	-4.700415	-4.903903	0.176507
	1	-4.892941	-4.455830	1.895148
$[Ni(bpb\text{-}(NMe_2)_2)(MeCN)_2]^0$	28	0.000224	0.318403	0.000906
	7	-1.533533	-0.827286	0.174564
	7	-1.245614	1.679058	-0.026366
	7	1.246238	1.678919	0.027677
	7	1.533809	-0.827550	-0.172292
	8	-3.498202	2.058491	-0.445273
	8	3.499423	2.058496	0.443265
	6	-1.637333	-2.114941	0.502608
	1	-0.715357	-2.614430	0.781158
	6	-2.837019	-2.793287	0.540663
	1	-2.827939	-3.838204	0.828009
	6	-4.035384	-2.119295	0.223311
	6	-3.917567	-0.739789	-0.046442
	1	-4.765287	-0.089483	-0.234489
	6	-2.679145	-0.150454	-0.047303
	6	-2.534906	1.339459	-0.211343
	6	2.535761	1.339378	0.211160
	6	2.679713	-0.150633	0.047785
	6	3.918135	-0.739981	0.045563
	1	4.766097	-0.089537	0.232042
	6	4.035605	-2.119633	-0.223570
	6	2.836796	-2.793822	-0.538798
	1	2.827316	-3.838881	-0.825608
	6	1.637178	-2.115430	-0.499596
	1	0.714854	-2.615107	-0.776631
	6	-0.706566	2.966259	-0.031861
	6	0.707355	2.966187	0.032420
	6	1.405968	4.170882	0.073729
	6	0.694572	5.367760	0.037533
	6	-0.693527	5.367818	-0.038699
	6	-1.405050	4.170989	-0.074038
	1	2.488584	4.154168	0.134053
	1	1.237332	6.309877	0.068778
	1	-1.236176	6.309975	-0.070655

	1	-2.487663	4.154352	-0.134442
	7	0.498504	-1.027431	3.030785
	7	-0.499978	-1.026086	-3.028464
	6	-0.493239	0.124024	-3.108213
	6	0.490691	0.122704	3.110055
	6	0.473710	1.569708	3.207464
	1	0.294074	1.875813	4.242843
	1	1.428893	1.982608	2.867704
	1	-0.316808	1.972898	2.565876
	6	-0.477642	1.571001	-3.206217
	1	-0.298588	1.876834	-4.241776
	1	-1.433123	1.983174	-2.866379
	1	0.312709	1.975215	-2.565071
	7	5.236274	-2.750989	-0.205901
	7	-5.236000	-2.750669	0.204254
	6	6.440546	-1.998982	0.074222
	1	6.632769	-1.233750	-0.690844
	1	6.381030	-1.503759	1.051861
	1	7.292566	-2.680540	0.094912
	6	5.320300	-4.151859	-0.550475
	1	4.698816	-4.764422	0.116553
	1	5.006610	-4.338610	-1.587737
	1	6.353228	-4.487478	-0.444955
	6	-6.439575	-1.999396	-0.080730
	1	-6.634111	-1.232886	0.682448
	1	-6.377354	-1.505833	-1.059060
	1	-7.291427	-2.681131	-0.102615
	6	-5.320356	-4.151652	0.548325
	1	-4.698788	-4.764269	-0.118622
	1	-5.006980	-4.338584	1.585621
	1	-6.353334	-4.487040	0.442612
$[Ni(bpb\text{-}(NMe_2)_2)]^{1-}$	28	-0.000010	0.154112	-0.038396
	7	-1.755716	-0.922794	-0.054676
	7	-1.305204	1.671079	-0.012748
	7	1.305267	1.671018	-0.014075
	7	1.755660	-0.922914	-0.054414
	8	-3.600733	2.144798	0.002860
	8	3.600817	2.144663	-0.001538
	6	-2.010461	-2.229781	-0.065050
	1	-1.137427	-2.880351	-0.066426

	6	-3.277366	-2.789868	-0.067634
	1	-3.369160	-3.870959	-0.064954
	6	-4.397561	-1.941207	-0.070181
	6	-4.129427	-0.565082	-0.044964
	1	-4.897470	0.201305	-0.021637
	6	-2.827587	-0.098904	-0.041796
	6	-2.602038	1.406584	-0.014281
	6	2.602084	1.406472	-0.016947
	6	2.827560	-0.099055	-0.043138
	6	4.129381	-0.565288	-0.046286
	1	4.897449	0.201103	-0.024394
	6	4.397474	-1.941456	-0.069454
	6	3.277244	-2.790066	-0.065089
	1	3.369000	-3.871154	-0.060680
	6	2.010361	-2.229926	-0.062871
	1	1.137309	-2.880464	-0.062755
	6	-0.717928	2.922913	0.009021
	6	0.718067	2.922883	0.008121
	6	1.397653	4.146047	0.029150
	6	0.695262	5.348105	0.050574
	6	-0.694976	5.348133	0.051542
	6	-1.397440	4.146103	0.031043
	1	2.482851	4.125092	0.028260
	1	1.243990	6.289633	0.066644
	1	-1.243647	6.289683	0.068401
	1	-2.482639	4.125192	0.031648
	7	-5.687302	-2.430770	-0.099957
	7	5.687142	-2.431105	-0.098888
	6	-5.904270	-3.832613	0.149369
	1	-5.560934	-4.151408	1.148775
	1	-6.972434	-4.051553	0.067777
	1	-5.387619	-4.447479	-0.598561
	6	-6.778688	-1.519019	0.149842
	1	-6.741711	-1.071743	1.156721
	1	-6.775904	-0.702835	-0.581153
	1	-7.725093	-2.056369	0.041593
	6	6.778683	-1.518994	0.148904
	1	6.775678	-0.704067	-0.583455
	1	6.742114	-1.070013	1.155011
	1	7.724985	-2.056565	0.041232

	6	5.904122	-3.832569	0.152739
	1	5.560660	-4.149639	1.152655
	1	5.387501	-4.448634	-0.594217
	1	6.972301	-4.051690	0.071656
$[Ni(bpb\text{-}(NMe_2)_2)]^{2-}$	28	0.001098	0.134550	-0.079774
	7	-1.710945	-0.926891	-0.094026
	7	-1.305008	1.648269	-0.035765
	7	1.299667	1.649752	0.033192
	7	1.719876	-0.916453	-0.170695
	8	-3.600532	2.140744	-0.079030
	8	3.592725	2.147230	0.127812
	6	-1.955059	-2.249939	-0.106796
	1	-1.066958	-2.882349	-0.108889
	6	-3.202246	-2.829001	-0.126090
	1	-3.299504	-3.911790	-0.183401
	6	-4.352751	-1.989458	-0.124229
	6	-4.120333	-0.620836	-0.110000
	1	-4.915115	0.121944	-0.102310
	6	-2.826807	-0.106865	-0.104394
	6	-2.615519	1.370281	-0.075104
	6	2.612180	1.375081	0.046697
	6	2.830904	-0.096270	-0.060783
	6	4.126509	-0.606633	-0.052666
	1	4.916485	0.133608	0.054558
	6	4.366361	-1.967232	-0.183569
	6	3.221870	-2.803682	-0.328655
	1	3.328655	-3.874130	-0.495543
	6	1.972096	-2.230141	-0.311018
	1	1.089293	-2.858599	-0.432152
	6	-0.726164	2.899654	0.039215
	6	0.715959	2.900011	0.082528
	6	1.388343	4.124604	0.164754
	6	0.683518	5.331087	0.203613
	6	-0.703719	5.330630	0.161476
	6	-1.403604	4.123352	0.079793
	1	2.473434	4.098326	0.197370
	1	1.233349	6.272068	0.267684
	1	-1.257190	6.271167	0.191277
	1	-2.488613	4.096054	0.044711
	7	-5.648128	-2.588557	-0.143753

	7	5.663844	-2.561865	-0.194769
	6	-5.948084	-3.364514	1.041820
	1	-6.120878	-2.731457	1.936405
	1	-6.848183	-3.975172	0.873278
	1	-5.114622	-4.036461	1.267811
	6	-6.732033	-1.698766	-0.468959
	1	-6.940724	-0.943990	0.316106
	1	-6.501541	-1.162131	-1.395399
	1	-7.648712	-2.286795	-0.620927
	6	6.758988	-1.644767	-0.372638
	1	6.576075	-1.020339	-1.253587
	1	6.920769	-0.970015	0.492392
	1	7.685620	-2.215185	-0.531124
	6	5.904544	-3.451636	0.922376
	1	6.027673	-2.910305	1.883005
	1	5.062864	-4.141447	1.036746
	1	6.814695	-4.043734	0.741888

Table S30. Cartesian coordinates of $[Ni(bpb\text{-}F)(MeCN)_2]^n$, n=2+, 1+, 0, and $[Ni(bpb\text{-}F)]^n$, n=1-, 2-.

Complexes	*Cartesian coordinates*			
$[Ni(bpb\text{-}F)(MeCN)_2]^{2+}$	28	-0.499436	0.077850	0.000651
	7	-1.217183	1.921690	0.172511
	7	1.163851	0.949385	0.040203
	7	0.579363	-1.467212	-0.019069
	7	-1.987024	-1.227722	-0.186439
	8	2.119059	3.030623	-0.156255
	8	0.449471	-3.757279	0.187597
	6	-2.467421	2.318952	0.405051
	1	-3.211145	1.547497	0.566533
	6	-2.807805	3.662808	0.475354
	1	-3.839599	3.942737	0.665428
	6	-1.816939	4.621392	0.308580
	1	-2.058398	5.680066	0.352908
	6	-0.506903	4.202500	0.111088
	1	0.324523	4.895605	0.011523
	6	-0.247981	2.845966	0.060086
	6	1.146934	2.329257	-0.045258
	6	-0.083175	-2.686005	0.062500

	6	-1.555865	-2.495263	-0.063443
	6	-2.411536	-3.579381	-0.121282
	1	-1.996633	-4.578078	-0.012212
	6	-3.764368	-3.347025	-0.337350
	1	-4.466615	-4.174898	-0.387348
	6	-4.198034	-2.040056	-0.514944
	1	-5.239932	-1.812481	-0.719315
	6	-3.276117	-1.004857	-0.436465
	1	-3.577712	0.022240	-0.605175
	6	2.263016	0.144061	0.014341
	6	1.917710	-1.260787	0.003488
	6	2.931877	-2.247301	0.004665
	6	4.236854	-1.838678	0.004654
	6	4.548214	-0.454119	0.004971
	6	3.597775	0.545121	0.009158
	1	2.669895	-3.297822	0.005057
	1	5.054945	-2.553721	0.002614
	1	3.888626	1.588089	0.009547
	7	-0.626262	-0.084313	2.148222
	7	-0.510104	0.269185	-2.147559
	6	-0.412691	0.355861	-3.289666
	6	-0.587303	-0.175545	3.293465
	6	-0.537485	-0.290553	4.732056
	1	-1.545949	-0.199925	5.149114
	1	-0.120316	-1.263515	5.012642
	1	0.094165	0.502511	5.146154
	6	-0.288950	0.465471	-4.724214
	1	-1.187668	0.931291	-5.141931
	1	0.582270	1.079680	-4.975594
	1	-0.164093	-0.529842	-5.163653
	9	5.814693	-0.122666	0.002073
$[Ni(bpb\text{-}F)(MeCN)_2]^{1+}$ $[Ni^{3+}]$	28	-0.583640	0.097226	-0.001476
	7	-1.312927	1.939920	0.195395
	7	1.070945	0.974439	0.049362
	7	0.465175	-1.453072	-0.059106
	7	-2.092315	-1.186163	-0.188905
	8	2.020924	3.059445	-0.154704
	8	0.330144	-3.736943	0.157133
	6	-2.557424	2.347193	0.442868
	1	-3.303227	1.577619	0.606211

	6	-2.887790	3.690683	0.524906
	1	-3.914456	3.979058	0.728063
	6	-1.887633	4.640275	0.352117
	1	-2.119432	5.700783	0.404558
	6	-0.585945	4.211787	0.138172
	1	0.254891	4.891647	0.031633
	6	-0.336201	2.852169	0.079131
	6	1.059601	2.323217	-0.040325
	6	-0.177031	-2.636076	0.040946
	6	-1.657460	-2.449715	-0.071664
	6	-2.517335	-3.532723	-0.123330
	1	-2.093856	-4.527565	-0.015942
	6	-3.868553	-3.299836	-0.331430
	1	-4.571744	-4.127199	-0.378127
	6	-4.306463	-1.992082	-0.506037
	1	-5.349175	-1.764972	-0.705071
	6	-3.383284	-0.961213	-0.431178
	1	-3.680526	0.068147	-0.596262
	6	2.187075	0.131466	0.009056
	6	1.849304	-1.230826	-0.022700
	6	2.846927	-2.202826	-0.034848
	6	4.178914	-1.805653	-0.023236
	6	4.486854	-0.456250	-0.000482
	6	3.518661	0.535146	0.016315
	1	2.578551	-3.251976	-0.052807
	1	4.980480	-2.537957	-0.033283
	1	3.794770	1.581970	0.034448
	7	-0.644754	-0.102249	2.178479
	7	-0.558548	0.308379	-2.180096
	6	-0.124901	0.314076	-3.243863
	6	-0.252343	-0.310339	3.237932
	6	0.252799	-0.578886	4.564423
	1	-0.574143	-0.633169	5.279288
	1	0.792064	-1.531757	4.564147
	1	0.938716	0.218904	4.866698
	6	0.434209	0.321994	-4.575867
	1	-0.231405	0.856157	-5.261031
	1	1.409167	0.820057	-4.559489
	1	0.564622	-0.706069	-4.928421
	9	5.770349	-0.083811	0.007327

$[Ni(bpb\text{-}F)(MeCN)_2]^{1+}$	28	-0.520724	0.237753	0.000793
	7	-2.199991	-0.460280	-0.651511
$[Ni^{2+}]$	7	0.111175	-1.471925	-0.318447
	7	1.263172	0.627140	0.298568
	7	-0.828731	2.023567	0.668152
	8	-0.680483	-3.647486	-0.459019
	8	2.674387	2.459677	0.450029
	6	-3.298644	0.190640	-1.024469
	1	-3.252953	1.274047	-0.991356
	6	-4.434631	-0.474938	-1.465803
	1	-5.309348	0.096144	-1.761050
	6	-4.422943	-1.861129	-1.527555
	1	-5.300848	-2.406840	-1.862557
	6	-3.259743	-2.538387	-1.182602
	1	-3.168068	-3.618633	-1.247879
	6	-2.167379	-1.802108	-0.765767
	6	-0.844592	-2.447748	-0.497215
	6	1.575023	1.950088	0.488680
	6	0.321074	2.717184	0.775305
	6	0.357527	4.031438	1.199701
	1	1.318476	4.534211	1.258549
	6	-0.835690	4.645386	1.560062
	1	-0.844790	5.676923	1.901589
	6	-2.011247	3.909936	1.505101
	1	-2.960331	4.338194	1.812128
	6	-1.965245	2.597248	1.055423
	1	-2.854307	1.976110	1.027142
	6	1.444639	-1.638083	-0.199027
	6	2.125418	-0.401388	0.131007
	6	3.520108	-0.375658	0.258020
	6	4.187165	-1.560788	0.069145
	6	3.546431	-2.782772	-0.242985
	6	2.184323	-2.828478	-0.377598
	1	4.051191	0.538630	0.494071
	9	5.505084	-1.581075	0.179666
	1	4.159685	-3.669704	-0.372817
	1	1.668448	-3.751393	-0.614879
	7	-0.183803	1.673704	-2.393912
	7	-1.528911	-0.870534	2.386183
	6	-1.291652	-1.937120	2.753262

	6	0.842236	2.063832	-2.745755
	6	2.145782	2.549964	-3.151300
	1	2.044247	3.494341	-3.694945
	1	2.768951	2.712742	-2.264716
	1	2.633849	1.818278	-3.802562
	6	-0.983173	-3.287436	3.179039
	1	-1.821902	-3.706440	3.743564
	1	-0.794501	-3.914797	2.300658
	1	-0.093590	-3.286553	3.816527
$[Ni(bpb\text{-}F)(MeCN)_2]^0$	28	-0.507605	0.060745	-0.005040
	7	-1.336891	1.765772	0.362685
	7	1.066814	1.012681	0.094584
	7	0.582982	-1.418710	-0.119026
	7	-1.928441	-1.198150	-0.356248
	8	1.875864	3.178701	-0.109604
	8	0.507163	-3.725713	0.121501
	6	-2.586685	2.047264	0.723817
	1	-3.239762	1.198871	0.897135
	6	-3.021931	3.351040	0.914125
	1	-4.050530	3.534729	1.208721
	6	-2.123435	4.392808	0.730025
	6	-0.806525	4.094375	0.406981
	1	-0.036039	4.850951	0.291007
	6	-0.446844	2.769070	0.250343
	6	0.984574	2.355981	0.042122
	6	-0.004314	-2.625711	-0.043025
	6	-1.486594	-2.464190	-0.239203
	6	-2.326802	-3.552920	-0.379062
	1	-1.902231	-4.545291	-0.258914
	6	-3.660893	-3.328489	-0.691037
	6	-4.095949	-2.024119	-0.881216
	1	-5.119565	-1.803669	-1.167816
	6	-3.194350	-0.983785	-0.707055
	1	-3.475615	0.048442	-0.885642
	6	2.223422	0.240897	0.027494
	6	1.950806	-1.144553	-0.058504
	6	2.997988	-2.061086	-0.092598
	6	4.312118	-1.601364	-0.057090
	6	4.551877	-0.240446	0.012180
	6	3.537156	0.702193	0.055195

	1	2.776378	-3.121171	-0.144763
	1	5.149870	-2.291887	-0.083646
	9	5.824580	0.192849	0.039859
	1	3.756540	1.762201	0.108453
	7	-1.838250	-0.558239	2.848535
	7	-1.559020	1.138109	-2.843118
	6	-0.426260	0.978436	-2.987866
	6	-0.741423	-0.889358	2.978764
	6	0.640766	-1.300040	3.131464
	1	0.902097	-1.357298	4.192636
	1	0.797326	-2.279166	2.666439
	1	1.296797	-0.577232	2.635124
	6	0.998458	0.770777	-3.159621
	1	1.242615	0.690606	-4.223388
	1	1.556570	1.605194	-2.722261
	1	1.300475	-0.148877	-2.647402
	1	-4.348615	-4.162167	-0.809192
	1	-2.440131	5.424430	0.861449
$[Ni(bpb\text{-}F)]^{1-}$	28	-0.687777	0.258242	-0.002967
	7	-0.924937	2.302111	-0.007111
	7	1.235601	0.801480	0.003662
	7	0.135709	-1.560265	-0.006234
	7	-2.406030	-0.878285	0.005523
	8	2.630490	2.682333	0.016414
	8	-0.400972	-3.840397	-0.012242
	6	-2.024839	3.058834	-0.016577
	1	-2.968760	2.518325	-0.026242
	6	-2.005986	4.446315	-0.015361
	1	-2.940089	5.002450	-0.023443
	6	-0.773406	5.088500	-0.003818
	1	-0.712728	6.175234	-0.002210
	6	0.376505	4.313627	0.004978
	1	1.378071	4.734074	0.013231
	6	0.272624	2.927077	0.002720
	6	1.541987	2.090441	0.008905
	6	-0.650118	-2.624755	-0.008016
	6	-2.107766	-2.195853	-0.001644
	6	-3.099769	-3.171038	-0.000615
	1	-2.772926	-4.206958	-0.006770
	6	-4.433835	-2.794073	0.008694

	1	-5.223972	-3.542771	0.009617
	6	-4.741025	-1.438434	0.017460
	1	-5.769104	-1.085079	0.025816
	6	-3.693872	-0.527802	0.015483
	1	-3.891382	0.541961	0.022936
	6	2.122844	-0.254604	0.001950
	6	1.519389	-1.557403	-0.004515
	6	2.342894	-2.685655	-0.007706
	6	3.731690	-2.560992	-0.004487
	6	4.283118	-1.294864	0.001950
	6	3.517925	-0.143382	0.005309
	1	1.870971	-3.662733	-0.012624
	1	4.379420	-3.434164	-0.006899
	1	3.978708	0.838362	0.010571
	9	5.637872	-1.172875	0.005176
$[Ni(bpb\text{-}F)]^{2-}$	28	0.707817	0.228901	-0.007542
	7	1.015219	2.215316	-0.085868
	7	-1.195439	0.841372	0.059575
	7	-0.195745	-1.557499	-0.072808
	7	2.325644	-0.934805	0.080537
	8	-2.542380	2.754997	0.180293
	8	0.232434	-3.858505	-0.173934
	6	2.143484	2.948564	-0.205178
	1	3.058568	2.366162	-0.317528
	6	2.196437	4.322924	-0.201275
	1	3.158710	4.823420	-0.304266
	6	0.987048	5.055691	-0.065696
	1	0.979904	6.144401	-0.054763
	6	-0.187637	4.318879	0.042553
	1	-1.165462	4.788187	0.135710
	6	-0.171933	2.932136	0.028526
	6	-1.446350	2.162708	0.103447
	6	0.563823	-2.656573	-0.105866
	6	2.013957	-2.280906	-0.025933
	6	2.990072	-3.261212	-0.027733
	1	2.640939	-4.288326	-0.115275
	6	4.341401	-2.933020	0.086376
	1	5.116076	-3.697913	0.085294
	6	4.657830	-1.560954	0.213677
	1	5.687935	-1.221648	0.319415

	6	3.639514	-0.635014	0.205867
	1	3.859460	0.426958	0.311694
	6	-2.124759	-0.169964	0.031472
	6	-1.576172	-1.503426	-0.051074
	6	-2.448033	-2.592550	-0.093235
	6	-3.837494	-2.415390	-0.056546
	6	-4.330508	-1.131199	0.022283
	6	-3.517549	-0.010682	0.067317
	1	-2.009874	-3.583857	-0.156106
	1	-4.520200	-3.262560	-0.089500
	1	-3.930454	0.990545	0.131505
	9	-5.691274	-0.950812	0.058909

Table S31. Cartesian coordinates of $[Ni(bpb\text{-}CF_3)(MeCN)_2]^n$, n=2+, 1+, 0, and $[Ni(bpb\text{-}CF_3)]^n$, n=1-, 2-.

Complexes	*Cartesian coordinates*			
$[Ni(bpb\text{-}CF_3)(MeCN)_2]^{2+}$	28	-1.095802	0.076324	-0.000158
	7	-1.803576	1.923785	0.180812
	7	0.576562	0.939913	0.026395
	7	-0.027433	-1.472812	-0.028060
	7	-2.589715	-1.219095	-0.180143
	8	1.531487	3.024630	-0.186711
	8	-0.165876	-3.761219	0.181681
	6	-3.049205	2.325959	0.427583
	1	-3.794194	1.557250	0.596407
	6	-3.383755	3.671389	0.503025
	1	-4.412377	3.954866	0.704826
	6	-2.391617	4.626519	0.326009
	1	-2.628460	5.686083	0.373796
	6	-1.085258	4.202850	0.113834
	1	-0.253050	4.893785	0.005982
	6	-0.832037	2.845159	0.058915
	6	0.560248	2.327499	-0.061293
	6	-0.694408	-2.688271	0.058790
	6	-2.166858	-2.489482	-0.059474
	6	-3.030254	-3.567380	-0.114025
	1	-2.621781	-4.569025	-0.007182
	6	-4.382313	-3.325127	-0.324508

	1	-5.090734	-4.147894	-0.371926
	6	-4.807395	-2.015008	-0.500369
	1	-5.848414	-1.780077	-0.700925
	6	-3.878107	-0.986215	-0.425436
	1	-4.172770	0.043018	-0.593521
	6	1.663426	0.130426	0.004558
	6	1.313797	-1.271509	-0.004722
	6	2.323824	-2.253174	-0.000743
	6	3.629682	-1.836178	0.001309
	6	3.970077	-0.453713	0.001038
	6	3.015517	0.528549	0.002038
	1	2.066451	-3.304708	0.000235
	1	4.428058	-2.574140	0.002039
	1	3.283197	1.577709	0.001079
	7	-1.199102	-0.085927	2.145895
	7	-1.111024	0.272493	-2.146014
	6	-1.014600	0.345566	-3.289246
	6	-1.146543	-0.189652	3.289607
	6	-1.079497	-0.320867	4.725979
	1	-2.086526	-0.269130	5.153104
	1	-0.627564	-1.282915	4.990074
	1	-0.470559	0.487438	5.144686
	6	-0.892025	0.437929	-4.724972
	1	-1.787456	0.906414	-5.146814
	1	-0.015974	1.041887	-4.984470
	1	-0.775807	-0.563420	-5.153033
	6	5.440152	-0.097508	0.000095
	9	5.628539	1.217279	0.006502
	9	6.025140	-0.608377	-1.083306
	9	6.029482	-0.619634	1.075714
$[Ni(bpb\text{-}CF_3)(MeCN)_2]^{1+}$	28	-1.155179	0.131774	-0.002289
	7	-2.650779	-0.917483	-0.624656
$[Ni^{2+}]$	7	-0.168604	-1.393146	-0.345808
	7	0.502314	0.904952	0.260801
	7	-1.839263	1.801705	0.682267
	8	-0.460796	-3.689698	-0.487554
	8	1.467517	3.011145	0.369119
	6	-3.874391	-0.524110	-0.969182
	1	-4.068771	0.542476	-0.928840
	6	-4.844845	-1.423070	-1.390919

	1	-5.830347	-1.058541	-1.663576
	6	-4.528978	-2.772483	-1.462540
	1	-5.272171	-3.497732	-1.782566
	6	-3.237726	-3.177096	-1.146980
	1	-2.911334	-4.210344	-1.221315
	6	-2.325555	-2.218882	-0.748768
	6	-0.886840	-2.556645	-0.512574
	6	0.512068	2.271625	0.442840
	6	-0.872778	2.736433	0.765591
	6	-1.121128	4.024815	1.198273
	1	-0.297278	4.731557	1.236386
	6	-2.412034	4.353740	1.593275
	1	-2.643990	5.356267	1.942351
	6	-3.392746	3.372356	1.563135
	1	-4.405276	3.575523	1.897721
	6	-3.064922	2.104089	1.102757
	1	-3.791807	1.298776	1.092940
	6	1.171029	-1.261490	-0.243830
	6	1.564014	0.091504	0.080825
	6	2.929364	0.425519	0.192300
	6	3.854804	-0.565777	-0.005864
	6	3.473019	-1.898352	-0.314389
	6	2.154808	-2.253576	-0.433345
	1	3.223595	1.441626	0.426513
	1	4.247708	-2.646797	-0.458717
	1	1.858413	-3.269331	-0.666787
	7	-1.112520	1.609337	-2.383211
	7	-1.805698	-1.149756	2.393661
	6	-1.322273	-2.130342	2.759036
	6	-0.201307	2.188902	-2.786588
	6	0.959775	2.916875	-3.257198
	1	0.648226	3.792631	-3.834838
	1	1.556911	3.247849	-2.400097
	1	1.574244	2.273381	-3.894470
	6	-0.702853	-3.370125	3.181835
	1	-1.405463	-3.958889	3.779682
	1	-0.408019	-3.951664	2.301130
	1	0.185293	-3.159711	3.785751
	6	5.327377	-0.270278	0.094592
	9	5.937756	-0.534877	-1.067058

	9	5.565680	1.003539	0.405061
	9	5.897214	-1.039701	1.029245
$[Ni(bpb\text{-}CF_3)(MeCN)_2]^0$	28	-1.103764	0.055243	0.001342
	7	-1.849887	1.799664	0.362077
	7	0.513918	0.930129	0.095605
	7	-0.082918	-1.474447	-0.108137
	7	-2.582205	-1.134644	-0.350630
	8	1.420463	3.050130	-0.165854
	8	-0.269722	-3.780454	0.083644
	6	-3.082130	2.143725	0.729431
	1	-3.773061	1.329043	0.917540
	6	-3.453247	3.468728	0.909092
	1	-4.469478	3.704202	1.209702
	6	-2.507879	4.464710	0.707036
	6	-1.209098	4.100809	0.377405
	1	-0.404818	4.818895	0.246997
	6	-0.914226	2.758374	0.232843
	6	0.493433	2.274499	0.016981
	6	-0.728526	-2.656801	-0.055825
	6	-2.202110	-2.421414	-0.246251
	6	-3.092677	-3.467597	-0.396017
	1	-2.715949	-4.480405	-0.287497
	6	-4.415163	-3.176917	-0.703159
	6	-4.787421	-1.851463	-0.879140
	1	-5.799803	-1.579574	-1.161424
	6	-3.837085	-0.856993	-0.696688
	1	-4.069754	0.189105	-0.863518
	6	1.633048	0.103747	0.019911
	6	1.291746	-1.268770	-0.061221
	6	2.291652	-2.236706	-0.107347
	6	3.623791	-1.836771	-0.087592
	6	3.954278	-0.488426	-0.025951
	6	2.965720	0.494354	0.029863
	1	2.018831	-3.284566	-0.160455
	1	4.410099	-2.584472	-0.130412
	1	3.217580	1.548412	0.069486
	7	-2.483414	-0.463154	2.849188
	7	-2.001882	1.180266	-2.839381
	6	-0.867394	1.038545	-2.989244
	6	-1.385654	-0.786343	2.990609

	6	-0.001996	-1.186519	3.157257
	1	0.257705	-1.215285	4.219935
	1	0.159172	-2.176701	2.718638
	1	0.651488	-0.473426	2.643836
	6	0.560084	0.854290	-3.164435
	1	0.809794	0.832092	-4.229660
	1	1.108685	1.670376	-2.682017
	1	0.869799	-0.088366	-2.701227
	1	-5.141891	-3.975586	-0.828578
	1	-2.774264	5.511497	0.829659
	6	5.386223	-0.063505	0.042291
	9	6.223113	-1.014299	-0.403341
	9	5.618533	1.042936	-0.683276
	9	5.764778	0.222975	1.303168
$[Ni(bpb\text{-}CF_3)]^{1-}$	28	-1.419801	0.165646	0.002735
	7	-1.960638	2.153698	-0.000861
	7	0.399789	0.988203	0.013092
	7	-0.325869	-1.509029	0.009028
	7	-2.941230	-1.214469	-0.005693
	8	1.497988	3.055467	0.016354
	8	-0.520040	-3.843164	0.009251
	6	-3.160352	2.738957	-0.007544
	1	-4.014195	2.065062	-0.011758
	6	-3.346822	4.113920	-0.009221
	1	-4.352784	4.525971	-0.014773
	6	-2.222674	4.931124	-0.003688
	1	-2.323277	6.014832	-0.004801
	6	-0.970852	4.335105	0.003265
	1	-0.042535	4.899164	0.007858
	6	-0.869107	2.948482	0.004561
	6	0.510232	2.308893	0.012275
	6	-0.946965	-2.681329	0.006270
	6	-2.453517	-2.473827	-0.002474
	6	-3.288952	-3.584763	-0.007104
	1	-2.813796	-4.561556	-0.004038
	6	-4.664520	-3.409225	-0.015356
	1	-5.335329	-4.266231	-0.019138
	6	-5.167511	-2.113732	-0.018729
	1	-6.236455	-1.915994	-0.025190
	6	-4.267244	-1.057973	-0.013633

	1	-4.620637	-0.029349	-0.016007
	6	1.438236	0.078899	0.018492
	6	1.035792	-1.301314	0.016489
	6	2.018845	-2.297144	0.026277
	6	3.367030	-1.963345	0.035696
	6	3.753258	-0.627113	0.041009
	6	2.797913	0.390295	0.029193
	1	1.698428	-3.333597	0.028928
	1	4.119714	-2.747600	0.048567
	1	3.088157	1.435823	0.038370
	6	5.193777	-0.266133	-0.012291
	9	5.626780	-0.003482	-1.270414
	9	5.998329	-1.250462	0.447708
	9	5.488201	0.834998	0.707224
$[Ni(bpb\text{-}CF_3)]^{2-}$	28	1.426605	0.172947	0.008617
	7	1.958013	2.088111	-0.073369
	7	-0.399846	0.998319	0.063709
	7	0.323412	-1.493128	-0.067279
	7	2.912363	-1.187660	0.089301
	8	-1.500382	3.062596	0.160773
	8	0.473233	-3.830078	-0.200040
	6	3.173054	2.672106	-0.192576
	1	4.007550	1.979040	-0.291596
	6	3.389644	4.031586	-0.201746
	1	4.406463	4.409837	-0.301819
	6	2.286686	4.904907	-0.083314
	1	2.414876	5.985811	-0.084051
	6	1.025166	4.319648	0.024432
	1	0.111303	4.904810	0.105172
	6	0.876009	2.944435	0.024484
	6	-0.493328	2.332488	0.097237
	6	0.959803	-2.684487	-0.113750
	6	2.430153	-2.488290	-0.031363
	6	3.286987	-3.582405	-0.045543
	1	2.818709	-4.560015	-0.145160
	6	4.660612	-3.423731	0.069847
	1	5.334729	-4.278712	0.058586
	6	5.149752	-2.095971	0.213318
	1	6.213141	-1.887125	0.322829
	6	4.248767	-1.056598	0.216035

	1	4.599545	-0.030600	0.333727
	6	-1.439848	0.096208	0.036192
	6	-1.029896	-1.289993	-0.045202
	6	-2.022272	-2.281914	-0.081265
	6	-3.372451	-1.950291	-0.041405
	6	-3.762949	-0.617757	0.041879
	6	-2.798918	0.401610	0.075887
	1	-1.696182	-3.315221	-0.140712
	1	-4.123706	-2.737390	-0.067006
	1	-3.082379	1.447371	0.145643
	6	-5.194185	-0.255751	0.020574
	9	-5.509752	0.763456	0.854708
	9	-5.644888	0.150881	-1.202420
	9	-6.014927	-1.282642	0.364199

Table S32. Cartesian coordinates of $[Ni(bpb\text{-}NO_2)(MeCN)_2]^n$, n=2+, 1+, 0, and $[Ni(bpb\text{-}NO_2)]^n$, n=1-, 2-.

Complexes	*Cartesian coordinates*			
$[Ni(bpb\text{-}NO_2)(MeCN)_2]^{2+}$	28	0.863371	0.077836	0.000273
	7	2.367340	-1.205039	0.179939
	7	-0.192675	-1.480705	0.029066
	7	-0.816624	0.927041	-0.025398
	7	1.553725	1.932604	-0.181778
	8	-0.035306	-3.767677	-0.179577
	8	-1.791430	3.002400	0.188971
	6	3.653585	-0.960397	0.424713
	1	3.939280	0.071501	0.591848
	6	4.591999	-1.981015	0.500287
	1	5.630927	-1.736579	0.700443
	6	4.178694	-3.295008	0.325647
	1	4.894422	-4.111408	0.373673
	6	2.828703	-3.549505	0.115577
	1	2.429362	-4.554973	0.009709
	6	1.955819	-2.479377	0.060290
	6	0.485490	-2.691468	-0.057392
	6	-0.813051	2.317307	0.061936
	6	0.574323	2.846092	-0.060129
	6	0.815491	4.205892	-0.116592
	1	-0.022527	4.889928	-0.009094

	6	2.118142	4.640709	-0.329864
	1	2.345687	5.702261	-0.378935
	6	3.118296	3.693945	-0.506178
	1	4.144402	3.986059	-0.708597
	6	2.795607	2.345506	-0.429265
	1	3.547547	1.583462	-0.597474
	6	-1.534681	-1.292073	0.005606
	6	-1.894839	0.109150	-0.003368
	6	-3.249383	0.498111	-0.000650
	6	-4.178091	-0.503815	0.000126
	6	-3.849110	-1.880524	-0.000652
	6	-2.536798	-2.281852	0.001103
	1	-3.551453	1.538652	0.000724
	1	-2.271276	-3.331357	-0.000241
	7	0.876705	0.276602	2.144351
	7	0.965772	-0.083315	-2.144233
	6	0.919223	-0.185082	-3.288456
	6	0.784996	0.353397	3.287807
	6	0.668423	0.450493	4.723642
	1	1.569988	0.911101	5.141107
	1	-0.200615	1.063835	4.984810
	1	0.543834	-0.548597	5.154654
	6	0.859694	-0.313616	-4.725328
	1	1.864109	-0.216732	-5.150817
	1	0.450095	-1.293263	-4.993829
	1	0.216971	0.469317	-5.141840
	7	-5.615943	-0.124500	-0.000082
	8	-6.406499	-1.037877	-0.005095
	8	-5.863183	1.056160	0.004624
	1	-4.657095	-2.606700	-0.001404
$[Ni(bpb\text{-}NO_2)(MeCN)_2]^{1+}$	28	0.937597	0.122790	0.004194
	7	1.602345	1.808803	-0.661789
$[Ni^{2+}]$	7	-0.726809	0.875474	-0.262691
	7	-0.030321	-1.417864	0.322628
	7	2.442919	-0.913281	0.621755
	8	-1.720450	2.969262	-0.338510
	8	0.286163	-3.710332	0.464472
	6	2.824374	2.131336	-1.078578
	1	3.560361	1.334686	-1.081248
	6	3.136856	3.409792	-1.521401

	1	4.146819	3.629101	-1.853573
	6	2.145079	4.380298	-1.537719
	1	2.365423	5.390239	-1.872634
	6	0.857875	4.030752	-1.148153
	1	0.025648	4.728109	-1.177394
	6	0.624941	2.733601	-0.733743
	6	-0.754137	2.247100	-0.421837
	6	0.702943	-2.575550	0.490171
	6	2.135934	-2.220547	0.730947
	6	3.061032	-3.170682	1.118038
	1	2.749044	-4.209164	1.180272
	6	4.347104	-2.752150	1.436416
	1	5.100592	-3.470802	1.747021
	6	4.644582	-1.397900	1.378886
	1	5.625657	-1.023087	1.653562
	6	3.661603	-0.507380	0.968395
	1	3.842366	0.561929	0.938696
	6	-1.778954	0.048819	-0.088923
	6	-1.369982	-1.304146	0.222001
	6	-2.341336	-2.311100	0.400951
	6	-3.666438	-1.975811	0.289098
	6	-4.042784	-0.645515	0.000005
	6	-3.145739	0.370768	-0.191701
	1	-2.032338	-3.325423	0.623645
	1	-4.447126	-2.718119	0.421585
	1	-3.476530	1.379201	-0.410557
	7	1.699646	-1.153332	-2.379889
	7	0.815714	1.534556	2.390597
	6	-0.087462	2.100929	2.829152
	6	1.064994	-1.999712	-2.837574
	6	0.262239	-3.075199	-3.383002
	1	0.830991	-3.624389	-4.139846
	1	-0.019915	-3.765907	-2.580611
	1	-0.643970	-2.672364	-3.846274
	6	-1.238199	2.815014	3.343690
	1	-0.915827	3.712125	3.881433
	1	-1.886835	3.112371	2.512174
	1	-1.802862	2.176235	4.029841
	7	-5.484182	-0.333590	-0.102294
	8	-6.252288	-1.262416	0.017251

	8	-5.785494	0.821670	-0.297272
$[Ni(bpb\text{-}NO_2)(MeCN)_2]^0$	28	0.876966	0.045942	-0.006145
	7	1.644593	1.783065	-0.355946
	7	-0.730326	0.936416	-0.121176
	7	-0.162210	-1.473358	0.087513
	7	2.337368	-1.158438	0.368814
	8	-1.617547	3.062558	0.149755
	8	0.004599	-3.782851	-0.080417
	6	2.884407	2.115216	-0.708702
	1	3.568903	1.293944	-0.891765
	6	3.270774	3.436901	-0.880728
	1	4.292577	3.662760	-1.169567
	6	2.333561	4.442079	-0.686502
	1	2.611961	5.486345	-0.803517
	6	1.027470	4.090822	-0.372028
	1	0.229425	4.817038	-0.248323
	6	0.717623	2.751203	-0.234365
	6	-0.696811	2.280963	-0.033370
	6	0.473991	-2.664651	0.055225
	6	1.946695	-2.442066	0.264807
	6	2.824346	-3.496247	0.432232
	1	2.439795	-4.506109	0.323755
	6	4.145315	-3.216911	0.756428
	1	4.862312	-4.021968	0.895813
	6	4.527775	-1.894345	0.931246
	1	5.538767	-1.630926	1.226219
	6	3.589906	-0.891421	0.731059
	1	3.830696	0.153097	0.895790
	6	-1.857892	0.122414	-0.054230
	6	-1.529907	-1.255755	0.029413
	6	-2.539616	-2.218577	0.062995
	6	-3.866207	-1.812445	0.031753
	6	-4.165761	-0.458498	-0.033612
	6	-3.184590	0.527826	-0.077669
	1	-2.275194	-3.268458	0.114448
	1	-4.674596	-2.535127	0.059792
	1	-3.449684	1.577070	-0.126415
	7	2.313671	-0.480828	-2.823217
	7	1.706491	1.167956	2.840179
	6	0.566003	1.078010	2.984199

	6	1.209371	-0.766319	-2.991812
	6	-0.182963	-1.118392	-3.191443
	1	-0.422839	-1.122585	-4.259118
	1	-0.384334	-2.109503	-2.772153
	1	-0.822160	-0.391499	-2.679343
	6	-0.869537	0.958521	3.149238
	1	-1.130318	0.977277	4.211858
	1	-1.379204	1.782644	2.638244
	1	-1.213740	0.016114	2.710532
	7	-5.569773	-0.046575	-0.057407
	8	-6.415729	-0.920839	-0.030991
	8	-5.808801	1.144366	-0.102138
$[Ni(bpb\text{-}NO_2)]^{1-}$	28	1.149980	-0.166705	-0.000012
	7	2.669905	1.207092	0.000046
	7	0.054931	1.510441	-0.000052
	7	-0.670789	-0.982735	0.000017
	7	1.686183	-2.159865	-0.000067
	8	0.259748	3.843083	-0.000130
	8	-1.775543	-3.044714	0.000052
	6	3.995456	1.044172	0.000127
	1	4.343752	0.014026	0.000195
	6	4.900176	2.095953	0.000133
	1	5.968098	1.892990	0.000201
	6	4.404204	3.393935	0.000051
	1	5.079128	4.247503	0.000051
	6	3.029255	3.576135	-0.000027
	1	2.560186	4.555809	-0.000086
	6	2.189562	2.469175	-0.000024
	6	0.684076	2.682962	-0.000079
	6	-0.785504	-2.304298	0.000024
	6	0.591691	-2.949529	-0.000019
	6	0.686883	-4.336574	-0.000020
	1	-0.243640	-4.896986	0.000021
	6	1.936208	-4.937518	-0.000074
	1	2.032235	-6.021574	-0.000076
	6	3.063911	-4.125302	-0.000131
	1	4.067986	-4.541797	-0.000179
	6	2.883442	-2.749657	-0.000126
	1	3.740260	-2.079567	-0.000176
	6	-1.300543	1.308598	-0.000032

	6	-1.708480	-0.074344	0.000015
	6	-3.066309	-0.381949	0.000053
	6	-4.003721	0.651352	0.000044
	6	-3.626733	1.990896	-0.000006
	6	-2.281262	2.313294	-0.000044
	1	-3.382446	-1.418387	0.000091
	1	-4.391398	2.760617	-0.000013
	1	-1.955300	3.347683	-0.000084
	7	-5.414830	0.323941	0.000087
	8	-5.740407	-0.852968	0.000127
	8	-6.225043	1.243537	0.000073
$[Ni(bpb\text{-}NO_2)]^{2-}$	28	1.142773	-0.183277	-0.001409
	7	2.669299	1.195474	-0.005279
	7	0.043771	1.481040	0.004050
	7	-0.685516	-1.017452	-0.002717
	7	1.690558	-2.103677	0.002063
	8	0.197820	3.828516	0.016265
	8	-1.753230	-3.104493	-0.009548
	6	3.998937	1.075260	-0.015808
	1	4.370043	0.048815	-0.024661
	6	4.899322	2.124333	-0.016797
	1	5.969501	1.928339	-0.026113
	6	4.372044	3.445842	-0.005419
	1	5.033181	4.312311	-0.004724
	6	3.005935	3.599684	0.004036
	1	2.527551	4.576745	0.011958
	6	2.148391	2.485556	0.003038
	6	0.694017	2.680006	0.008779
	6	-0.773753	-2.340461	-0.005619
	6	0.617204	-2.946299	-0.001465
	6	0.762988	-4.324164	0.000968
	1	-0.153266	-4.909343	-0.002510
	6	2.033196	-4.896547	0.008448
	1	2.164583	-5.977391	0.010827
	6	3.132458	-4.035729	0.013621
	1	4.153174	-4.414479	0.020443
	6	2.911843	-2.669569	0.010115
	1	3.745951	-1.971481	0.014518
	6	-1.293853	1.281531	0.003455
	6	-1.727313	-0.107669	-0.001361

	6	-3.079021	-0.409990	-0.003532
	6	-4.036182	0.623935	-0.000803
	6	-3.631443	1.966040	0.004147
	6	-2.291955	2.287178	0.006325
	1	-3.394401	-1.446782	-0.007224
	1	-4.394089	2.739314	0.006241
	1	-1.957879	3.319348	0.010433
	7	-5.418738	0.315974	-0.002818
	8	-5.774644	-0.870585	-0.007565
	8	-6.246759	1.245714	-0.000354

Table S33. Cartesian coordinates of the $[Ni(bpb)(DMF)_2]^n$ complexes, n=2+, 1+, 0, 1-, 2- charge states.

Complexes	*Cartesian coordinates*			
$[Ni(bpb)(DMF)_2]^{2+}$	Ni	0.00011300	0.00048200	-0.33896700
	N	0.08381400	1.60018700	-1.47314900
	N	-0.07146400	1.24005700	1.05970300
	N	0.07098900	-1.24329400	1.05601600
	N	-0.08302700	-1.59579200	-1.47800300
	O	-0.62119100	3.44702000	1.45101700
	O	0.62034600	-3.45146400	1.44096200
	C	0.39027500	1.69094100	-2.76442900
	H	0.69927300	0.77808300	-3.26185400
	C	0.35066800	2.90492200	-3.43806400
	H	0.60528000	2.93956900	-4.49303800
	C	-0.00766200	4.05263800	-2.74335200
	H	-0.05425700	5.01290100	-3.25000800
	C	-0.27527000	3.95882800	-1.38243500
	H	-0.51696800	4.82349700	-0.77003600
	C	-0.20636100	2.71583400	-0.78254400
	C	-0.35012400	2.55228200	0.69397800
	C	0.34973900	-2.55442400	0.68648300
	C	0.20671500	-2.71352500	-0.79059700
	C	0.27583900	-3.95472100	-1.39417600
	H	0.51716700	-4.82123900	-0.78424800
	C	0.00890900	-4.04443400	-2.75550200
	H	0.05568900	-5.00317900	-3.26500800
	C	-0.34898600	-2.89461500	-3.44695200
	H	-0.60307200	-2.92608300	-4.50215300
	C	-0.38884400	-1.68265600	-2.76970300
	H	-0.69751600	-0.76828900	-3.26455200
	C	-0.06975300	0.71773900	2.31255600

	C	0.06859500	-0.72473800	2.31042900
	C	0.15856000	-1.43044500	3.52541700
	C	0.08104800	-0.71328600	4.69432000
	C	-0.08350500	0.69910800	4.69640300
	C	-0.16038100	1.41977600	3.52961400
	H	0.28212400	-2.50643500	3.52318500
	H	0.14336800	-1.23710900	5.64463000
	H	-0.28393800	2.49577000	3.53056800
	C	-2.94952200	-0.42055400	0.15795000
	O	-2.09066400	0.16579600	-0.53194000
	H	-2.65476300	-1.16645200	0.90921500
	N	-4.24752400	-0.21599000	0.06393800
	C	-4.79872300	0.74643900	-0.87607400
	H	-5.46673800	0.23520200	-1.57738600
	H	-5.37143200	1.50423200	-0.33109000
	H	-3.98620400	1.22363600	-1.42385200
	C	-5.19770000	-0.93944400	0.89342100
	H	-5.77632200	-0.23349500	1.49836000
	H	-4.67235800	-1.62926800	1.55832800
	H	-5.88551800	-1.51050200	0.26102800
	O	2.09097100	-0.16426600	-0.53146900
	C	2.94952200	0.42023400	0.16036800
	N	4.24757100	0.21597100	0.06633200
	C	4.79918600	-0.74389200	-0.87605500
	H	3.98692000	-1.21945700	-1.42562500
	H	5.37149800	-1.50327900	-0.33287400
	H	5.46765800	-0.23077300	-1.57555500
	C	5.19738500	0.93723000	0.89813700
	H	5.77593300	0.22966200	1.50125200
	H	4.67173700	1.62508400	1.56484100
	H	5.88530800	1.51018000	0.26756900
	H	2.65442200	1.16407800	0.91353300
	H	-0.14633800	1.22006700	5.64825300
$[Ni(bpb)(DMF)_2]^{1+}$	Ni	0.00093800	-0.43920000	0.00053100
	N	0.31255500	-1.58737400	1.57770900
$[Ni^{3+}]$	N	0.34784500	0.95031200	1.19777300
	N	-0.34979600	0.94677300	-1.19979600
	N	-0.30737400	-1.59160500	-1.57413500
	O	1.31621300	1.35960800	3.24813900
	O	-1.32166900	1.34859500	-3.25001100
	C	0.06953700	-2.88488400	1.74552600
	H	-0.41015800	-3.40333600	0.92278400
	C	0.38086400	-3.54112500	2.92670600
	H	0.16938100	-4.60151900	3.02358500
	C	0.95116000	-2.81741400	3.96690400
	H	1.21186900	-3.30770000	4.90117100

	C	1.15412200	-1.45450900	3.80442800
	H	1.55362200	-0.81513300	4.58667900
	C	0.80807000	-0.87280500	2.59808500
	C	0.87807600	0.61045100	2.39588600
	C	-0.88010900	0.60266400	-2.39670100
	C	-0.80550100	-0.88077300	-2.59583600
	C	-1.15005000	-1.46611300	-3.80085100
	H	-1.55187400	-0.82968600	-4.58431600
	C	-0.94267500	-2.82868400	-3.96055000
	H	-1.20207500	-3.32177000	-4.89370800
	C	-0.36964100	-3.54835100	-2.91905100
	H	-0.15469800	-4.60824100	-3.01381200
	C	-0.06009400	-2.88864300	-1.73933400
	H	0.42158900	-3.40377100	-0.91566800
	C	0.20383600	2.24176700	0.67023400
	C	-0.21068000	2.23973200	-0.67465700
	C	-0.42280100	3.43987400	-1.35022600
	C	-0.21471700	4.63479300	-0.66803200
	C	0.19874200	4.63678600	0.65915000
	C	0.41141500	3.44393100	1.34355700
	H	-0.74959000	3.42608800	-2.38315000
	H	-0.38064200	5.57527100	-1.18712700
	H	0.73831900	3.43319100	2.37649200
	C	2.75300900	0.35940500	-0.69943600
	O	2.08202600	-0.65260100	-0.44371300
	H	2.28833900	1.35369000	-0.73756800
	N	4.05580900	0.35385100	-0.94335300
	C	4.82172600	-0.87627400	-0.92499600
	H	5.28905700	-1.03980400	-1.90271700
	H	5.60762700	-0.81464900	-0.16365100
	H	4.15475200	-1.70762400	-0.69460800
	C	4.77555300	1.57966200	-1.22673600
	H	5.55966600	1.73837400	-0.47771600
	H	4.09003200	2.43053400	-1.20337900
	H	5.23934700	1.52549400	-2.21824800
	O	-2.07957500	-0.65707700	0.44464300
	C	-2.75362600	0.35328100	0.69883100
	N	-4.05658300	0.34426900	0.94175500
	C	-4.81898400	-0.88805100	0.92410900
	H	-4.14926800	-1.71788000	0.69621100
	H	-5.60378700	-0.82992000	0.16135200
	H	-5.28747600	-1.05127500	1.90131900
	C	-4.78008100	1.56836700	1.22299100
	H	-5.56411000	1.72380700	0.47319500
	H	-4.09701600	2.42119200	1.19896800
	H	-5.24442600	1.51416600	2.21424000

	H	-2.29183200	1.34893900	0.73615500
	H	0.36101400	5.57885000	1.17652600
$[Ni(bpb)(DMF)_2]^{1+}$	Ni	0.69261200	-0.00165300	0.00078400
	N	1.83404400	1.53864700	0.23622000
$[Ni^{2+}]$	N	-0.67983700	1.17536800	0.38286600
	N	-0.68630800	-1.17026800	-0.38352600
	N	1.82503600	-1.54883000	-0.23301700
	O	-1.05025400	3.23711700	1.37735200
	O	-1.06785800	-3.22973300	-1.37842600
	C	3.11629800	1.70069200	-0.08126800
	H	3.59830700	0.87053400	-0.58615000
	C	3.79870100	2.88027300	0.18375300
	H	4.84524900	2.96674300	-0.09149900
	C	3.12212300	3.92926000	0.79027700
	H	3.63478900	4.85974600	1.01851800
	C	1.77009900	3.78005900	1.07148600
	H	1.16768300	4.57464300	1.50198900
	C	1.16072100	2.57795300	0.76581800
	C	-0.31751900	2.39696800	0.90546400
	C	-0.33067800	-2.39409400	-0.90544400
	C	1.14620900	-2.58409400	-0.76348700
	C	1.74871200	-3.78990500	-1.06817200
	H	1.14208500	-4.58085400	-1.49945500
	C	3.09942300	-3.94725000	-0.78511800
	H	3.60676000	-4.88085600	-1.01252400
	C	3.78151300	-2.90226300	-0.17786600
	H	4.82717000	-2.99497300	0.09873600
	C	3.10587700	-1.71856600	0.08612600
	H	3.59225100	-0.89128500	0.59154000
	C	-1.93274100	0.69008500	0.23424100
	C	-1.93649500	-0.67749200	-0.23675900
	C	-3.15886600	-1.33795500	-0.47377500
	C	-4.32836100	-0.65456100	-0.23843700
	C	-4.32467400	0.68169100	0.23258000
	C	-3.15141800	1.35796600	0.46954300
	H	-3.15569600	-2.35740300	-0.84129800
	H	-5.27972900	-1.14878400	-0.41778000
	H	-3.14227300	2.37741000	0.83702200
	C	-0.26381200	0.21916000	-3.06612000
	O	0.87485600	0.24356900	-2.62264900
	H	-0.69700500	-0.69768800	-3.50791000
	N	-1.13358500	1.24591300	-3.05872400
	C	-0.73239600	2.55385300	-2.59433700
	H	-0.76693000	3.27717100	-3.41858600
	H	-1.40415200	2.90080500	-1.79944900
	H	0.28921100	2.49065000	-2.21520700

	C	-2.47653400	1.11040800	-3.57429100
	H	-3.20960400	1.34738400	-2.79239200
	H	-2.64537500	0.08232300	-3.90653900
	H	-2.63864500	1.78534300	-4.42374400
	O	0.86664500	-0.25187600	2.62341300
	C	-0.27259600	-0.22001100	3.06494500
	N	-1.14906700	-1.24101800	3.05585900
	C	-0.75571100	-2.55142900	2.59172600
	H	0.26716100	-2.49497000	2.21494500
	H	-1.42799300	-2.89346900	1.79515000
	H	-0.79701300	-3.27486900	3.41555300
	C	-2.49200000	-1.09680300	3.56910700
	H	-3.22522900	-1.32873300	2.78584900
	H	-2.65464900	-0.06772300	3.90136300
	H	-2.66003200	-1.77089600	4.41807600
	H	-0.70052100	0.69961200	3.50610300
	H	-5.27329800	1.18146700	0.41050200
$[Ni(bpb)(DMF)_2]^0$	Ni	0.39627000	-0.02290900	-0.09347800
	N	1.67354900	-1.44159500	-0.43309800
	N	-0.75480800	-1.41057900	0.30551600
	N	-0.98061900	1.06286300	0.47959400
	N	1.23803200	1.64203000	-0.60747800
	O	-1.07287100	-3.66309700	-0.16596600
	O	-1.51957500	3.28438800	0.88939600
	C	2.99657500	-1.39858100	-0.58661600
	H	3.47183100	-0.44185400	-0.40283300
	C	3.73961500	-2.53075400	-0.89151200
	H	4.81624400	-2.44730300	-1.00409700
	C	3.08713900	-3.74743700	-1.03309900
	H	3.64348400	-4.64750200	-1.28308700
	C	1.71702600	-3.80071300	-0.81674000
	H	1.14141200	-4.72062000	-0.86124900
	C	1.04895100	-2.63245900	-0.49970500
	C	-0.40357600	-2.63968600	-0.11141000
	C	-0.78600200	2.39057500	0.48429100
	C	0.52414000	2.70180500	-0.18297200
	C	0.91953900	4.00427400	-0.42625700
	H	0.29104400	4.80559000	-0.04873000
	C	2.07797300	4.22417400	-1.15911100
	H	2.41769600	5.23562500	-1.36773100
	C	2.77971200	3.13002400	-1.64451800
	H	3.67244600	3.25139500	-2.25002800
	C	2.32247800	1.85407700	-1.34825700
	H	2.82318700	0.97413700	-1.73344600
	C	-2.01208400	-1.01505000	0.76072000
	C	-2.13293900	0.38845900	0.87945400

	C	-3.32546000	0.95340500	1.32757100
	C	-4.39278100	0.11608900	1.64743600
	C	-4.27401500	-1.26373600	1.52752900
	C	-3.08511600	-1.84055700	1.08590500
	H	-3.40072800	2.03104100	1.42112800
	H	-5.32517600	0.55361900	1.99677000
	H	-2.98142400	-2.91434300	0.97709500
	C	2.35832600	0.98477400	2.27411400
	O	3.37183000	0.63941100	1.69244800
	H	2.04004900	2.04479900	2.31789800
	N	1.50044000	0.17457700	2.92631100
	C	1.72354800	-1.25067200	2.97884900
	H	1.75657100	-1.59117000	4.02153000
	H	0.91373200	-1.77719000	2.45754400
	H	2.67609900	-1.47229600	2.49384900
	C	0.29295700	0.67269500	3.54137200
	H	-0.58748300	0.18312200	3.10846900
	H	0.19973900	1.74875200	3.36782300
	H	0.30618200	0.48799400	4.62370900
	O	-0.10026100	-0.17693200	-2.91624900
	C	-1.21353800	-0.63055800	-2.70487100
	N	-2.33200000	0.08713800	-2.48501400
	C	-2.29994900	1.52955900	-2.50391300
	H	-1.26558100	1.85086200	-2.64103000
	H	-2.68817100	1.93139900	-1.56032200
	H	-2.91190400	1.91579500	-3.33029600
	C	-3.60118900	-0.54376100	-2.21280600
	H	-3.99791800	-0.20303200	-1.24850000
	H	-3.47089700	-1.62831100	-2.15869500
	H	-4.32916000	-0.31007800	-3.00194900
	H	-1.40510300	-1.72008800	-2.66680900
	H	-5.11486500	-1.90577500	1.77978700
$[Ni(bpb)(DMF)_2]^{1-}$	Ni	-0.00028300	-0.23597400	-0.00038700
	N	1.54578300	-1.30720800	0.81136200
	N	1.18490400	1.28195000	0.54211700
	N	-1.18516700	1.28255200	-0.54210000
	N	-1.54667600	-1.30630500	-0.81238500
	O	3.46197700	1.70012700	0.88391100
	O	-3.46231300	1.70138900	-0.88272000
	C	1.66049700	-2.61247600	1.05555500
	H	0.76829000	-3.20476200	0.87204000
	C	2.82699700	-3.19013600	1.53740900
	H	2.86644700	-4.26130800	1.71582100
	C	3.92435600	-2.37150100	1.77167700
	H	4.85897100	-2.78956200	2.14124500
	C	3.80551800	-1.00978400	1.53138100

	H	4.61621300	-0.30518700	1.69324600
	C	2.59990300	-0.50587200	1.06056500
	C	2.44795300	0.98675100	0.81756500
	C	-2.44839500	0.98778600	-0.81715900
	C	-2.60072700	-0.50468100	-1.06089300
	C	-3.80654300	-1.00819300	-1.53163100
	H	-4.61719800	-0.30340400	-1.69286200
	C	-3.92559200	-2.36976200	-1.77266000
	H	-4.86036500	-2.78751500	-2.14218100
	C	-2.82825600	-3.18867500	-1.53919900
	H	-2.86790000	-4.25973300	-1.71825000
	C	-1.66155300	-2.61141800	-1.05733200
	H	-0.76935900	-3.20389700	-0.87432500
	C	0.66531700	2.53559000	0.26917200
	C	-0.66526400	2.53590100	-0.26842000
	C	-1.30662400	3.75710300	-0.49697400
	C	-0.65038400	4.95914800	-0.24308300
	C	0.65104000	4.95885300	0.24508600
	C	1.30698000	3.75651100	0.49836000
	H	-2.32311800	3.73797100	-0.87758200
	H	-1.16394300	5.90079900	-0.43344100
	H	2.32345100	3.73694700	0.87900700
	C	1.46508800	-1.28833500	-2.40882500
	O	1.71875600	-2.47235800	-2.26223000
	H	0.43766300	-0.92263700	-2.58906400
	N	2.36359300	-0.27734800	-2.38962800
	C	3.75682000	-0.53795600	-2.13149600
	H	4.36659800	-0.34490900	-3.02703000
	H	4.11316600	0.10597500	-1.31752500
	H	3.86198200	-1.58694700	-1.84441300
	C	1.95990900	1.09394600	-2.59413100
	H	2.40558400	1.73635700	-1.82693300
	H	0.87315600	1.17925600	-2.49428300
	H	2.27104500	1.45554000	-3.58596500
	O	-1.71653600	-2.47374500	2.26212000
	C	-1.46342800	-1.28966000	2.40906100
	N	-2.36246800	-0.27910800	2.39008700
	C	-3.75549400	-0.54048100	2.13163400
	H	-3.85979000	-1.58899500	1.84249300
	H	-4.11247600	0.10469900	1.31894500
	H	-4.36540800	-0.34978700	3.02758800
	C	-1.95963300	1.09234300	2.59497000
	H	-2.40454700	1.73463200	1.82722700
	H	-0.87279000	1.17808400	2.49653700
	H	-2.27216100	1.45385700	3.58639900
	H	-0.43622700	-0.92350500	2.58958700

	H	1.16484400	5.90027300	0.43592200
$[Ni(bpb)(DMF)_2]^{2-}$	Ni	0.00010500	-0.31450400	0.00015300
	N	1.47420500	-1.35245000	0.88003700
	N	1.14519900	1.22027700	0.61235100
	N	-1.14519700	1.22007800	-0.61220000
	N	-1.47404000	-1.35273800	-0.87973600
	O	3.37378300	1.68789400	1.16261000
	O	-3.37371700	1.68751900	-1.16292900
	C	1.60721600	-2.66761100	1.13886100
	H	0.75051500	-3.27962200	0.86058300
	C	2.72097700	-3.24122000	1.71129900
	H	2.74535400	-4.31691500	1.87537100
	C	3.81569500	-2.41579700	2.05156300
	H	4.72056200	-2.82995200	2.49313100
	C	3.69083700	-1.05498100	1.80266200
	H	4.47882700	-0.34165200	2.03544200
	C	2.53202900	-0.54021000	1.23899500
	C	2.39001500	0.92746500	1.00890200
	C	-2.38992700	0.92712300	-1.00905500
	C	-2.53177300	-0.54049900	-1.23913700
	C	-3.69040700	-1.05537600	-1.80313700
	H	-4.47831200	-0.34207500	-2.03629900
	C	-3.81524100	-2.41620600	-2.05182200
	H	-4.71994100	-2.83038900	-2.49370900
	C	-2.72067700	-3.24165700	-1.71091800
	H	-2.74508000	-4.31740100	-1.87466000
	C	-1.60710800	-2.66794500	-1.13822100
	H	-0.75058500	-3.27997800	-0.85942300
	C	0.64594800	2.47287900	0.31971800
	C	-0.64623900	2.47275000	-0.31939600
	C	-1.26248300	3.69758100	-0.59903700
	C	-0.62829600	4.90518700	-0.29521800
	C	0.62748200	4.90529800	0.29591300
	C	1.26193800	3.69778100	0.59953500
	H	-2.24634400	3.67285800	-1.05780100
	H	-1.12771300	5.84694400	-0.52824700
	H	2.24582300	3.67320000	1.05825800
	C	2.26878300	-1.31811000	-2.28114800
	O	2.79236500	-2.41958800	-2.27279200
	H	1.18256800	-1.17440400	-2.43778300
	N	2.91262300	-0.14172400	-2.09947300
	C	4.30450800	-0.12214800	-1.72980300
	H	4.93679600	0.22829100	-2.56309600
	H	4.44096000	0.54172500	-0.86633800
	H	4.59841600	-1.13931300	-1.45955700
	C	2.24763500	1.12387800	-2.29809000

	H	2.54748900	1.83069500	-1.51826800
	H	1.16254100	0.99609700	-2.21502300
	H	2.49812000	1.55224700	-3.28317300
	O	-2.79096100	-2.41958500	2.27408500
	C	-2.26825500	-1.31767100	2.28156900
	N	-2.91291300	-0.14198700	2.09842800
	C	-4.30473300	-0.12380900	1.72849200
	H	-4.59763500	-1.14127900	1.45830800
	H	-4.44154300	0.53979800	0.86489000
	H	-4.93757000	0.22614600	2.56158300
	C	-2.24920000	1.12430100	2.29700900
	H	-2.54964300	1.83069100	1.51701700
	H	-1.16397400	0.99753400	2.21418300
	H	-2.50029500	1.55255800	3.28199300
	H	-1.18219600	-1.17296600	2.43839800
	H	1.12668300	5.84713700	0.52907100

Table S34. Cartesian coordinates of the $[Ni(bpb\text{-}(NMe_2)_2)(DMF)_2]^n$ complexes, n=2+, 1+, 0.

Complexes	*Cartesian coordinates*			
$[Ni(bpb\text{-}(NMe_2)_2)(DMF)_2]^{2+}$	Ni	0.00006000	0.21694200	0.00010600
	N	1.57488800	-0.91297300	0.06269900
	N	1.24544500	1.61136200	-0.07321000
	N	-1.24525800	1.61139600	0.07359000
	N	-1.57482600	-0.91291400	-0.06274300
	O	3.45331000	2.00210500	-0.62426600
	O	-3.45324400	2.00220500	0.62409700
	C	1.69404100	-2.21406200	0.35060900
	H	0.79409500	-2.72384800	0.67857600
	C	2.88683500	-2.89154500	0.27968200
	H	2.89385900	-3.94329400	0.54065100
	C	4.07372300	-2.21769600	-0.11544600
	C	3.93645800	-0.82165700	-0.33910000
	H	4.77664100	-0.17505800	-0.57009700
	C	2.70482700	-0.23623200	-0.23656900
	C	2.55394300	1.24492000	-0.35942100
	C	-2.55383800	1.24499900	0.35942700
	C	-2.70477100	-0.23613800	0.23643500
	C	-3.93643800	-0.82151600	0.33880700
	H	-4.77661600	-0.17489100	0.56975700
	C	-4.07373800	-2.21754200	0.11510100
	C	-2.88682600	-2.89144100	-0.27986700
	H	-2.89385600	-3.94319300	-0.54082300
	C	-1.69399600	-2.21400300	-0.35064800

	H	-0.79403100	-2.72383300	-0.67849300
	C	0.72121100	2.86148300	-0.06881400
	C	-0.72099700	2.86150500	0.06916700
	C	-1.42308800	4.07923100	0.15963700
	C	-0.70550400	5.24723900	0.08265500
	C	0.70577800	5.24721800	-0.08236600
	C	1.42333000	4.07919000	-0.15932000
	H	-2.49890300	4.07667100	0.28366300
	H	-1.22820600	6.19804000	0.14566800
	H	2.49914500	4.07660100	-0.28335300
	C	-0.72896200	0.48821500	-2.90778200
	O	0.16200900	0.13819600	-2.11471800
	H	-1.73274800	0.75493700	-2.54721500
	N	-0.57445500	0.57418900	-4.21674700
	C	0.69664500	0.26604600	-4.84917000
	H	0.56737900	-0.56142700	-5.55523000
	H	1.05871700	1.14365000	-5.39545400
	H	1.42102000	-0.01423800	-4.08427700
	C	-1.66095900	0.98203100	-5.08940500
	H	-1.37921800	1.88547000	-5.64093500
	H	-2.55872900	1.19222200	-4.50274900
	H	-1.88481800	0.18577800	-5.80756400
	O	-0.16183800	0.13747300	2.11487100
	C	0.72873300	0.48835700	2.90800600
	N	0.57416600	0.57375500	4.21700100
	C	-0.69653000	0.26385500	4.84937800
	H	-1.42058400	-0.01709600	4.08442500
	H	-1.05966600	1.14085000	5.39593500
	H	-0.56621600	-0.56366600	5.55518900
	C	1.66019500	0.98268800	5.08974100
	H	1.37735500	1.88561700	5.64154300
	H	2.55768400	1.19415900	4.50311500
	H	1.88506400	0.18650400	5.80766000
	H	1.73217000	0.75643200	2.54747700
	H	1.22850500	6.19800500	-0.14540000
	N	5.24873100	-2.84486400	-0.24190500
	N	-5.24879400	-2.84466000	0.24139300
	C	5.35156400	-4.26933200	0.03434900
	H	6.37293100	-4.59883400	-0.15386300
	H	5.11208300	-4.49104800	1.08199600
	H	4.68573100	-4.84661000	-0.61864100
	C	6.43825900	-2.10002200	-0.62903500
	H	6.29314100	-1.60040500	-1.59406500
	H	6.70556700	-1.35215500	0.12787000
	H	7.27519200	-2.79008300	-0.73195700
	C	-5.35163200	-4.26914000	-0.03479500

	H	-6.37303900	-4.59859700	0.15327700
	H	-5.11200100	-4.49092200	-1.08239400
	H	-4.68591800	-4.84640600	0.61832600
	C	-6.43833400	-2.09977200	0.62839600
	H	-6.29338300	-1.60032600	1.59354100
	H	-6.70540600	-1.35175500	-0.12844200
	H	-7.27535800	-2.78976900	0.73100800
$[Ni(bpb\text{-}(NMe_2)_2)(DMF)_2]^{1+}$ $[Ni^{3+}]$	Ni	-0.00001600	0.09977800	-0.00002700
	N	1.57878000	-1.04540800	0.13897500
	N	1.25011200	1.48640400	0.00781100
	N	-1.25014100	1.48641000	-0.00778700
	N	-1.57881400	-1.04538100	-0.13907200
	O	3.49666800	1.89554600	-0.30303100
	O	-3.49669200	1.89554700	0.30312300
	C	1.69343900	-2.34492500	0.41641900
	H	0.77552700	-2.87727500	0.64167100
	C	2.90083500	-3.00365000	0.45751500
	H	2.90220300	-4.06038400	0.69768000
	C	4.10150600	-2.29646200	0.20561500
	C	3.96204900	-0.90678900	-0.01323000
	H	4.80294800	-0.23447200	-0.14809300
	C	2.71418800	-0.34188400	-0.02755900
	C	2.55128000	1.14478600	-0.14199600
	C	-2.55131600	1.14479800	0.14197400
	C	-2.71422300	-0.34187000	0.02751700
	C	-3.96208000	-0.90678700	0.01320900
	H	-4.80298200	-0.23447900	0.14808900
	C	-4.10152900	-2.29645700	-0.20566300
	C	-2.90085700	-3.00363100	-0.45760700
	H	-2.90222200	-4.06036600	-0.69776600
	C	-1.69346700	-2.34489600	-0.41653300
	H	-0.77555400	-2.87723000	-0.64181800
	C	0.70450400	2.77696600	-0.00698000
	C	-0.70452200	2.77697200	0.00708400
	C	-1.40909000	3.97941700	0.01693400
	C	-0.69493500	5.17404700	0.00956300
	C	0.69493000	5.17404200	-0.00932300
	C	1.40907800	3.97940900	-0.01676500
	H	-2.49237200	3.96549600	0.03571800
	H	-1.23862600	6.11537100	0.01915100
	H	2.49236000	3.96548300	-0.03555700
	C	0.09296600	0.93895600	-2.83946300
	O	0.15792300	-0.08342600	-2.14548000
	H	-0.07995000	1.92511100	-2.38757000
	N	0.22258100	0.96053000	-4.16178500
	C	0.46225800	-0.25387100	-4.91302400

	H	-0.34353300	-0.41082300	-5.63953000
	H	1.41377700	-0.17911400	-5.45228400
	H	0.50020700	-1.09630400	-4.22134500
	C	0.14442700	2.19980600	-4.90664000
	H	1.08151400	2.37710900	-5.44723600
	H	-0.02974600	3.03569600	-4.22419200
	H	-0.67776400	2.15737200	-5.63058800
	O	-0.15773000	-0.08350600	2.14543300
	C	-0.09292300	0.93884800	2.83946700
	N	-0.22252100	0.96033600	4.16179400
	C	-0.46202500	-0.25413600	4.91297200
	H	-0.49988200	-1.09653600	4.22124800
	H	-1.41354200	-0.17953100	5.45225900
	H	0.34380200	-0.41102500	5.63945000
	C	-0.14458200	2.19959100	4.90670600
	H	-1.08176800	2.37681400	5.44715900
	H	0.02965400	3.03551700	4.22431800
	H	0.67749500	2.15718400	5.63078300
	H	0.07985100	1.92505200	2.38762300
	H	1.23862800	6.11536500	-0.01885800
	N	5.30600600	-2.90172200	0.20016400
	N	-5.30602200	-2.90172900	-0.20019900
	C	6.50790500	-2.12025900	-0.02520100
	H	7.37096000	-2.78649000	-0.04149200
	H	6.46596900	-1.59869300	-0.98932500
	H	6.66347000	-1.37916200	0.76984100
	C	5.40826600	-4.32017900	0.47854300
	H	5.06311500	-4.55989500	1.49349100
	H	4.82622100	-4.90970200	-0.24160700
	H	6.45082900	-4.62873200	0.39641900
	C	-6.50791200	-2.12031800	0.02539200
	H	-6.46580400	-1.59874500	0.98950300
	H	-6.66367200	-1.37923300	-0.76962400
	H	-7.37093200	-2.78659000	0.04187600
	C	-5.40828800	-4.32014700	-0.47877800
	H	-5.06301200	-4.55972800	-1.49371400
	H	-4.82635500	-4.90979200	0.24136500
	H	-6.45087400	-4.62867600	-0.39684900
$[Ni(bpb\text{-}(NMe_2)_2)(DMF)_2]^{1+}$	Ni	0.00023800	-0.10347400	0.00019300
	N	1.54019900	-1.23837600	-0.07194700
$[Ni^{2+}]$	N	1.22672400	1.26771400	0.12874100
	N	-1.22586200	1.26827500	-0.12493400
	N	-1.53991000	-1.23805200	0.07007600
	O	3.44689500	1.64809300	0.67504600
	O	-3.44488900	1.64994000	-0.67504800
	C	1.66470400	-2.52714700	-0.39872300

	H	0.76192900	-3.02724500	-0.73372500
	C	2.86007700	-3.20816300	-0.35992400
	H	2.86565600	-4.25176000	-0.65172500
	C	4.04398800	-2.54440700	0.04094500
	C	3.90988700	-1.16129100	0.30782000
	H	4.75053300	-0.52115900	0.55228500
	C	2.67565100	-0.57259000	0.23220000
	C	2.53024600	0.90761600	0.39178100
	C	-2.52900100	0.90876700	-0.39124200
	C	-2.67490500	-0.57164800	-0.23436400
	C	-3.90918100	-1.16000700	-0.31185700
	H	-4.74948700	-0.51947700	-0.55645900
	C	-4.04381700	-2.54333600	-0.04631300
	C	-2.86058500	-3.20756500	0.35583300
	H	-2.86681900	-4.25125800	0.64727600
	C	-1.66504500	-2.52692300	0.39619400
	H	-0.76274600	-3.02729300	0.73208200
	C	0.71709200	2.52032500	0.09346400
	C	-0.71599200	2.52064000	-0.08714600
	C	-1.41258500	3.74158800	-0.18861200
	C	-0.69982300	4.91431800	-0.09308900
	C	0.70140300	4.91397800	0.10318800
	C	1.41392900	3.74091600	0.19684000
	H	-2.48521600	3.73422600	-0.34251700
	H	-1.22338600	5.86399100	-0.16837700
	H	2.48657300	3.73309100	0.35063500
	C	-0.49208800	1.00166900	-3.05620300
	O	-0.45210400	-0.16933900	-2.71804800
	H	-1.44973400	1.52120300	-3.25164500
	N	0.57174700	1.81589500	-3.20718900
	C	1.91577100	1.30955000	-3.05652800
	H	2.44020600	1.32075600	-4.02079100
	H	2.47825400	1.92940000	-2.34755900
	H	1.86198200	0.28333700	-2.68776300
	C	0.42284100	3.19896700	-3.59336100
	H	0.86857800	3.85653900	-2.83577400
	H	-0.63860400	3.44671100	-3.68297500
	H	0.91092900	3.39157600	-4.55720400
	O	0.44569900	-0.18690500	2.71781000
	C	0.48881300	0.98319200	3.05878000
	N	-0.57278900	1.80000600	3.21162100
	C	-1.91820500	1.29770000	3.05983300
	H	-1.86726700	0.27191900	2.68953100
	H	-2.47878300	1.92020200	2.35170200
	H	-2.44279800	1.30895400	4.02397300
	C	-0.42007500	3.18187200	3.60065800

	H	-0.86395600	3.84229000	2.84443500
	H	0.64206700	3.42650000	3.69084300
	H	-0.90768100	3.37384800	4.56487500
	H	1.44783500	1.49966300	3.25570600
	H	1.22520400	5.86340200	0.17996200
	N	5.22902000	-3.17727100	0.13635900
	N	-5.22880500	-3.17595500	-0.14392300
	C	6.41566600	-2.44032600	0.52991500
	H	6.28013800	-1.96603800	1.50976800
	H	7.25719300	-3.12948600	0.60491500
	H	6.67354200	-1.66691600	-0.20559300
	C	5.32871100	-4.58615400	-0.19125800
	H	4.66979700	-5.18973400	0.44612300
	H	5.07584600	-4.77479300	-1.24320100
	H	6.35271200	-4.92205700	-0.02624800
	C	-6.41453400	-2.43880500	-0.53988800
	H	-6.27651500	-1.96379700	-1.51904800
	H	-7.25580600	-3.12798600	-0.61758700
	H	-6.67437800	-1.66594600	0.19552200
	C	-5.32936700	-4.58485100	0.18338200
	H	-4.66977000	-5.18860200	-0.45315000
	H	-5.07799100	-4.77364900	1.23565700
	H	-6.35326000	-4.92040500	0.01698500
$[Ni(bpb\text{-}(NMe_2)_2)(DMF)_2]^0$	Ni	0.05204600	0.17110600	-0.11343600
	N	1.63589500	-0.92626600	0.01447300
	N	1.25743500	1.55915800	-0.30779100
	N	-1.23130000	1.46346200	-0.42434100
	N	-1.46115200	-0.97760100	0.21664400
	O	3.46051000	2.11115000	0.17887300
	O	-3.51707700	1.76053400	-0.70635500
	C	1.81250000	-2.24764500	-0.04219500
	H	0.94112500	-2.83949400	-0.29917900
	C	3.03684900	-2.85782400	0.13494500
	H	3.08608300	-3.93762400	0.05516500
	C	4.18277200	-2.07698400	0.39249400
	C	3.99432600	-0.67971400	0.37249200
	H	4.80058300	0.03800700	0.48119800
	C	2.73853600	-0.16456300	0.17303200
	C	2.53725500	1.31956200	0.02690300
	C	-2.51998200	1.09172400	-0.45441700
	C	-2.62678900	-0.35535300	-0.05821300
	C	-3.85529500	-0.95090700	0.08002300
	H	-4.71866400	-0.34075000	-0.16376000
	C	-3.94184500	-2.27750700	0.55128600
	C	-2.72043500	-2.88909200	0.90205300
	H	-2.68039200	-3.88789400	1.32043200

	C	-1.53502300	-2.20969600	0.71922600
	H	-0.59687600	-2.66737400	1.00966100
	C	0.67948900	2.80602800	-0.53845400
	C	-0.73171000	2.74882600	-0.62186000
	C	-1.46663900	3.91140700	-0.84894100
	C	-0.79349600	5.12429500	-0.98479200
	C	0.59296200	5.17976100	-0.90125500
	C	1.33942800	4.02431200	-0.68011600
	H	-2.54712700	3.85017100	-0.91587900
	H	-1.36515500	6.03289500	-1.16046600
	H	2.42034900	4.05328200	-0.60038400
	C	-0.73512700	-1.72116200	-2.73797800
	O	-0.20266400	-2.75446100	-2.37466000
	H	-1.83360200	-1.58269600	-2.70129700
	N	-0.10615100	-0.62908700	-3.21946500
	C	1.33277800	-0.59112000	-3.32909100
	H	1.63031800	-0.40541400	-4.36943000
	H	1.73531900	0.20826800	-2.69399100
	H	1.73246000	-1.55270000	-3.00166400
	C	-0.82335400	0.56303600	-3.60393100
	H	-0.46644100	1.42646100	-3.03031200
	H	-1.89103600	0.44072900	-3.39890800
	H	-0.68928900	0.76680300	-4.67497900
	O	0.25254000	0.16100300	2.88362600
	C	0.54440900	1.34083300	2.79646900
	N	-0.32597600	2.36966600	2.73070300
	C	-1.74819400	2.13188400	2.76669600
	H	-1.91939100	1.05461900	2.71638900
	H	-2.23519800	2.62089000	1.91471300
	H	-2.18326200	2.52494500	3.69627800
	C	0.11798500	3.73979500	2.64869700
	H	-0.32687800	4.23499900	1.77711300
	H	1.20515900	3.76937800	2.53190200
	H	-0.15962900	4.29609100	3.55549000
	H	1.59625200	1.68519500	2.75591700
	H	1.10668000	6.13255300	-1.00857500
	N	5.40100100	-2.63129700	0.61747500
	N	-5.13283100	-2.91490600	0.67805800
	C	6.55361900	-1.77910500	0.81467600
	H	6.40192500	-1.09699400	1.66084400
	H	7.42439500	-2.39867600	1.03590300
	H	6.77801400	-1.17847000	-0.07817900
	C	5.56363600	-4.06559000	0.55234800
	H	4.91069400	-4.57328500	1.27485900
	H	5.34414800	-4.45932400	-0.45081000
	H	6.59535500	-4.32274500	0.79794200

	C	-6.35949200	-2.21750900	0.35746500
	H	-6.35808000	-1.86766000	-0.68292900
	H	-7.20268900	-2.89921700	0.48063200
	H	-6.52110800	-1.35084100	1.01365300
	C	-5.18030900	-4.25361900	1.22053400
	H	-4.58105400	-4.94930300	0.61791200
	H	-4.81639500	-4.28874100	2.25759400
	H	-6.21193800	-4.60918800	1.21262200

Table S35. Cartesian coordinates of the $[Ni(bpb\text{-}F)(DMF)_2]^n$ complexes, n=2+, 1+, 0.

Complexes	*Cartesian coordinates*			
$[Ni(bpb\text{-}F)(DMF)_2]^{2+}$	Ni	0.46105300	0.18103500	0.06495100
	N	1.23120100	0.27327500	1.86872600
	N	-1.04460800	-0.46275800	0.96201600
	N	-0.55331400	-0.20185100	-1.46251200
	N	1.74144100	0.91934500	-1.22861200
	O	-2.01566900	-0.44779800	3.05186900
	O	-0.29652000	-0.50343400	-3.73614700
	C	2.49948200	0.45922800	2.22570900
	H	3.23408600	0.49941300	1.42880800
	C	2.87208900	0.56183400	3.55962200
	H	3.91680400	0.71529200	3.81266500
	C	1.89792800	0.45829900	4.54397200
	H	2.16320500	0.54394800	5.59436200
	C	0.58290500	0.21268900	4.16569700
	H	-0.22049200	0.07667300	4.88492300
	C	0.29328700	0.11514800	2.81822300
	C	-1.06465800	-0.27454600	2.33434000
	C	0.12312800	-0.11883600	-2.67652200
	C	1.43061000	0.57495600	-2.49019300
	C	2.22360200	0.92180900	-3.56758900
	H	1.91972000	0.61148000	-4.56381300
	C	3.36753000	1.67587000	-3.33194800
	H	4.01669200	1.96532400	-4.15400300
	C	3.65007300	2.07920900	-2.03371100
	H	4.51177900	2.70095200	-1.81030000
	C	2.80686200	1.68309800	-1.00333400
	H	2.97307900	2.00594400	0.01858400
	C	-2.09009200	-0.83357300	0.17214100
	C	-1.77775100	-0.73361800	-1.23909400
	C	-2.71988900	-1.14599100	-2.21055900
	C	-3.93581800	-1.60538400	-1.78442000
	C	-4.22792100	-1.66371000	-0.39701600
	C	-3.34018000	-1.29049800	0.58933600

	H	-2.47351800	-1.09084500	-3.26389300
	H	-4.69814000	-1.92930800	-2.48760600
	H	-3.60924300	-1.35784100	1.63640000
	C	-1.03770000	2.78930500	-0.24783600
	O	-0.20787100	2.14466200	0.42508900
	H	-1.44033800	2.38195500	-1.18583100
	N	-1.50134800	3.97790700	0.08020500
	C	-1.07552700	4.64990900	1.29722000
	H	-0.59843800	5.60266500	1.04410600
	H	-1.94685500	4.84807100	1.93026300
	H	-0.36902400	4.01586900	1.83274300
	C	-2.46243400	4.68126900	-0.75407800
	H	-3.37884000	4.87210100	-0.18584500
	H	-2.70887000	4.08379900	-1.63514100
	H	-2.04253500	5.63844700	-1.08056200
	O	1.46386400	-1.64711200	-0.23019700
	C	1.15698100	-2.74601900	0.27574200
	N	1.75152900	-3.88758300	-0.00447100
	C	2.84245900	-3.96103000	-0.96293100
	H	3.03936100	-2.96716400	-1.36466600
	H	2.57010300	-4.64092200	-1.77707300
	H	3.74141800	-4.34411800	-0.46845400
	C	1.35001300	-5.13421500	0.62744900
	H	1.02688200	-5.85059600	-0.13510800
	H	0.52498300	-4.95899900	1.32213200
	H	2.19412200	-5.56130800	1.17897300
	H	0.34203500	-2.81887900	1.00931700
	F	-5.41245300	-2.10455900	-0.05036800
$[Ni(bpb\text{-}F)(DMF)_2]^{1+}$	Ni	-0.59212200	-0.09398300	0.06552500
	N	-2.02148100	-0.03815500	-1.29625600
$[Ni^{3+}]$	N	0.49422100	0.45187800	-1.34969200
	N	1.01411000	-0.15770300	1.01497400
	N	-1.39143800	-0.54868500	1.81466000
	O	0.38703700	1.40639800	-3.44158000
	O	1.92087000	-0.97286300	2.96927600
	C	-3.26475600	-0.50923600	-1.24138500
	H	-3.54604300	-1.04481900	-0.34150100
	C	-4.15253500	-0.35607800	-2.29554500
	H	-5.15974700	-0.75218600	-2.21097000
	C	-3.72422800	0.29694000	-3.44495200
	H	-4.40111100	0.43710100	-4.28362600
	C	-2.41115100	0.73966600	-3.51607800
	H	-1.99165500	1.21829700	-4.39659800
	C	-1.58538000	0.54058100	-2.42442500
	C	-0.12603900	0.87459600	-2.47478600
	C	0.97539400	-0.69937700	2.25603400

	C	-0.44348500	-0.88032500	2.70311700
	C	-0.74903100	-1.28222300	3.99071400
	H	0.06993300	-1.53955800	4.65662400
	C	-2.08113500	-1.31266000	4.37820200
	H	-2.35790000	-1.62368100	5.38210600
	C	-3.05426200	-0.90997600	3.47184400
	H	-4.10484400	-0.88188900	3.74396300
	C	-2.66594900	-0.52821500	2.19648300
	H	-3.38864500	-0.17219800	1.47072300
	C	1.86031900	0.55213600	-1.04708400
	C	2.15367600	0.18684800	0.28012200
	C	3.46386100	0.20651700	0.75105100
	C	4.45538900	0.59882500	-0.13443200
	C	4.19166500	0.96664000	-1.44340300
	C	2.88216900	0.94326800	-1.91012100
	H	3.70457800	-0.08080600	1.76717900
	H	2.64670800	1.23083100	-2.92783100
	C	-0.20039500	2.78171900	0.60812900
	O	-1.09892400	1.92868400	0.53674100
	H	0.85392800	2.50811400	0.46827300
	N	-0.40488300	4.06856000	0.84933100
	C	-1.73986100	4.59576800	1.05072100
	H	-1.81318800	5.05725200	2.04200500
	H	-1.95934100	5.35424600	0.29075200
	H	-2.46033400	3.78104200	0.97215100
	C	0.69610900	5.00957400	0.91099300
	H	0.57319200	5.78436900	0.14578200
	H	1.64248900	4.49126300	0.73739400
	H	0.73003800	5.48781100	1.89646100
	O	-0.50854000	-2.19338800	-0.32775800
	C	0.54095100	-2.68940400	-0.76678700
	N	0.72073200	-3.98175300	-0.99781900
	C	-0.32940900	-4.94733200	-0.74166400
	H	-1.20834900	-4.42559800	-0.36180300
	H	0.01089600	-5.67996600	-0.00109500
	H	-0.58598100	-5.47409300	-1.66777800
	C	1.98020400	-4.49038300	-1.50432200
	H	2.41107400	-5.20517100	-0.79413600
	H	2.68618300	-3.66844700	-1.64727300
	H	1.82535600	-4.99534700	-2.46456000
	H	1.41251300	-2.06155100	-0.99545700
	H	5.01044300	1.26891300	-2.08928400
	F	5.71823500	0.62008300	0.30215800
$[Ni(bpb\text{-}F)(DMF)_2]^{1+}$	Ni	-0.83922600	-0.11740500	0.00819500
	N	-1.67016800	-1.84694100	0.25447900
$[Ni^{2+}]$	N	0.73479500	-1.02502400	0.35705300

	N	0.29646200	1.28693000	-0.39127000
	N	-2.24033500	1.19816100	-0.20084000
	O	1.51074800	-2.99708100	1.29277800
	O	0.27679600	3.37125300	-1.40527800
	C	-2.91059000	-2.24000900	-0.02567400
	H	-3.55298700	-1.50962700	-0.50557800
	C	-3.35726500	-3.52594600	0.24658100
	H	-4.37859900	-3.80092400	0.00208800
	C	-2.48153500	-4.43649200	0.82083800
	H	-2.80773800	-5.44650200	1.05364600
	C	-1.17188500	-4.04293100	1.06465500
	H	-0.42113100	-4.71539800	1.46941800
	C	-0.80248800	-2.74791500	0.75498000
	C	0.62116900	-2.29834700	0.86005600
	C	-0.28919800	2.42123500	-0.91348700
	C	-1.77213900	2.33828100	-0.74400500
	C	-2.59005500	3.41301700	-1.03674100
	H	-2.14700800	4.30019300	-1.47968400
	C	-3.94127900	3.32203600	-0.72763000
	H	-4.61471700	4.14653600	-0.94529900
	C	-4.40929200	2.17246000	-0.10705500
	H	-5.44876800	2.07387100	0.19004800
	C	-3.52389800	1.13268100	0.14362000
	H	-3.84094300	0.23246600	0.65888400
	C	1.87367800	-0.31536300	0.19262700
	C	1.61553800	1.03296200	-0.26790900
	C	2.69201100	1.91225900	-0.52156900
	C	3.97220000	1.46659100	-0.31376900
	C	4.19612100	0.14971400	0.14632600
	C	3.19011000	-0.74995300	0.40271400
	H	2.49224400	2.91467600	-0.88149000
	H	4.83149900	2.10567600	-0.49537800
	H	3.40493800	-1.74898100	0.76287100
	C	0.13527600	-0.11443300	-3.10522100
	O	-0.97613400	-0.29108100	-2.62860300
	H	0.42507600	0.85019000	-3.56238000
	N	1.13789000	-1.01193700	-3.11893300
	C	0.93396500	-2.35940600	-2.63864000
	H	1.03765200	-3.07661000	-3.46246700
	H	1.67397100	-2.60670200	-1.86745500
	H	-0.07284100	-2.43262400	-2.22363800
	C	2.42994400	-0.70159700	-3.68623900
	H	3.21789500	-0.83904700	-2.93453000
	H	2.44883100	0.33874500	-4.02277500
	H	2.64747700	-1.35166100	-4.54267200
	O	-1.06069500	0.10247300	2.62127700

	C	0.06895400	0.26002200	3.05988700
	N	0.76006500	1.41509400	3.05603600
	C	0.14698400	2.64290500	2.60430000
	H	-0.85299900	2.41586400	2.23019800
	H	0.74725400	3.10109000	1.80841500
	H	0.06788800	3.35665900	3.43377700
	C	2.10566300	1.49958700	3.57543900
	H	2.78956600	1.87121200	2.80134900
	H	2.44578400	0.50949400	3.89155200
	H	2.15114100	2.17804200	4.43630400
	H	0.64744200	-0.57667100	3.49416400
	F	5.45319800	-0.22102800	0.33551200
$[Ni(bpb\text{-}F)(DMF)_2]^0$	Ni	-0.54520000	0.09930100	-0.10028700
	N	-1.49996200	1.76912000	-0.35731400
	N	0.91518200	1.19730500	0.17309100
	N	0.60212000	-1.26383900	0.37662800
	N	-1.77523700	-1.33951300	-0.50419800
	O	1.68336400	3.32363800	-0.35514800
	O	0.67956700	-3.54786700	0.78011500
	C	-2.80777700	2.02130500	-0.40105600
	H	-3.46619500	1.19440200	-0.16119300
	C	-3.30464500	3.29048000	-0.66457300
	H	-4.37859400	3.44798900	-0.68700100
	C	-2.41375400	4.33227900	-0.88084500
	H	-2.77610300	5.33352000	-1.10037600
	C	-1.05255200	4.07983300	-0.77831800
	H	-0.29380200	4.84949700	-0.88622000
	C	-0.63575400	2.79222300	-0.49687100
	C	0.80975600	2.47677800	-0.23092200
	C	0.12357300	-2.51553900	0.42302000
	C	-1.27631000	-2.53061600	-0.12148000
	C	-1.96917700	-3.71374000	-0.30107500
	H	-1.50065700	-4.63372800	0.03633600
	C	-3.20952400	-3.67294600	-0.92326100
	H	-3.78113100	-4.58456400	-1.07912700
	C	-3.69501900	-2.45138900	-1.36748500
	H	-4.64446200	-2.37298000	-1.88764800
	C	-2.94301000	-1.30763300	-1.14050500
	H	-3.27195200	-0.33925900	-1.49728200
	C	2.08552500	0.53520800	0.52775000
	C	1.90461100	-0.86039600	0.66763900
	C	2.97813900	-1.66995200	1.02999800
	C	4.23017500	-1.09552700	1.24370900
	C	4.38048800	0.27132000	1.09564700
	C	3.33550300	1.11008600	0.74077400
	H	2.82499700	-2.73745300	1.14175500

	H	5.08537300	-1.70167600	1.52794900
	H	3.48729900	2.17625800	0.62163900
	C	-2.55129100	-0.50701200	2.42356900
	O	-3.47701000	0.10009000	1.91453500
	H	-2.52333600	-1.61385000	2.45493600
	N	-1.46284400	0.04063200	3.00051500
	C	-1.29899500	1.47401000	3.06024600
	H	-1.15856600	1.79604600	4.09981400
	H	-0.42420900	1.77838900	2.47113900
	H	-2.19571600	1.94406600	2.65205900
	C	-0.38896900	-0.76557500	3.53063900
	H	0.55537800	-0.52339200	3.02872900
	H	-0.59786200	-1.82622100	3.36256100
	H	-0.27265100	-0.59421600	4.60890900
	O	-0.25979600	0.16949000	-2.94194600
	C	0.94413100	0.34385600	-2.84294800
	N	1.87530600	-0.62077600	-2.71284200
	C	1.50018300	-2.01415200	-2.70462800
	H	0.41104000	-2.07949900	-2.74214200
	H	1.86686200	-2.50208700	-1.79355700
	H	1.92450600	-2.53022900	-3.57643700
	C	3.27758400	-0.31191100	-2.56947300
	H	3.66870700	-0.73981000	-1.63806400
	H	3.41584800	0.77219000	-2.52763800
	H	3.85357200	-0.71276500	-3.41470700
	H	1.39183800	1.35620000	-2.84339700
	F	5.59300900	0.81705700	1.30090000

Table S36. Cartesian coordinates of the $[Ni(bpb\text{-}CF_3)(DMF)_2]^n$ complexes, n=2+, 1+, 0.

Complexes	*Cartesian coordinates*			
$[Ni(bpb\text{-}CF_3)(DMF)_2]^{2+}$	Ni	1.02013900	0.01237200	0.07967000
	N	1.74041600	-0.11793200	1.90099700
	N	-0.63576100	-0.00743700	0.94962900
	N	-0.03426300	-0.03311400	-1.46445500
	N	2.50223500	0.19699200	-1.19269900
	O	-1.56196400	0.44196800	3.01539800
	O	0.13601900	-0.49752500	-3.71842500
	C	2.98240500	-0.39713700	2.28718700
	H	3.69325500	-0.65434200	1.50944900
	C	3.34557400	-0.39389400	3.62798300
	H	4.36983600	-0.62465900	3.90490400
	C	2.38613600	-0.10160200	4.58830100
	H	2.64762900	-0.08429700	5.64297000
	C	1.07847600	0.13794300	4.18126900

	H	0.27039700	0.32814400	4.88275500
	C	0.79470900	0.10844900	2.82916200
	C	-0.60374100	0.22556400	2.32252300
	C	0.64672000	-0.24752800	-2.65896400
	C	2.11304100	-0.06157900	-2.45306600
	C	2.99852100	-0.06289600	-3.51390000
	H	2.62299000	-0.28173800	-4.51004300
	C	4.33202600	0.24031000	-3.26324900
	H	5.05785300	0.24675900	-4.07203900
	C	4.71519200	0.56498900	-1.96863500
	H	5.73810600	0.84514300	-1.73581800
	C	3.76629400	0.53657000	-0.95461800
	H	4.01710700	0.81694100	0.06264700
	C	-1.72286300	-0.01546000	0.14157000
	C	-1.37334900	-0.08736600	-1.26058000
	C	-2.38441800	-0.17787300	-2.23755600
	C	-3.69014400	-0.16259300	-1.82019200
	C	-4.03014700	-0.06014400	-0.44143500
	C	-3.07533900	0.01419200	0.53808800
	H	-2.12544300	-0.25375600	-3.28646300
	H	-4.48914900	-0.22642300	-2.55466000
	H	-3.34121500	0.09003800	1.58544600
	C	0.60352200	2.96968400	-0.37808100
	C	-5.49957000	-0.03916300	-0.08604400
	F	-5.68860200	0.04696600	1.22621600
	F	-6.08664200	-1.15139600	-0.52968800
	F	-6.09018900	1.00286200	-0.67189200
	O	1.07242400	2.09453400	0.37914700
	H	0.18881800	2.70083300	-1.35955600
	N	0.55706600	4.25440700	-0.09305500
	C	1.05603800	4.77124200	1.17126000
	H	1.86125600	5.48920500	0.98288700
	H	0.24595800	5.28033100	1.70397500
	H	1.43293400	3.94689800	1.77647700
	C	0.00444400	5.22514100	-1.02455800
	H	-0.84105500	5.74461300	-0.56164300
	H	-0.34014100	4.72571400	-1.93325800
	H	0.76918500	5.96174100	-1.29213400
	O	1.30529400	-2.06169200	-0.12962600
	C	0.57152900	-2.96550200	0.32184000
	N	0.73204200	-4.24887700	0.07462700
	C	1.80651400	-4.73014300	-0.77881700
	H	2.38806600	-3.88294800	-1.14208100
	H	1.38286700	-5.27815700	-1.62698000
	H	2.45391100	-5.40550000	-0.20950100
	C	-0.15485000	-5.25353800	0.63996600

	H	-0.64304000	-5.81321800	-0.16466100
	H	-0.92103100	-4.77913800	1.25788200
	H	0.41907500	-5.95106600	1.25889000
	H	-0.27981000	-2.72597800	0.97409600
$[Ni(bpb\text{-}CF_3)(DMF)_2]^{1+}$	Ni	1.10942400	0.05528600	0.07524700
	N	1.83980000	0.47940800	1.86098200
$[Ni^{3+}]$	N	-0.53744600	0.23293600	0.93309500
	N	0.06690600	-0.43359100	-1.39365400
	N	2.60477100	-0.11189700	-1.20365400
	O	-1.49446400	1.14878300	2.81707700
	O	0.23153700	-1.42700000	-3.46568600
	C	3.08708100	0.37937900	2.31369700
	H	3.82272600	-0.03643800	1.63422300
	C	3.42924100	0.75438900	3.60423700
	H	4.45867900	0.65928900	3.93555200
	C	2.43742100	1.23667400	4.44940400
	H	2.67895400	1.54442000	5.46334900
	C	1.12972100	1.29045800	3.98801800
	H	0.29496100	1.61419700	4.60347500
	C	0.86926900	0.88896600	2.69054900
	C	-0.53104500	0.79792800	2.16505100
	C	0.72184900	-0.91540200	-2.47873700
	C	2.19561100	-0.67754200	-2.34832400
	C	3.06570600	-0.94731000	-3.38882800
	H	2.66596700	-1.41079300	-4.28645600
	C	4.39951200	-0.59098000	-3.24803700
	H	5.11145000	-0.78793200	-4.04520600
	C	4.80493900	0.04927900	-2.08349300
	H	5.82990900	0.37967100	-1.94626300
	C	3.87179600	0.27617200	-1.08299900
	H	4.13748900	0.80499700	-0.17444100
	C	-1.65663800	-0.03432100	0.13455400
	C	-1.31334500	-0.44449200	-1.16891500
	C	-2.30560200	-0.78917200	-2.08343800
	C	-3.63673500	-0.70715100	-1.69046900
	C	-3.97110800	-0.29184000	-0.40775300
	C	-2.98910900	0.04836000	0.52007300
	H	-2.03287100	-1.11690500	-3.07923600
	H	-4.41950600	-0.97624300	-2.39325600
	H	-3.24755900	0.36600400	1.52331300
	C	0.18882000	2.72004700	-0.81049700
	C	-5.41066700	-0.14231100	-0.01100100
	F	-5.61111600	-0.49992100	1.26482400
	F	-6.22004200	-0.88598000	-0.77413800
	F	-5.81632400	1.13290500	-0.12752400
	O	1.18491100	2.15034600	-0.33663600

	H	-0.71578800	2.15568300	-1.07333000
	N	0.10705200	4.02187600	-1.04071600
	C	1.21158300	4.91199900	-0.74266500
	H	1.53566500	5.42293500	-1.65633800
	H	0.89639900	5.66375500	-0.01024600
	H	2.03890700	4.32983700	-0.33570500
	C	-1.09647300	4.61829100	-1.58705300
	H	-1.49986700	5.35964700	-0.88818300
	H	-1.85324100	3.84834300	-1.75694200
	H	-0.87424500	5.11328800	-2.53913700
	O	1.45260600	-1.99198500	0.58071500
	C	0.48952400	-2.76966300	0.67348200
	N	0.59312100	-4.06618800	0.92463900
	C	1.88430200	-4.69781800	1.11201000
	H	2.66727800	-3.94573900	1.01041000
	H	2.02792100	-5.48112500	0.35922400
	H	1.93672000	-5.15128900	2.10828000
	C	-0.57919600	-4.91482300	1.01291100
	H	-0.53049500	-5.70449700	0.25471000
	H	-1.48358200	-4.32396200	0.84749300
	H	-0.63604500	-5.37883300	2.00404500
	H	-0.54124300	-2.41302200	0.54605400
$[Ni(bpb\text{-}CF_3)(DMF)_2]^{1+}$	Ni	-1.15071900	0.00835700	0.11621900
	N	-1.78207200	0.06236800	2.07543200
$[Ni^{2+}]$	N	0.60889900	0.03259700	0.98025800
	N	-0.02537600	-0.05532600	-1.48535000
	N	-2.64889300	-0.01649500	-1.29942900
	O	1.62726500	0.13465100	3.06434900
	O	-0.13483900	-0.16882200	-3.80197900
	C	-3.01299000	0.05992800	2.57794000
	H	-3.82687100	0.01763000	1.86177500
	C	-3.26969000	0.10512900	3.94129100
	H	-4.29521900	0.10113500	4.29793800
	C	-2.19749000	0.15418200	4.82076700
	H	-2.36186300	0.19100400	5.89440300
	C	-0.91015100	0.15306700	4.30316000
	H	-0.02624600	0.18671100	4.93304100
	C	-0.74310200	0.10545400	2.92792700
	C	0.64609000	0.09429400	2.35176100
	C	-0.65109700	-0.10854400	-2.70513900
	C	-2.14690700	-0.08062300	-2.54510100
	C	-2.95386600	-0.11510500	-3.67161300
	H	-2.48088600	-0.16729900	-4.64767100
	C	-4.33136000	-0.07995500	-3.50907600
	H	-4.99103100	-0.10565100	-4.37236700
	C	-4.84945600	-0.00913800	-2.22373800

	H	-5.91998800	0.02343400	-2.04535600
	C	-3.96966500	0.02066100	-1.15061900
	H	-4.33962100	0.07911800	-0.13234000
	C	1.66408000	-0.00030100	0.14865100
	C	1.30107500	-0.05468800	-1.26563000
	C	2.32768700	-0.09822200	-2.24030900
	C	3.63626400	-0.08611400	-1.84019800
	C	3.98421100	-0.03113400	-0.46509300
	C	3.03190600	0.01135600	0.51670800
	H	2.05743700	-0.14022800	-3.28849000
	H	4.42791900	-0.11888700	-2.58432600
	H	3.29951300	0.05336700	1.56563700
	C	-0.45177700	-2.98676600	0.02629500
	C	5.44722200	-0.02095100	-0.11716500
	F	5.65649500	0.03073800	1.19770800
	F	6.05581500	1.03460800	-0.67454500
	F	6.05051200	-1.12033300	-0.58901300
	O	-1.38661600	-2.21477000	0.24092900
	H	0.54866800	-2.61340200	-0.24786900
	N	-0.52331000	-4.31553200	0.09413300
	C	-1.76321100	-4.97892800	0.44109800
	H	-2.07632700	-5.64089300	-0.37471100
	H	-1.62740300	-5.57832500	1.34887400
	H	-2.53122500	-4.22396200	0.61363600
	C	0.62508300	-5.15466400	-0.17357300
	H	0.86783800	-5.76151000	0.70667100
	H	1.49187700	-4.53620400	-0.42175800
	H	0.41969500	-5.82511400	-1.01639500
	O	-1.35306700	2.23809900	0.12840900
	C	-0.41663800	2.98298300	-0.16188200
	N	-0.46563100	4.31446100	-0.15656400
	C	-1.67994000	5.01465900	0.20817700
	H	-2.45105600	4.28197200	0.44930100
	H	-2.01729200	5.64143000	-0.62557100
	H	-1.49718900	5.65502000	1.07898500
	C	0.68318300	5.12020500	-0.51103000
	H	0.45333900	5.75276200	-1.37652400
	H	1.52970600	4.47594300	-0.76350300
	H	0.97098200	5.76443400	0.32810800
	H	0.56594800	2.57993100	-0.45767300
$[Ni(bpb\text{-}CF_3)(DMF)_2]^0$	Ni	-1.39625200	-0.16907200	0.01444000
	N	-2.08385500	-2.01143900	0.37655800
	N	0.28841300	-0.99697000	0.27074300
	N	-0.34219200	1.34598500	-0.42208200
	N	-2.90612400	1.12487600	-0.19027200
	O	1.28386900	-2.97476900	0.96594300

	O	-0.49079700	3.54808800	-1.14333400
	C	-3.31242700	-2.51281400	0.28325800
	H	-4.08579200	-1.83302100	-0.05739700
	C	-3.60605700	-3.83743700	0.57167800
	H	-4.62701400	-4.19539400	0.47951500
	C	-2.57418900	-4.67776400	0.96708900
	H	-2.77021500	-5.72014700	1.20606000
	C	-1.28608900	-4.16798300	1.03112900
	H	-0.42184900	-4.76763500	1.30124100
	C	-1.07675600	-2.83456900	0.71917800
	C	0.32049200	-2.27057600	0.68128900
	C	-0.96484100	2.47836100	-0.77613200
	C	-2.45555500	2.31727000	-0.61947200
	C	-3.30366300	3.38848800	-0.84820100
	H	-2.86430600	4.31843600	-1.19682300
	C	-4.66044300	3.23374300	-0.60650200
	H	-5.35104700	4.05605900	-0.77664000
	C	-5.11785800	2.01546300	-0.12312600
	H	-6.16635800	1.85290900	0.10751400
	C	-4.20323500	0.99119200	0.07274600
	H	-4.51680900	0.03493400	0.47692400
	C	1.38211900	-0.17381000	0.04951900
	C	1.02604800	1.14246700	-0.35930400
	C	2.02384600	2.07520300	-0.63978000
	C	3.36253900	1.71655200	-0.50325300
	C	3.70496200	0.43454300	-0.09500100
	C	2.72233200	-0.51859800	0.18323100
	H	1.73985300	3.06978300	-0.96465700
	H	4.14010700	2.44281400	-0.72106400
	H	2.98392700	-1.52151000	0.50285100
	C	-0.57082900	-0.36284600	-2.92154500
	C	5.13444700	0.06397800	0.12523700
	F	5.41321300	-1.17098900	-0.32610000
	F	5.45618300	0.06933100	1.43526300
	F	5.98414400	0.90871400	-0.48158800
	O	-1.59746300	-0.86331800	-2.48204200
	H	-0.53430900	0.68825500	-3.26189900
	N	0.61677500	-0.97913400	-3.04165200
	C	0.78092000	-2.35940000	-2.64987700
	H	1.13392200	-2.95326400	-3.50281800
	H	1.50970600	-2.44505300	-1.83446100
	H	-0.18505100	-2.74186300	-2.31529200
	C	1.79879800	-0.27204800	-3.47371800
	H	2.55089400	-0.26324900	-2.67455000
	H	1.54420500	0.76416400	-3.71301900
	H	2.22974900	-0.74775400	-4.36424500

	O	-1.58845600	0.39822500	2.55261100
	C	-0.40163800	0.41154300	2.85094100
	N	0.39719100	1.49183000	2.85349200
	C	-0.11632800	2.79464400	2.50120700
	H	-1.18544700	2.70324100	2.30128200
	H	0.38983600	3.18041400	1.60773400
	H	0.04217800	3.49805800	3.32885400
	C	1.81272800	1.38953400	3.11940500
	H	2.38950800	1.70793900	2.24187900
	H	2.07590100	0.35121100	3.33918100
	H	2.09145300	2.01633900	3.97646700
	H	0.13553000	-0.50780000	3.14796300

Table S37. Cartesian coordinates of the $[Ni(bpb\text{-}NO_2)(DMF)_2]^n$ complexes, n=2+, 1+, 0.

Complexes	*Cartesian coordinates*			
$[Ni(bpb\text{-}NO_2)(DMF)_2]^{2+}$	Ni	0.81050400	0.00960700	0.08052900
	N	1.51195100	-0.18913100	1.90371600
	N	-0.85361300	-0.04474900	0.93302900
	N	-0.22944700	0.02227300	-1.47478600
	N	2.30375200	0.24400100	-1.16906400
	O	-1.80156600	0.33623000	3.00272900
	O	-0.03615400	-0.35958400	-3.74231100
	C	2.75034300	-0.48163000	2.29194100
	H	3.46892800	-0.71111900	1.51257000
	C	3.10050200	-0.52557300	3.63559500
	H	4.12215900	-0.76596900	3.91402600
	C	2.13197500	-0.26692300	4.59645500
	H	2.38337400	-0.28663200	5.65354600
	C	0.82814900	-0.01270300	4.18564800
	H	0.01355100	0.15324500	4.88584900
	C	0.55745700	0.00539500	2.83065000
	C	-0.83577900	0.14180700	2.31541600
	C	0.46440500	-0.14817300	-2.67026000
	C	1.92813900	0.03122000	-2.44225100
	C	2.82483100	0.06893000	-3.49293300
	H	2.46026600	-0.11360900	-4.50044100
	C	4.15548400	0.36312700	-3.21713000
	H	4.88988700	0.39918000	-4.01737200
	C	4.52467100	0.64029600	-1.90748100
	H	5.54494800	0.91178900	-1.65369200
	C	3.56516300	0.57460900	-0.90516600
	H	3.80514400	0.81730300	0.12435000
	C	-1.93116700	-0.02501200	0.11530100
	C	-1.56924000	-0.04275500	-1.28725900

	C	-2.57086800	-0.09846800	-2.27652000
	C	-3.88349900	-0.10114700	-1.87626300
	C	-4.21395400	-0.05450100	-0.50069000
	C	-3.28618000	-0.01550300	0.50199200
	H	-2.30225700	-0.13419400	-3.32510900
	H	-4.69115500	-0.13462300	-2.60194700
	H	-3.58762000	0.01562100	1.54261000
	C	0.43756300	2.98543100	-0.28982300
	O	0.84397200	2.07648800	0.46447400
	H	0.10597300	2.76073500	-1.31307600
	N	0.36565600	4.25604200	0.04667000
	C	0.75711700	4.71609200	1.36968200
	H	1.57232300	5.44178100	1.27986600
	H	-0.09592600	5.20097500	1.85583700
	H	1.08627700	3.86590400	1.96687900
	C	-0.10885300	5.26838500	-0.88400600
	H	-0.98988100	5.76956100	-0.46981700
	H	-0.37695200	4.81039700	-1.83912700
	H	0.67497900	6.01364400	-1.05468700
	O	1.08966800	-2.05314800	-0.21477300
	C	0.40281200	-2.97823900	0.26745800
	N	0.54641200	-4.25054300	-0.03841000
	C	1.54268900	-4.69381300	-1.00071500
	H	2.08754500	-3.83064100	-1.38269600
	H	1.04719100	-5.21343300	-1.82751800
	H	2.23971200	-5.38533900	-0.51571100
	C	-0.28169800	-5.28145900	0.56803500
	H	-0.83627900	-5.81715600	-0.20939200
	H	-0.99253500	-4.83512900	1.26760900
	H	0.34875500	-5.99499900	1.10870200
	H	-0.38812700	-2.76836100	1.00093700
	N	-5.65150700	-0.05025000	-0.12376100
	O	-5.90116300	-0.06163900	1.05673100
	O	-6.44191000	-0.03432200	-1.03745100
$[Ni(bpb\text{-}NO_2)(DMF)_2]^{1+}$	Ni	-0.90522600	-0.04853300	0.06976200
	N	-1.65227500	-0.50611000	1.83990600
$[Ni^{3+}]$	N	0.73262500	-0.26796600	0.93325500
	N	0.15580700	0.45966200	-1.38037200
	N	-2.38862600	0.16931500	-1.21500700
	O	1.66616600	-1.25031000	2.79455800
	O	0.01743600	1.50645500	-3.42828100
	C	-2.90097700	-0.39933000	2.28716000
	H	-3.62585100	0.04548200	1.61450000
	C	-3.25692800	-0.80316100	3.56528100
	H	-4.28692400	-0.70109600	3.89277400
	C	-2.27810400	-1.32266100	4.40347100

	H	-2.53086000	-1.65336100	5.40739000
	C	-0.96821300	-1.38415100	3.94893500
	H	-0.14256800	-1.73680600	4.56086700
	C	-0.69356000	-0.95236900	2.66434900
	C	0.71119000	-0.86822700	2.14964000
	C	-0.48686900	0.97750200	-2.45882600
	C	-1.96432100	0.75607500	-2.34347400
	C	-2.82339900	1.06232400	-3.38287200
	H	-2.41192100	1.54143800	-4.26689000
	C	-4.16301300	0.72127100	-3.25895500
	H	-4.86679700	0.94676700	-4.05577500
	C	-4.58480100	0.05972600	-2.11240000
	H	-5.61511900	-0.25948300	-1.98947400
	C	-3.66162800	-0.20354100	-1.11139300
	H	-3.94074100	-0.74948600	-0.21711600
	C	1.86039000	0.00422100	0.15004200
	C	1.53110000	0.45287700	-1.14644100
	C	2.53352600	0.81076600	-2.04826100
	C	3.86053800	0.70585800	-1.65417500
	C	4.16142300	0.25081900	-0.37963700
	C	3.18720900	-0.10656200	0.54406100
	H	2.26989600	1.16387200	-3.03766100
	H	4.66667000	0.97276200	-2.32948000
	H	3.45989900	-0.45833500	1.53132700
	C	-0.02295700	-2.71127000	-0.87097700
	O	-1.00928100	-2.13119500	-0.38901800
	H	0.89267300	-2.15918600	-1.12172200
	N	0.03486000	-4.00973000	-1.12429200
	C	-1.08682000	-4.88425600	-0.84349900
	H	-1.41890800	-5.37237100	-1.76667200
	H	-0.78650800	-5.65479800	-0.12454600
	H	-1.90372100	-4.29429600	-0.42691500
	C	1.22771700	-4.61962700	-1.67946900
	H	1.61673700	-5.37927700	-0.99226100
	H	1.99865800	-3.86126300	-1.83711800
	H	0.99689300	-5.09512500	-2.63934700
	O	-1.22609000	1.99043500	0.61470800
	C	-0.25622900	2.75042900	0.76732400
	N	-0.35092200	4.04261000	1.04122200
	C	-1.63953600	4.69146200	1.18375600
	H	-2.42884700	3.95516200	1.02951400
	H	-1.73571400	5.49333200	0.44312600
	H	-1.73035300	5.12382800	2.18658200
	C	0.82898200	4.87031300	1.20147400
	H	0.82703700	5.67633800	0.45916300
	H	1.73114700	4.26880300	1.06500000

	H	0.84690800	5.31233200	2.20399700
	H	0.77349200	2.37915600	0.67891200
	N	5.57252200	0.14039800	0.02200100
	O	6.40915800	0.45786200	-0.79667400
	O	5.80197300	-0.25995400	1.14356000
$[Ni(bpb\text{-}NO_2)(DMF)_2]^{1+}$	Ni	-1.02638200	0.24443900	-0.02758400
	N	-1.79723600	-1.23375700	-1.01164600
$[Ni^{2+}]$	N	0.50954800	-0.77362300	-0.06124300
	N	0.08646300	1.54617600	0.66629300
	N	-2.43486700	1.48931100	0.38280600
	O	1.22115300	-2.95962600	-0.36185300
	O	0.13153800	3.84853700	0.94681400
	C	-2.96380100	-1.30721000	-1.64741900
	H	-3.56614300	-0.40534800	-1.65883500
	C	-3.38206200	-2.46646500	-2.28709000
	H	-4.34125000	-2.48092500	-2.79529600
	C	-2.55817400	-3.58365300	-2.26872300
	H	-2.86548700	-4.50513200	-2.75588100
	C	-1.32189400	-3.49451100	-1.64091800
	H	-0.61066700	-4.31540800	-1.62168000
	C	-0.97412700	-2.30069000	-1.03721300
	C	0.38284800	-2.08998900	-0.44501000
	C	-0.46567800	2.81400300	0.77170100
	C	-1.95198000	2.71439700	0.66234600
	C	-2.77259600	3.79252600	0.93180800
	H	-2.31890200	4.75615400	1.14415900
	C	-4.14683000	3.58829000	0.94916700
	H	-4.82438600	4.41187300	1.15728800
	C	-4.63692900	2.30768400	0.73495500
	H	-5.70000400	2.09445800	0.78860300
	C	-3.74616000	1.27823900	0.46079900
	H	-4.07314700	0.25083100	0.34663500
	C	1.64091700	-0.13845200	0.31848400
	C	1.39632500	1.22925900	0.71694600
	C	2.47672400	2.05981900	1.08604400
	C	3.74627800	1.54087700	1.06646200
	C	3.95867200	0.19984100	0.68290300
	C	2.94934400	-0.65231800	0.31484300
	H	2.29122800	3.08675400	1.37671700
	H	4.60435400	2.14396000	1.34569700
	H	3.15478700	-1.67943800	0.03668600
	C	0.52922500	1.96520300	-2.32798800
	O	-0.61816700	1.55643800	-2.20655100
	H	0.80483300	3.00523000	-2.07495000
	N	1.58171200	1.23457800	-2.73581300
	C	1.40823100	-0.12787100	-3.18662000

	H	1.68093900	-0.21379400	-4.24552300
	H	2.04682900	-0.80717700	-2.60837500
	H	0.36003200	-0.40562700	-3.06310600
	C	2.90928000	1.79933000	-2.83270100
	H	3.61243100	1.22053400	-2.22071800
	H	2.90274200	2.83118600	-2.47141300
	H	3.26127300	1.79104300	-3.87130900
	O	-2.51820100	-1.17808800	1.83256700
	C	-2.10483300	-2.32205600	1.94371200
	N	-0.89956900	-2.68147900	2.42255300
	C	-0.00433500	-1.70044800	2.99534600
	H	-0.38734300	-0.70104500	2.77972400
	H	0.99897900	-1.81538000	2.56876000
	H	0.06188400	-1.83441800	4.08231300
	C	-0.48759700	-4.06614600	2.49392500
	H	0.43565200	-4.21402400	1.91985600
	H	-1.26736000	-4.70780000	2.07374200
	H	-0.31199600	-4.36614900	3.53407900
	H	-2.70665800	-3.19230700	1.61996300
	N	5.34412000	-0.31423000	0.67104800
	O	6.21850200	0.46662800	0.97573000
	O	5.50014900	-1.47127900	0.35237700
$[Ni(bpb\text{-}NO_2)(DMF)_2]^0$	Ni	-0.89172400	0.07206200	-0.09586400
	N	-1.66913200	-1.61271800	-0.64739200
	N	0.65777100	-0.91183200	0.08153600
	N	0.19654100	1.52338800	0.26025600
	N	-2.32536000	1.37614800	-0.00692800
	O	1.46318600	-3.08710000	0.09750800
	O	0.22320300	3.83434700	0.03767400
	C	-2.85903200	-1.86522300	-1.18628400
	H	-3.50547600	-1.01201600	-1.35160000
	C	-3.25293700	-3.14601600	-1.54622500
	H	-4.23666600	-3.30140200	-1.97793900
	C	-2.37431400	-4.20072100	-1.34519000
	H	-2.65923300	-5.21592400	-1.60968700
	C	-1.11509800	-3.93223300	-0.82503200
	H	-0.35608700	-4.69489300	-0.67652900
	C	-0.79542100	-2.62710400	-0.50076400
	C	0.58688500	-2.24727700	-0.05121800
	C	-0.33684300	2.75019100	0.07928600
	C	-1.83032500	2.62798000	-0.03521600
	C	-2.64305200	3.74523300	-0.07353100
	H	-2.16883600	4.72177000	-0.10496700
	C	-4.01977600	3.56870000	-0.04161200
	H	-4.68838900	4.42544500	-0.06985600
	C	-4.52574200	2.28086500	0.06195500

	H	-5.59258700	2.09477100	0.13897200
	C	-3.64465500	1.20812700	0.08054200
	H	-3.99246100	0.19190700	0.22767100
	C	1.79993500	-0.16791600	0.35381200
	C	1.53832900	1.22452000	0.42862200
	C	2.58011100	2.12490500	0.65316400
	C	3.87333100	1.64496300	0.80889400
	C	4.10787700	0.27868000	0.73975000
	C	3.09216600	-0.64814200	0.51559500
	H	2.36796100	3.18685500	0.69404800
	H	4.70464800	2.31937000	0.98406500
	H	3.30532200	-1.70908200	0.46721600
	C	0.27686600	1.12467400	-2.82415200
	O	-0.83511600	0.62174300	-2.84854900
	H	0.42406000	2.21501000	-2.70279600
	N	1.44630400	0.46388500	-2.92212000
	C	1.47572800	-0.96754400	-3.10571900
	H	1.92444100	-1.21925000	-4.07581800
	H	2.06577800	-1.44300700	-2.31279800
	H	0.45060100	-1.34153800	-3.07463800
	C	2.71529500	1.14670100	-2.84645500
	H	3.31464800	0.74728100	-2.01887200
	H	2.55252500	2.21267200	-2.66405900
	H	3.27888500	1.02725700	-3.78139700
	O	-3.49669500	-1.18925000	2.02415900
	C	-2.41581200	-1.63222100	2.37102900
	N	-1.42896200	-0.93861500	2.97245000
	C	-1.59240800	0.46269000	3.28231000
	H	-2.61009000	0.75993400	3.02238600
	H	-0.87763200	1.06494800	2.70655700
	H	-1.42345500	0.63484300	4.35268100
	C	-0.15861200	-1.54070200	3.30111700
	H	0.65901700	-1.00023700	2.80941000
	H	-0.13400500	-2.57828700	2.95501800
	H	0.00916700	-1.52627400	4.38597100
	H	-2.13829500	-2.69226000	2.20917600
	N	5.47588500	-0.21214800	0.90225200
	O	6.35357300	0.61010200	1.08952200
	O	5.65786800	-1.41248600	0.83762700