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Preparation of Compounds. The ligand H_2(TC-3,5) was prepared following a procedure previously described.[1] All operations, excluding ligand synthesis, were carried out under a pure dinitrogen or argon atmosphere by using standard Schlenk and glove-box techniques. Solvents were dried according to standard procedures and were degassed prior to use. Unless otherwise specified, all reagents were obtained from commercial suppliers and were thoroughly degassed and dried before use. NMR spectra were recorded on a Bruker AM-250 spectrometer at room temperature. Compounds **1**, **2**, **4**, and **5** are unstable in solution at room temperature and insoluble at low temperature, prohibiting the collection of ^{13}C NMR data.

Synthesis of 2. A 100 mL flask containing a slurry of 130 mg (0.184 mmol) of $[Hf(CH_2Ph)_2(TC\text{-}3,5)]$[2] in 4 mL of toluene was charged with carbon monoxide at room temperature. The slurry rapidly darkened and red-brown crystals deposited directly from the reaction mixture. The product was collected by filtration, washed with ether, and dried to afford 87 mg (64%) of micro-crystalline material. ^1H NMR (CD_2Cl_2): δ 7.18 (m, 4H), 7.01 (d, 4H), 6.78 (t, 8H), 4.66 (t, 2H), 3.86 (d, 2H), 3.52 (m, 4H), 3.36 (m, 4H), 2.93 (m, 2H), 2.15 (m, 2H), 1.25 (m, 2H), 1.00 (m, 2H). IR KBr (cm^{-1}) 2924 (m), 1591 (vs), 1508 (s), 1437 (m), 1421 (m), 1395 (m), 1388 (w), 1339 (w), 1320 (w), 1275v (s), 1223 (m), 1029 (m), 980 (w), 935 (w), 887 (w), 736 (s), 700 (s), 584 (m), 473 (m). $[Hf(\eta^2\text{-}OC(CH_2Ph)_2)(TC\text{-}3,5)]\cdot C_6H_5Cl$: Anal. Calcd. for $C_{43}H_{45}N_4ClOHf$: C, 60.92; H, 5.35; N, 6.61. Found: C, 60.81; H, 5.38; N, 6.65.

Synthesis of 3. A solution of 20 mg (0.027 mmol) of **2** in 1 mL of dichloromethane was stored at room temperature overnight. Small yellow crystals deposited directly from the mixture and were characterized by x-ray crystallography. $[HfCl(OCH(CH_2Ph)_2)(TC\text{-}3,5)]$: ^1H NMR (CD_2Cl_2): δ 7.15 (br, m), 6.99 (br, m), 6.78 (br, m), 6.63 (d, 11.2 Hz), 6.46 (t, 10.3Hz), 4.42 (m), 3.65 (m), 3.28 (br, s), 2.87 (m), 2.64 (m), 2.11 (br, m), 1.25 (br, m) 0.57 (br, m).

Synthesis of 4. A solution of 75 mg (0.102 mmol) of **2** in 2 mL of dichloromethane was treated with 25.4 μL (0.204 mmol) cyclohexylisocyanide at -30°C. The dark red-brown solution was filtered, and several volume equivalents of pentane were diffused into the solution at -30°C resulting in the formation of large rhombohedral crystals. The crystals were collected, washed with ether and dried to afford 36 mg (37%) of product.. ^{1}H NMR (CD_2Cl_2): δ 7.20 (m), 7.14 (m), 7.01 (m), 6.75 (d, 11.7Hz), 6.54 (m) 6.46 (d of t, 3.2Hz, 9.1Hz), 4.20 (d of t, 3.1Hz, 14.0Hz), 3.63 (s, br), 3.57 (s, br), 3.43 (m), 3.26 (m), 3.03 (br, s), 2.98 (br, s), 2.89 (br, s), 2.84 (br, s), 2.75 (br, s), 2.70 (br, s), 2.49 (d, 12.9Hz), 2.03 (br, s), 1.97 (br, s), 1.71 (br, m), 1.40 (br, m), 1.26 (br, m), 1.02 (br, m). IR KBr (cm^{-1}) 3056 (w), 3023 (w), 2923 (s), 2850 (m), 1997 (w), 1950 (m), 1591 (s), 1509 (s), 1472 (m), 1444 (m), 1425 (m), 1388 (m), 1368 (w), 1340 (m), 1322 (w), 1275 (m), 1229 (m), 1135 (m), 1111 (w), 1086 (m), 1061 (w), 1025 (m), 992 (w), 977 (w), 967 (m), 937 (m), 878 (m), 861 (w), 839 (w), 821 (m), 793 (w), 760 (w), 748 (m), 722 (s), 698 (s), 669 (w), 614 (m), 600 (w), 562 (m), 545 (w), 515 (w), 477 (w), 468 (w). Anal. Calcd. for $C_{51}H_{62}N_6OHf$: C, 64.24; H, 6.55; N, 8.81. Found: C, 63.73; H, 6.57; N, 8.63.

Synthesis of 5. Dropwise addition of 24.8 μL (0.204 mmol) of benzylisocyanide to a stirred solution of 75 mg (0.102 mmol) of **2** in 2 mL of dichloromethane resulted in a slow color change from deep red to bright yellow-orange. The reaction mixture was stirred for 10 min, and filtered through celite, and the filtrate was cooled to -30°C. Over the course of several days, ether was slowly diffused into the reaction mixture resulting in the formation of yellow microcrystals. The material was collected, washed with pentane, and dried to afford 31 mg (31%) of product. ^{1}H NMR (CD_2Cl_2): δ 7.58 (br, m), 7.41 (br, m), 7.31 (br, m), 7.14 (br, m), 6.88 (br, s) 6.68 (m), 6.59 (br, s), 6.48 (m), 6.12 (t, 9Hz), 5.44 (d, 11.1Hz), 5.09 (br, m), 2.91 (d, 14.3Hz), 2.64 (12.9Hz), 2.28 (br, m), 1.76 (br, m), 1.26 (br, m), 0.72 (br, m). IR KBr (cm^{-1}) 3204 (w), 3003 (w), 2921 (m), 2902 (w), 2067 (m), 1954 (w), 1623 (m), 1589 (s), 1506 (vs), 1470 (m), 1444 (m), 1423 (m), 1388 (m), 1374 (w), 1337 (m), 1317 (m), 1267 (m), 1226 (m), 1167 (w), 1111 (m), 1079 (m), 1061 (w), 1025 (s),

976 (m), 942 (w), 933 (m), 889 (m), 839 (w), 826 (m), 817 (m), 763 (w), 751 (w), 730 (vs), 704 (s), 634 (s), 615 (m), 536 (m), 484 (m), 472 (w), 456 (w), 433 (w).

Sytheses of 6 and 7. A solution of 30 mg (0.0408 mmol) of **2** in 1.5 mL of dichloromethane was treated with 2 equiv (0.0816 mmol) of cyclohexenone or *trans*-4-phenyl-3-buten-2-one at -30°C. The solution rapidly turned from red-brown to bright yellow and the mixture was allowed to warm to room temperature. The mixture was filtered and several volume equiv of ether were diffused into the filtrate at -30°C over the course of several days. The resulting yellow crystals were collected, washed with ether, and dried to afford of **6** (16 mg, 47%) or **7** (14 mg, 39%). **6**: ^{1}H NMR (CD_2Cl_2): δ 7.23 (m), 7.14 (m), 6.78 (m), 6.51 (d of t, 2.7Hz, 9.2Hz), 5.85 (d, 10.3 Hz), 5.57 (m), 5.0 (br, s), 3.77 (br, m), 3.61 (br, s), 3.56 (br, s), 3.12 (d, 13.7 Hz), 2.91 (d, 13.5 Hz), 2.65 (br, m), 2.20 (br, m), 1.89 (br, m), 1.64 (br, s), 1.46 (br, m), 1.04 (br, m). ^{13}C NMR (CD_2Cl_2): δ 165.43, 165.08, 164.69, 164.53, 141.41, 141.21, 135.43, 135.35, 134.92, 132.09, 131.90, 130.07, 129.61, 128.79, 128.62, 127.48, 126.04, 125.49, 125.37, 120.56, 114.35, 113.96, 113.73, 88.53, 66.26, 52.76, 48.03, 42.78, 35.00, 28.71, 28.47, 26.25, 20.86, 18.55, 15.73, 14.45. Anal. Calcd. for $C_{43}H_{48}N_4O_2Hf$: C, 62.12; H, 5.82; N, 6.74. Found: C, 61.65; H, 6.05; N, 6.59. **7**: ^{1}H NMR (CD_2Cl_2): δ 7.25 (m,), 7.19 (m), 7.15 (m), 6.75 (m), 6.58 (m), 6.46 (m), 5.39 (br, s), 5.15 (br, s), 3.88 (br, s), 3.83 (br, s), 3.67 (br, m), 3.62 (br, m), 3.05 (d, 5.8Hz), 3.00 (d, 5.9Hz), 2.75 (d, 13.5Hz), 2.57 (d, 13.5Hz), 2.23 (m), 1.94 (br, d, 12.6 Hz), 1.53 (br, s), 1.27 (s), 1.05 (m). ^{13}C NMR (CD_2Cl_2): δ 165.35, 165.16, 164.81, 164.58, 141.41, 141.13, 140.08, 139.15, 135.54, 135.43, 135.35, 131.90, 131.74, 128.98, 128.79, 127.74, 127.27, 127.00, 126.85, 126.62, 125.76, 120.87, 120.75, 114.58, 114.16, 113.97, 92.22, 66.26, 52.68, 48.18, 44.83, 44.49, 28.77, 28.34, 27.29, 26.98, 18.48. Anal. Calcd. for $C_{47}H_{50}N_4O_2Hf$: C, 64.05; H, 5.72; N, 6.36. Found: C, 64.80; H, 6.09; N, 6.04.

Indentification of 8. Slow diffusion of ether into solutions of **4** in wet dichloromethane yielded small yellow needle-like crystals of **8** at -30°C. The complex was characterized by CCA.

Collection and Reduction of X-ray Data. Crystals were obtained by vapor diffusion of ether or pentane into a saturated solutions of the complexes at -30°C. Specific crystallization solvents were reported above. Single crystals were coated with Paratone-N oil, attached to a glass fiber, and rapidly transferred to the cold stream (185K) goniostat of a Siemens CCD X-ray diffraction system controlled by a pentium-based PC running the SMART software package.

The structures were solved by direct methods using SIR-92 or SHELXS and were refined by full matrix least-squares and Fourier techniques using the SHELXTL-PLUS program package. Space groups for all of the structures were determined from an examination of the systematic absences in the data, and these assignments were confirmed by the successful solution and refinement of the structure. Non-hydrogen atoms were usually refined anisotropically. Hydrogen atoms were assigned idealized positions and given a thermal parameter 1.2 times the thermal parameter of the carbon atoms to which they were attached. The structure of **2** contains a chlorobenzene molecule of crystallization situated in a channel and disordered beyond recognition. The identification of this residue was made by a combination of elemental analysis and NMR spectroscopy. Atoms arbitrarily assigned as carbon were introduced to account for electron density in this region and refined isotropically. There is no chemical meaing to the positioning of these atoms. The dichloromethane molecule in **3** is disordered across the mirror plane and was refined with two positions, each having a site occupancy of 0.5. Crystals of **5** grew as aggregates of thin plates. After numerous attempts to find a suitable specimen, data were collected on a crystal with structure in the peak profile plots. The integration window was expanded to 1.75° to assure that the intensity of the entire peak was measured. The poor quality of the data is reflected in the large anisotropic thermal parameters of the atoms and the high residuals. In the structure of **7**, three areas of electron density were modeled as disordered tetrahydrofuran molecules, and these atoms were refined isotropically. Final refinement yielded the residuals given in

Tables SI-SVII. ORTEP diagrams (30% probability ellipsoids) showing atom labeling scheme can be found in Figures S1-SVII.

1. Zask, A.; Gonnella, N.; Nakanishi, K.; Turner, C. J.; Imajo, S.; Nozoe, T. *Inorg. Chem.* **1986**, *25*, 3400-3406.

2. Scott, M. J.; Lippard, S. J., manuscript in preparation.

Table S-I. Crystal data and structure refinement for 2.

Identification code	Hf(TC-3,5)(benzyl)2 + CO
Empirical formula	C43 H45 Cl Hf N4 O
Formula weight	847.77
Temperature	188(2) K
Wavelength	0.71073 A
Crystal system	Orthorhombic
Space group	Cmc2(1)
Unit cell dimensions	a = 17.32120(10) A alpha = 90 deg. b = 20.7671(2) A beta = 90 deg. c = 10.06500(10) A gamma = 90 deg.
Volume, Z	3620.49(5) A^3, 4
Density (calculated)	1.555 Mg/m^3
Absorption coefficient	2.995 mm^-1
F(000)	1712
Crystal size	0.4 x 0.2 x 0.15 mm
Theta range for data collection	1.53 to 28.27 deg.
Limiting indices	-23<=h<=14, -26<=k<=26, -13<=l<=13
Reflections collected	10901
Independent reflections	4131 [R(int) = 0.0314]
Absorption correction	Semi-empirical from psi-scans
Max. and min. transmission	0.8831 and 0.5769
Refinement method	Full-matrix least-squares on F^2
Data / restraints / parameters	4131 / 1 / 208
Goodness-of-fit on F^2	1.185
Final R indices [I>2sigma(I)]	R1 = 0.0294, wR2 = 0.0874
R indices (all data)	R1 = 0.0317, wR2 = 0.0892
Absolute structure parameter	-0.001(14)
Largest diff. peak and hole	2.230 and -0.647 e.A^-3

Table S-II. Crystal data and structure refinement for 3.

Identification code	[Hf(OC(CH2Ph)2)(TC-3,5)] + CH2CL2
Empirical formula	C42 H54 Cl3 Hf N4 O2
Formula weight	931.73
Temperature	188(2) K
Wavelength	0.71073 A
Crystal system	Orthorhombic
Space group	Cmc2(1)
Unit cell dimensions	a = 17.7705(3) A alpha = 90 deg. b = 20.8034(5) A beta = 90 deg. c = 9.7159(2) A gamma = 90 deg.
Volume, Z	3591.84(13) A^3, 4
Density (calculated)	1.723 Mg/m^3
Absorption coefficient	3.173 mm^-1
F(000)	1892
Crystal size	0.4 x 0.15 x 0.15 mm
Theta range for data collection	1.51 to 28.28 deg.
Limiting indices	-23<=h<=22, -27<=k<=13, -12<=l<=12
Reflections collected	11178
Independent reflections	4291 [R(int) = 0.0297]
Absorption correction	Semi-empirical from psi-scans
Max. and min. transmission	0.9349 and 0.6021
Refinement method	Full-matrix least-squares on F^2
Data / restraints / parameters	4291 / 1 / 232
Goodness-of-fit on F^2	1.088
Final R indices [I>2sigma(I)]	R1 = 0.0226, wR2 = 0.0516
R indices (all data)	R1 = 0.0244, wR2 = 0.0527
Absolute structure parameter	-0.013(8)
Largest diff. peak and hole	0.771 and -0.571 e.A^-3

Table S-III. Crystal data and structure refinement for 4.

Identification code	[Hf(OC(CH2Ph)2(TC-3,5)] + 2cync
Empirical formula	C52 H62 Cl2 Hf N6 O
Formula weight	1036.47
Temperature	188(2) K
Wavelength	0.71073 A
Crystal system	Triclinic
Space group	P-1
Unit cell dimensions	a = 11.1134(2) A alpha = 102.99 deg. b = 12.84620(10) A beta = 90.1600(10) deg. c = 16.6664(3) A gamma = 94.2160(10) deg.
Volume, Z	2311.78(6) A^3, 2
Density (calculated)	1.489 Mg/m^3
Absorption coefficient	2.417 mm^-1
F(000)	1060
Crystal size	0.25 x 0.10 x 0.10 mm
Theta range for data collection	1.25 to 23.28 deg.
Limiting indices	-12<=h<=12, -7<=k<=14, -18<=l<=18
Reflections collected	9425
Independent reflections	6485 [R(int) = 0.0378]
Absorption correction	Semi-empirical from psi-scans
Max. and min. transmission	0.6852 and 0.5513
Refinement method	Full-matrix least-squares on F^2
Data / restraints / parameters	6484 / 0 / 559
Goodness-of-fit on F^2	1.081
Final R indices [I>2sigma(I)]	R1 = 0.0378, wR2 = 0.0962
R indices (all data)	R1 = 0.0420, wR2 = 0.1005
Largest diff. peak and hole	1.648 and -1.597 e.A^-3

Table S-IV. Crystal data and structure refinement for 5.

Identification code	[Hf(OC(CH2Ph)2(TC-3,5) + 2 benzylnc
Empirical formula	C54 H54 Cl Hf N6 O
Formula weight	1016.97
Temperature	188(2) K
Wavelength	0.71073 A
Crystal system	Orthorhombic
Space group	Pbca
Unit cell dimensions	a = 21.6337(2) A alpha = 90 deg. b = 10.71340(10) A beta = 90 deg. c = 39.8952(3) A gamma = 90 deg.
Volume, Z	9246.53(14) A^3, 8
Density (calculated)	1.461 Mg/m^3
Absorption coefficient	2.360 mm^-1
F(000)	4136
Crystal size	0.3 x 0.15 x 0.1 mm
Theta range for data collection	2.25 to 23.26 deg.
Limiting indices	-24<=h<=14, -11<=k<=11, -44<=l<=44
Reflections collected	32398
Independent reflections	6608 [R(int) = 0.1139]
Absorption correction	Semi-empirical from psi-scans
Max. and min. transmission	0.9406 and 0.7208
Refinement method	Full-matrix least-squares on F^2
Data / restraints / parameters	6608 / 0 / 562
Goodness-of-fit on F^2	1.101
Final R indices [I>2sigma(I)]	R1 = 0.1061, wR2 = 0.2488
R indices (all data)	R1 = 0.1626, wR2 = 0.2849
Largest diff. peak and hole	2.219 and -2.018 e.A^-3

Table S-V. Crystal data and structure refinement for 6.

Identification code	[Hf(OC(CH2Ph)2(TC-3,5)}+cyclohexenone
Empirical formula	C43 H48 Hf N4 O2
Formula weight	831.34
Temperature	188(2) K
Wavelength	0.71073 A
Crystal system	Monoclinic
Space group	C2/c
Unit cell dimensions	a = 35.9839(3) A alpha = 90 deg. b = 9.3874(2) A beta = 97.622(2) deg. c = 21.3384(4) A gamma = 90 deg.
Volume, Z	7144.3(2) A^3, 8
Density (calculated)	1.546 Mg/m^3
Absorption coefficient	2.964 mm^-1
F(000)	3376
Crystal size	0.2 x 0.1 x 0.15 mm
Theta range for data collection	1.14 to 23.25 deg.
Limiting indices	-33<=h<=39, -10<=k<=8, -22<=l<=23
Reflections collected	13152
Independent reflections	5059 [R(int) = 0.0901]
Absorption correction	Semi-empirical from psi-scans
Max. and min. transmission	0.7099 and 0.5390
Refinement method	Full-matrix least-squares on F^2
Data / restraints / parameters	5059 / 0 / 451
Goodness-of-fit on F^2	1.102
Final R indices [I>2sigma(I)]	R1 = 0.0598, wR2 = 0.1155
R indices (all data)	R1 = 0.0984, wR2 = 0.1310
Largest diff. peak and hole	1.621 and -1.534 e.A^-3

Table S-VI. Crystal data and structure refinement for 7.

Identification code	Hf(OC(CH2Ph)2(TC-3,5)+ 4-phenyl-3-buten-2-one
Empirical formula	C48 H52 Hf N4 O2.25
Formula weight	899.43
Temperature	188(2) K
Wavelength	0.71073 A
Crystal system	Triclinic
Space group	P-1
Unit cell dimensions	a = 12.9669(2) A alpha = 96.8020(10) deg. b = 15.978(1) A beta = 90.3630(10) deg. c = 23.5610(4) A gamma = 102.4760(10) deg.
Volume, Z	4730.25(11) A^3, 4
Density (calculated)	1.263 Mg/m^3
Absorption coefficient	2.244 mm^-1
F(000)	1832
Crystal size	0.35 x 0.10 x 0.10 mm
Theta range for data collection	1.32 to 23.28 deg.
Limiting indices	-14<=h<=13, -17<=k<=17, -13<=l<=26
Reflections collected	18277
Independent reflections	12777 [R(int) = 0.0562]
Absorption correction	Semi-empirical from psi-scans
Max. and min. transmission	0.6839 and 0.4950
Refinement method	Full-matrix least-squares on F^2
Data / restraints / parameters	12772 / 0 / 1033
Goodness-of-fit on F^2	1.099
Final R indices [I>2sigma(I)]	R1 = 0.0685, wR2 = 0.1525
R indices (all data)	R1 = 0.1023, wR2 = 0.1796
Largest diff. peak and hole	1.228 and -1.821 e.A^-3

Table S-VII. Crystal data and structure refinement for 8.

Identification code	hf35cync ketenimine + h2o
Empirical formula	C51 H60 Cl2 Hf N6 O2
Formula weight	1038.44
Temperature	188(2) K
Wavelength	0.71073 A
Crystal system	Monoclinic
Space group	P2(1)/c
Unit cell dimensions	a = 12.95950(10) A alpha = 90 deg. b = 39.5559(3) A beta = 105.9370(10) deg. c = 9.81700(10) A gamma = 90 deg.
Volume, Z	4839.01(7) A^3, 4
Density (calculated)	1.425 Mg/m^3
Absorption coefficient	2.311 mm^-1
F(000)	2120
Crystal size	0.2 x 0.2 x 0.15 mm
Theta range for data collection	1.63 to 23.25 deg.
Limiting indices	-14<=h<=14, -43<=k<=24, -10<=l<=10
Reflections collected	18453
Independent reflections	6845 [R(int) = 0.0594]
Absorption correction	Semi-empirical from psi-scans
Max. and min. transmission	0.9510 and 0.6792
Refinement method	Full-matrix least-squares on F^2
Data / restraints / parameters	6845 / 0 / 568
Goodness-of-fit on F^2	1.258
Final R indices [I>2sigma(I)]	R1 = 0.0677, wR2 = 0.1146
R indices (all data)	R1 = 0.0884, wR2 = 0.1215
Largest diff. peak and hole	0.768 and -0.915 e.A^-3

Table S-VIII.Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters (A^2 x 10^3) for 2. U(eq) is defined as one third of the trace of the orthogonalized Uij tensor.

	x	y	z	U(eq)
Hf(1)	5000	1439(1)	9174(1)	22(1)
O(1)	5000	2378(2)	9583(5)	31(1)
N(1)	4155(3)	663(2)	9588(4)	27(1)
N(2)	4042(3)	1508(2)	7783(5)	25(1)
C(1)	5000	340(4)	11471(8)	39(2)
C(2)	4268(4)	218(3)	10708(6)	35(1)
C(3)	3516(3)	603(2)	8851(4)	26(1)
C(4)	2984(3)	100(3)	9033(9)	36(1)
C(5)	2272(4)	-32(3)	8487(7)	47(2)
C(6)	1859(4)	299(4)	7500(8)	53(2)
C(7)	2116(4)	840(4)	6853(7)	46(2)
C(8)	2800(4)	1178(3)	6967(6)	36(1)
C(9)	3443(3)	1102(2)	7808(5)	24(1)
C(10)	4027(4)	2071(3)	6875(6)	28(1)
C(11)	4248(3)	1910(3)	5440(5)	31(1)
C(12)	5000	1542(3)	5251(7)	26(1)
C(13)	5000	2100(4)	10903(7)	33(2)
C(14)	4277(5)	2312(5)	11675(7)	52(2)
C(15)	3504(4)	2130(3)	11094(6)	41(2)
C(16)	3109(5)	1625(4)	11588(8)	51(2)
C(17)	2410(7)	1441(4)	11065(11)	66(3)
C(18)	2083(5)	1777(5)	10000(9)	62(2)
C(19)	2487(5)	2296(5)	9490(7)	64(2)
C(20)	3193(4)	2469(2)	10029(5)	49(2)
C(111)	0(4)	-678(2)	9236(5)	29(2)
C(112)	0(4)	-71(2)	8620(5)	13(2)
C(113)	0(4)	69(2)	4862(5)	119(16)
C(114)	0(4)	1083(2)	9271(5)	42(3)
C(115)	0(4)	-392(2)	4651(5)	44(4)
C(116)	0(4)	-1168(2)	8202(5)	46(4)

Table S-IX. Bond lengths [A] and angles [deg] for 2.

Hf(1)-O(1)	1.993(5)
Hf(1)-N(2)#1	2.176(5)
Hf(1)-N(2)	2.176(5)
Hf(1)-N(1)	2.216(4)
Hf(1)-N(1)#1	2.216(4)
Hf(1)-C(13)	2.217(8)
O(1)-C(13)	1.448(9)
N(1)-C(3)	1.338(7)
N(1)-C(2)	1.469(6)
N(2)-C(9)	1.338(7)
N(2)-C(10)	1.485(7)
C(1)-C(2)#1	1.504(8)
C(1)-C(2)	1.504(8)
C(3)-C(4)	1.406(7)
C(3)-C(9)	1.481(7)
C(4)-C(5)	1.377(9)
C(5)-C(6)	1.403(11)
C(6)-C(7)	1.373(11)
C(7)-C(8)	1.382(10)
C(8)-C(9)	1.407(8)
C(10)-C(11)	1.532(7)
C(11)-C(12)	1.523(7)
C(12)-C(11)#1	1.523(7)
C(13)-C(14)#1	1.538(9)
C(13)-C(14)	1.538(9)
C(14)-C(15)	1.509(11)
C(15)-C(16)	1.349(11)
C(15)-C(20)	1.390(9)
C(16)-C(17)	1.37(2)
C(17)-C(18)	1.40(2)
C(18)-C(19)	1.384(14)
C(19)-C(20)	1.385(12)
C(111)-C(112)	1.40
C(111)-C(113)#2	1.413(9)
C(111)-C(116)	1.45
C(112)-C(113)#2	1.25
C(112)-C(115)#2	1.414(7)
C(113)-C(115)	0.98
C(113)-C(112)#3	1.25
C(113)-C(111)#3	1.413(9)
C(114)-C(115)#2	1.486(10)
C(115)-C(112)#3	1.414(7)
C(115)-C(114)#3	1.486(10)
O(1)-Hf(1)-N(2)#1	93.91(14)
O(1)-Hf(1)-N(2)	93.91(14)
N(2)#1-Hf(1)-N(2)	99.3(2)
O(1)-Hf(1)-N(1)	132.27(13)
N(2)#1-Hf(1)-N(1)	132.3(2)
N(2)-Hf(1)-N(1)	70.5(2)
O(1)-Hf(1)-N(1)#1	132.27(13)
N(2)#1-Hf(1)-N(1)#1	70.5(2)
N(2)-Hf(1)-N(1)#1	132.3(2)
N(1)-Hf(1)-N(1)#1	82.6(2)
O(1)-Hf(1)-C(13)	39.8(2)
N(2)#1-Hf(1)-C(13)	117.7(2)
N(2)-Hf(1)-C(13)	117.7(2)
N(1)-Hf(1)-C(13)	107.6(2)
N(1)#1-Hf(1)-C(13)	107.6(2)
C(13)-O(1)-Hf(1)	78.5(4)
C(3)-N(1)-C(2)	118.5(5)
C(3)-N(1)-Hf(1)	120.6(3)

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C(2)-N(1)-Hf(1)	120.9(4)
C(9)-N(2)-C(10)	119.6(5)
C(9)-N(2)-Hf(1)	122.6(3)
C(10)-N(2)-Hf(1)	117.5(4)
C(2)#1-C(1)-C(2)	115.0(7)
N(1)-C(2)-C(1)	113.5(5)
N(1)-C(3)-C(4)	122.6(5)
N(1)-C(3)-C(9)	113.6(4)
C(4)-C(3)-C(9)	123.8(5)
C(5)-C(4)-C(3)	133.1(7)
C(4)-C(5)-C(6)	129.9(7)
C(7)-C(6)-C(5)	124.7(6)
C(6)-C(7)-C(8)	130.9(7)
C(7)-C(8)-C(9)	132.1(7)
N(2)-C(9)-C(8)	122.2(5)
N(2)-C(9)-C(3)	112.8(4)
C(8)-C(9)-C(3)	125.0(5)
N(2)-C(10)-C(11)	113.8(5)
C(12)-C(11)-C(10)	116.2(5)
C(11)-C(12)-C(11)#1	117.7(7)
O(1)-C(13)-C(14)#1	110.4(5)
O(1)-C(13)-C(14)	110.4(5)
C(14)#1-C(13)-C(14)	109.1(8)
O(1)-C(13)-Hf(1)	61.7(3)
C(14)#1-C(13)-Hf(1)	125.0(4)
C(14)-C(13)-Hf(1)	125.0(4)
C(15)-C(14)-C(13)	117.1(6)
C(16)-C(15)-C(20)	118.8(7)
C(16)-C(15)-C(14)	120.1(7)
C(20)-C(15)-C(14)	121.1(7)
C(15)-C(16)-C(17)	121.4(8)
C(16)-C(17)-C(18)	120.9(9)
C(19)-C(18)-C(17)	117.9(8)
C(18)-C(19)-C(20)	120.2(7)
C(19)-C(20)-C(15)	120.9(7)
C(112)-C(111)-C(113)#2	52.7(2)
C(112)-C(111)-C(116)	108.2
C(113)#2-C(111)-C(116)	160.8(2)
C(113)#2-C(112)-C(111)	64.0(5)
C(113)#2-C(112)-C(115)#2	42.6(2)
C(111)-C(112)-C(115)#2	106.6(3)
C(115)-C(113)-C(112)#3	77.7(5)
C(115)-C(113)-C(111)#3	141.0(2)
C(112)#3-C(113)-C(111)#3	63.3(3)
C(113)-C(115)-C(112)#3	59.7(3)
C(113)-C(115)-C(114)#3	177.56(11)
C(112)#3-C(115)-C(114)#3	117.8(2)

Symmetry transformations used to generate equivalent atoms:
#1 -x+1,y,z #2 -x,-y,z+1/2 #3 -x,-y,z-1/2

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Table S-X. Anisotropic displacement parameters (A^2 x 10^3) for 2. The anisotropic displacement factor exponent takes the form: -2 pi^2 [h^2 a*^2 U11 + ... + 2 h k a* b* U12]

	U11	U22	U33	U23	U13	U12
Hf(1)	31(1)	19(1)	16(1)	0(1)	0	0
O(1)	54(3)	23(2)	17(2)	-1(2)	0	0
N(1)	33(2)	23(2)	24(2)	5(2)	6(2)	2(2)
N(2)	31(2)	23(2)	20(2)	1(2)	5(2)	4(2)
C(1)	48(5)	45(5)	24(4)	14(3)	0	0
C(2)	46(3)	28(3)	32(3)	14(2)	9(2)	4(2)
C(3)	32(2)	24(2)	23(3)	-4(2)	8(2)	5(2)
C(4)	37(2)	34(2)	38(4)	0(3)	5(3)	-7(2)
C(5)	42(3)	44(4)	54(4)	1(3)	5(3)	-15(3)
C(6)	43(4)	56(4)	59(4)	-8(3)	0(3)	-16(3)
C(7)	30(3)	65(4)	43(4)	-9(3)	-4(3)	-2(3)
C(8)	31(3)	51(4)	27(3)	-3(3)	0(2)	11(3)
C(9)	24(2)	27(2)	22(2)	-3(2)	5(2)	2(2)
C(10)	37(3)	28(3)	20(2)	4(2)	-3(2)	6(2)
C(11)	35(3)	40(3)	19(2)	4(2)	-1(2)	6(2)
C(12)	34(4)	25(3)	17(3)	-2(3)	0	0
C(13)	50(5)	35(4)	15(3)	-3(3)	0	0
C(14)	63(5)	65(5)	29(3)	-15(3)	7(3)	6(4)
C(15)	47(4)	50(4)	27(3)	-10(2)	9(3)	19(3)
C(16)	65(5)	44(4)	45(4)	0(3)	9(4)	13(4)
C(17)	70(6)	53(5)	76(7)	-12(4)	28(5)	11(4)
C(18)	46(4)	71(6)	70(5)	-26(4)	12(4)	18(4)
C(19)	64(4)	90(6)	37(5)	-6(3)	3(3)	46(4)
C(20)	59(4)	53(4)	35(3)	0(3)	11(3)	26(3)

Table S-XI. Hydrogen coordinates (x 10^4) and isotropic displacement parameters (A^2 x 10^3) for 2.

	x	y	z	U(eq)
H(1A)	5000	784(4)	11769(8)	47
H(1B)	5000	68(4)	12254(8)	47
H(2A)	4275(4)	-219(3)	10372(6)	42
H(2B)	3832(4)	257(3)	11308(6)	42
H(4A)	3145(3)	-206(3)	9647(9)	43
H(5A)	2028(4)	-396(3)	8823(7)	56
H(6A)	1375(4)	140(4)	7269(8)	63
H(7A)	1772(4)	1006(4)	6232(7)	55
H(8A)	2843(4)	1521(3)	6377(6)	44
H(10A)	4380(4)	2396(3)	7210(6)	34
H(10B)	3512(4)	2255(3)	6882(6)	34
H(11A)	3833(3)	1658(3)	5052(5)	37
H(11E)	4282(3)	2309(3)	4944(5)	37
H(12A)	5000	1363(3)	4361(7)	31
H(12B)	5000	1182(3)	5867(7)	31
H(14A)	4307(5)	2131(5)	12562(7)	62
H(14B)	4292(5)	2777(5)	11765(7)	62
H(16A)	3314(5)	1395(4)	12299(8)	61
H(17A)	2152(7)	1089(4)	11424(11)	80
H(18A)	1609(5)	1656(5)	9647(9)	75
H(19A)	2284(5)	2529(5)	8782(7)	77
H(20A)	3462(4)	2816(2)	9674(5)	58

Table S-XII. Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters (A^2 x 10^3) for 3. U(eq) is defined as one third of the trace of the orthogonalized Uij tensor.

	x	y	z	U(eq)
Hf(1)	5000	1866(1)	4309(1)	17(1)
Cl(1)	5000	2335(1)	6679(1)	34(1)
O(1)	5000	2750(2)	3718(3)	23(1)
N(1)	4196(2)	1131(1)	5136(3)	24(1)
N(2)	4118(2)	1679(1)	2793(3)	19(1)
C(1)	5000	435(3)	6625(5)	32(1)
C(2)	4290(2)	843(2)	6512(3)	28(1)
C(3)	3690(2)	869(1)	4285(7)	23(1)
C(4)	3345(2)	260(2)	4532(4)	30(1)
C(5)	2712(2)	-16(2)	3961(4)	40(1)
C(6)	2214(3)	230(2)	3015(5)	43(1)
C(7)	2306(2)	792(2)	2256(5)	37(1)
C(8)	2916(2)	1206(2)	2217(4)	30(1)
C(9)	3571(2)	1248(2)	3025(3)	21(1)
C(10)	4075(2)	2060(2)	1514(3)	22(1)
C(11)	4271(2)	1672(2)	215(4)	28(1)
C(12)	5000	1293(3)	275(5)	29(1)
C(13)	5000	3414(2)	3530(5)	26(1)
C(14)	4285(2)	3707(2)	4183(8)	38(1)
C(15)	3577(2)	3469(2)	3516(4)	35(1)
C(16)	3124(2)	3022(2)	4197(10)	43(1)
C(17)	2464(3)	2807(3)	3599(7)	60(2)
C(18)	2238(3)	3037(3)	2349(9)	77(2)
C(19)	2687(4)	3464(3)	1648(7)	73(2)
C(20)	3355(3)	3679(2)	2222(5)	52(1)
Cl(2)	5000	4636(2)	7327(3)	104(1)
Cl(3)	4638(4)	3744(4)	9565(7)	182(3)
C(21)	4767(10)	3985(16)	8275(25)	220(17)

Table S-XIII. Bond lengths [A] and angles [deg] for 3.

Hf(1)-O(1)	1.926(3)
Hf(1)-N(2)#1	2.186(3)
Hf(1)-N(2)	2.186(3)
Hf(1)-N(1)	2.242(3)
Hf(1)-N(1)#1	2.242(3)
Hf(1)-Cl(1)	2.5011(12)
O(1)-C(13)	1.395(6)
N(1)-C(3)	1.338(5)
N(1)-C(2)	1.474(4)
N(2)-C(9)	1.342(4)
N(2)-C(10)	1.476(4)
C(1)-C(2)	1.525(5)
C(1)-C(2)#1	1.525(5)
C(3)-C(4)	1.427(4)
C(3)-C(9)	1.471(6)
C(4)-C(5)	1.380(5)
C(5)-C(6)	1.376(6)
C(6)-C(7)	1.391(6)
C(7)-C(8)	1.386(5)
C(8)-C(9)	1.406(5)
C(10)-C(11)	1.538(5)
C(11)-C(12)	1.518(5)
C(12)-C(11)#1	1.518(5)
C(13)-C(14)	1.545(5)
C(13)-C(14)#1	1.545(5)
C(14)-C(15)	1.500(6)
C(15)-C(20)	1.388(6)
C(15)-C(16)	1.397(8)
C(16)-C(17)	1.382(7)
C(17)-C(18)	1.366(10)
C(18)-C(19)	1.375(10)
C(19)-C(20)	1.385(8)
Cl(2)-C(21)	1.69(2)
Cl(2)-C(21)#1	1.69(2)
Cl(3)-Cl(3)#1	1.286(13)
Cl(3)-C(21)	1.37(2)
Cl(3)-C(21)#1	1.71(2)
C(21)-C(21)#1	0.83(4)
C(21)-Cl(3)#1	1.71(2)
O(1)-Hf(1)-N(2)#1	88.21(10)
O(1)-Hf(1)-N(2)	88.21(10)
N(2)#1-Hf(1)-N(2)	91.65(14)
O(1)-Hf(1)-N(1)	139.31(8)
N(2)#1-Hf(1)-N(1)	125.25(11)
N(2)-Hf(1)-N(1)	70.32(10)
O(1)-Hf(1)-N(1)#1	139.31(8)
N(2)#1-Hf(1)-N(1)#1	70.32(10)
N(2)-Hf(1)-N(1)#1	125.25(11)
N(1)-Hf(1)-N(1)#1	79.2(2)
O(1)-Hf(1)-Cl(1)	84.37(10)
N(2)#1-Hf(1)-Cl(1)	133.61(7)
N(2)-Hf(1)-Cl(1)	133.61(7)
N(1)-Hf(1)-Cl(1)	86.36(8)
N(1)#1-Hf(1)-Cl(1)	86.36(8)
C(13)-O(1)-Hf(1)	170.2(3)
C(3)-N(1)-C(2)	118.1(3)
C(3)-N(1)-Hf(1)	119.0(3)
C(2)-N(1)-Hf(1)	122.0(2)
C(9)-N(2)-C(10)	117.6(3)
C(9)-N(2)-Hf(1)	121.7(2)
C(10)-N(2)-Hf(1)	120.6(2)

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C(2)-C(1)-C(2)#1	111.7(4)
N(1)-C(2)-C(1)	112.6(3)
N(1)-C(3)-C(4)	123.1(5)
N(1)-C(3)-C(9)	113.1(3)
C(4)-C(3)-C(9)	123.6(4)
C(5)-C(4)-C(3)	130.7(4)
C(6)-C(5)-C(4)	129.7(4)
C(5)-C(6)-C(7)	126.2(4)
C(8)-C(7)-C(6)	129.0(4)
C(7)-C(8)-C(9)	132.1(4)
N(2)-C(9)-C(8)	123.1(3)
N(2)-C(9)-C(3)	113.2(3)
C(8)-C(9)-C(3)	123.4(3)
N(2)-C(10)-C(11)	113.4(3)
C(12)-C(11)-C(10)	115.8(3)
C(11)#1-C(12)-C(11)	117.2(4)
O(1)-C(13)-C(14)	109.7(3)
O(1)-C(13)-C(14)#1	109.7(3)
C(14)-C(13)-C(14)#1	110.6(5)
C(15)-C(14)-C(13)	112.5(5)
C(20)-C(15)-C(16)	118.3(5)
C(20)-C(15)-C(14)	121.8(5)
C(16)-C(15)-C(14)	119.9(5)
C(17)-C(16)-C(15)	120.3(8)
C(18)-C(17)-C(16)	120.7(7)
C(17)-C(18)-C(19)	119.7(5)
C(18)-C(19)-C(20)	120.5(6)
C(19)-C(20)-C(15)	120.4(6)
C(21)-Cl(2)-C(21)#1	28.3(12)
Cl(3)#1-Cl(3)-C(21)	80.4(9)
Cl(3)#1-Cl(3)-C(21)#1	52.0(6)
C(21)-Cl(3)-C(21)#1	28.4(13)
C(21)#1-C(21)-Cl(3)	99.6(9)
C(21)#1-C(21)-Cl(2)	75.8(6)
Cl(3)-C(21)-Cl(2)	146(3)
C(21)#1-C(21)-Cl(3)#1	52.0(6)
Cl(3)-C(21)-Cl(3)#1	47.7(8)
Cl(2)-C(21)-Cl(3)#1	119(2)

Symmetry transformations used to generate equivalent atoms:
#1 -x+1,y,z

Table S-IX. Anisotropic displacement parameters (A^2 x 10^3) for 3. The anisotropic displacement factor exponent takes the form: -2 pi^2 [h^2 a*^2 U11 + ... + 2 h k a* b* U12]

	U11	U22	U33	U23	U13	U12
Hf(1)	20(1)	16(1)	14(1)	0(1)	0	0
Cl(1)	57(1)	27(1)	19(1)	-5(1)	0	0
O(1)	31(2)	18(2)	21(2)	1(1)	0	0
N(1)	26(1)	25(2)	19(1)	2(1)	2(1)	-4(1)
N(2)	22(1)	18(1)	18(1)	1(1)	1(1)	0(1)
C(1)	52(3)	24(2)	20(2)	8(2)	0	0
C(2)	39(2)	28(2)	16(2)	4(1)	2(1)	-8(2)
C(3)	24(1)	26(1)	20(1)	-2(3)	4(3)	-2(1)
C(4)	39(2)	28(2)	22(3)	2(1)	2(2)	-9(1)
C(5)	48(2)	30(2)	40(3)	1(2)	4(2)	-15(2)
C(6)	39(2)	43(3)	47(2)	-4(2)	-1(2)	-19(2)
C(7)	28(2)	40(2)	43(2)	-1(2)	-5(2)	-7(2)
C(8)	29(2)	36(2)	24(2)	0(2)	-2(1)	-9(2)
C(9)	22(2)	21(2)	21(2)	-3(1)	4(1)	-2(1)
C(10)	24(2)	23(2)	19(2)	5(1)	-1(1)	0(1)
C(11)	30(2)	37(2)	17(2)	2(1)	-5(1)	-8(2)
C(12)	38(3)	29(3)	19(2)	-2(2)	0	0
C(13)	35(3)	19(2)	25(3)	-1(2)	0	0
C(14)	39(2)	22(2)	51(3)	-9(2)	3(3)	6(1)
C(15)	32(2)	31(2)	41(2)	-10(2)	-1(2)	14(2)
C(16)	37(2)	48(2)	44(3)	-14(3)	9(3)	6(2)
C(17)	32(2)	65(4)	84(4)	-34(3)	12(2)	0(2)
C(18)	34(3)	85(5)	112(6)	-54(4)	-20(3)	23(3)
C(19)	70(4)	79(4)	70(4)	-27(3)	-37(3)	53(4)
C(20)	61(3)	41(3)	53(3)	-2(2)	-9(2)	28(2)
Cl(2)	99(2)	93(2)	119(2)	-52(2)	0	0
Cl(3)	167(5)	281(8)	97(5)	30(5)	45(4)	40(5)
C(21)	70(14)	376(37)	214(24)	235(26)	15(12)	36(15)

Table S-XV. Hydrogen coordinates (x 10^4) and isotropic displacement parameters (A^2 x 10^3) for 3.

	x	y	z	U(eq)
H(1A)	5000	114(3)	5900(5)	38
H(1B)	5000	211(3)	7501(5)	38
H(2A)	3855(2)	577(2)	6715(3)	33
H(2B)	4309(2)	1183(2)	7193(3)	33
H(4A)	3588(2)	8(2)	5186(4)	36
H(5A)	2607(2)	-431(2)	4261(4)	48
H(6A)	1773(3)	-1(2)	2868(5)	52
H(7A)	1902(2)	904(2)	1697(5)	44
H(8A)	2887(2)	1515(2)	1527(4)	36
H(10A)	4419(2)	2421(2)	1585(3)	27
H(10B)	3570(2)	2231(2)	1419(3)	27
H(11A)	3862(2)	1374(2)	35(4)	34
H(11B)	4298(2)	1966(2)	-558(4)	34
H(12A)	5000	1044(3)	1120(5)	34
H(12B)	5000	990(3)	-484(5)	34
H(13A)	5000	3508(2)	2541(5)	31
H(14A)	4274(2)	3600(2)	5155(8)	45
H(14B)	4307(2)	4171(2)	4104(8)	45
H(16A)	3266(2)	2868(2)	5057(10)	51
H(17A)	2172(3)	2504(3)	4053(7)	73
H(18A)	1782(3)	2905(3)	1974(9)	92
H(19A)	2542(4)	3609(3)	782(7)	88
H(20A)	3656(3)	3966(2)	1738(5)	62

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Table S-XVI.Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters (A^2 x 10^3) for 4. U(eq) is defined as one third of the trace of the orthogonalized Uij tensor.

	x	y	z	U(eq)
Hf(1)	-133(1)	-3871(1)	-2427(1)	21(1)
O(1)	-461(3)	-3315(3)	-3436(2)	23(1)
N(1)	-1716(4)	-5018(4)	-2962(3)	23(1)
N(2)	481(4)	-5494(3)	-2824(3)	22(1)
N(3)	726(4)	-4068(4)	-1243(3)	26(1)
N(4)	-1194(4)	-3142(3)	-1386(3)	24(1)
N(5)	1774(4)	42(4)	-2874(3)	27(1)
N(6)	1126(4)	-2488(3)	-2330(3)	20(1)
C(1)	-1547(5)	-6060(4)	-3257(3)	23(1)
C(2)	-2498(5)	-6815(5)	-3622(3)	31(1)
C(3)	-2579(6)	-7912(5)	-3886(4)	37(2)
C(4)	-1713(6)	-8631(5)	-3917(3)	35(2)
C(5)	-523(6)	-8360(5)	-3743(4)	39(2)
C(6)	89(6)	-7372(4)	-3468(3)	30(1)
C(7)	-292(5)	-6330(4)	-3184(3)	23(1)
C(8)	1728(5)	-5721(5)	-2675(3)	29(1)
C(9)	2417(5)	-4800(5)	-2100(4)	29(1)
C(10)	1881(5)	-4554(5)	-1249(3)	30(1)
C(11)	258(5)	-3635(4)	-526(3)	25(1)
C(12)	744(6)	-3760(5)	229(3)	37(2)
C(13)	550(6)	-3241(6)	1048(4)	45(2)
C(14)	-229(7)	-2462(6)	1354(4)	48(2)
C(15)	-1068(6)	-2103(5)	905(4)	43(2)
C(16)	-1359(6)	-2377(5)	69(3)	34(1)
C(17)	-813(5)	-3043(4)	-597(3)	24(1)
C(18)	-2231(5)	-2563(4)	-1557(3)	27(1)
C(19)	-3456(5)	-3066(5)	-1370(4)	35(1)
C(20)	-3619(6)	-4290(5)	-1598(4)	35(1)
C(21)	-3869(5)	-4789(5)	-2508(4)	38(2)
C(22)	-2884(5)	-4617(5)	-3106(4)	30(1)
C(23)	199(5)	-2462(4)	-3671(3)	22(1)
C(24)	-667(5)	-1702(4)	-3933(3)	25(1)
C(25)	-1626(5)	-1275(4)	-3336(3)	26(1)
C(26)	-2827(5)	-1674(5)	-3484(4)	32(1)
C(27)	-3738(6)	-1247(5)	-2966(4)	44(2)
C(28)	-3451(6)	-406(5)	-2304(4)	42(2)
C(29)	-2272(6)	4(5)	-2150(4)	37(2)
C(30)	-1367(5)	-435(5)	-2661(3)	31(1)
C(31)	961(5)	-2893(4)	-4437(3)	24(1)
C(32)	1945(5)	-3603(4)	-4353(3)	26(1)
C(33)	1784(5)	-4710(5)	-4656(4)	33(1)
C(34)	2706(6)	-5363(5)	-4625(4)	41(2)
C(35)	3830(6)	-4940(5)	-4303(4)	40(2)
C(36)	3997(6)	-3857(5)	-4010(4)	44(2)
C(37)	3079(6)	-3193(5)	-4038(4)	39(2)
C(38)	999(4)	-1887(4)	-2930(3)	19(1)
C(39)	1549(5)	-935(4)	-2934(3)	25(1)
C(40)	2909(5)	433(4)	-3224(3)	26(1)
C(41)	3245(6)	1563(5)	-2747(4)	37(2)
C(42)	4276(7)	2119(6)	-3129(4)	50(2)
C(43)	4032(6)	2062(5)	-4036(5)	47(2)
C(44)	3788(7)	927(6)	-4495(4)	46(2)
C(45)	2721(6)	386(6)	-4129(4)	44(2)
C(46)	1822(5)	-1933(4)	-1577(3)	24(1)
C(47)	3146(5)	-1628(5)	-1702(4)	31(1)
C(48)	3799(6)	-1210(5)	-867(4)	41(2)

C(49)	3185(6)	-267(6)	-343(4)	51(2)
C(50)	1848(6)	-568(5)	-235(4)	44(2)
C(51)	1202(5)	-978(4)	-1076(3)	28(1)
Cl(1)	-5900(2)	3720(2)	-781(1)	58(1)
Cl(2)	-3881(3)	2414(3)	-1224(2)	129(1)
C(52)	-4459(8)	3445(7)	-532(6)	71(2)

Table S-XVII. Bond lengths [A] and angles [deg] for 4.

Hf(1)-O(1)	2.012(3)
Hf(1)-N(6)	2.155(4)
Hf(1)-N(4)	2.173(4)
Hf(1)-N(2)	2.201(4)
Hf(1)-N(1)	2.254(4)
Hf(1)-N(3)	2.262(4)
O(1)-C(23)	1.402(6)
N(1)-C(1)	1.345(7)
N(1)-C(22)	1.470(7)
N(2)-C(7)	1.350(7)
N(2)-C(8)	1.468(7)
N(3)-C(11)	1.324(7)
N(3)-C(10)	1.468(7)
N(4)-C(17)	1.356(7)
N(4)-C(18)	1.477(7)
N(5)-C(39)	1.243(7)
N(5)-C(40)	1.492(7)
N(6)-C(38)	1.406(6)
N(6)-C(46)	1.481(6)
C(1)-C(2)	1.418(8)
C(1)-C(7)	1.472(8)
C(2)-C(3)	1.375(8)
C(3)-C(4)	1.375(9)
C(4)-C(5)	1.356(9)
C(5)-C(6)	1.376(8)
C(6)-C(7)	1.413(8)
C(8)-C(9)	1.503(8)
C(9)-C(10)	1.516(8)
C(11)-C(12)	1.415(8)
C(11)-C(17)	1.477(8)
C(12)-C(13)	1.403(9)
C(13)-C(14)	1.383(10)
C(14)-C(15)	1.362(10)
C(15)-C(16)	1.389(8)
C(16)-C(17)	1.408(8)
C(18)-C(19)	1.527(8)
C(19)-C(20)	1.529(8)
C(20)-C(21)	1.524(9)
C(21)-C(22)	1.520(8)
C(23)-C(38)	1.532(7)
C(23)-C(24)	1.548(7)
C(23)-C(31)	1.552(7)
C(24)-C(25)	1.509(8)
C(25)-C(30)	1.386(8)
C(25)-C(26)	1.395(8)
C(26)-C(27)	1.395(9)
C(27)-C(28)	1.377(10)
C(28)-C(29)	1.377(9)
C(29)-C(30)	1.388(8)
C(31)-C(32)	1.501(8)
C(32)-C(37)	1.384(8)
C(32)-C(33)	1.396(8)
C(33)-C(34)	1.379(9)
C(34)-C(35)	1.383(9)
C(35)-C(36)	1.365(9)
C(36)-C(37)	1.384(9)
C(38)-C(39)	1.328(7)
C(40)-C(45)	1.508(8)
C(40)-C(41)	1.510(8)
C(41)-C(42)	1.522(8)
C(42)-C(43)	1.519(10)
C(43)-C(44)	1.492(10)

C(44)-C(45)	1.528(9)
C(46)-C(47)	1.522(8)
C(46)-C(51)	1.531(8)
C(47)-C(48)	1.534(8)
C(48)-C(49)	1.532(10)
C(49)-C(50)	1.529(10)
C(50)-C(51)	1.538(8)
Cl(1)-C(52)	1.734(9)
Cl(2)-C(52)	1.715(9)

O(1)-Hf(1)-N(6)	75.1(2)
O(1)-Hf(1)-N(4)	112.3(2)
N(6)-Hf(1)-N(4)	94.7(2)
O(1)-Hf(1)-N(2)	108.4(2)
N(6)-Hf(1)-N(2)	120.2(2)
N(4)-Hf(1)-N(2)	131.8(2)
O(1)-Hf(1)-N(1)	80.89(14)
N(6)-Hf(1)-N(1)	155.7(2)
N(4)-Hf(1)-N(1)	91.1(2)
N(2)-Hf(1)-N(1)	70.9(2)
O(1)-Hf(1)-N(3)	161.3(2)
N(6)-Hf(1)-N(3)	86.2(2)
N(4)-Hf(1)-N(3)	70.3(2)
N(2)-Hf(1)-N(3)	79.4(2)
N(1)-Hf(1)-N(3)	117.8(2)
C(23)-O(1)-Hf(1)	125.7(3)
C(1)-N(1)-C(22)	119.2(4)
C(1)-N(1)-Hf(1)	119.5(3)
C(22)-N(1)-Hf(1)	120.6(3)
C(7)-N(2)-C(8)	117.4(4)
C(7)-N(2)-Hf(1)	121.0(3)
C(8)-N(2)-Hf(1)	121.6(3)
C(11)-N(3)-C(10)	118.9(4)
C(11)-N(3)-Hf(1)	119.9(4)
C(10)-N(3)-Hf(1)	120.8(3)
C(17)-N(4)-C(18)	119.5(4)
C(17)-N(4)-Hf(1)	122.0(4)
C(18)-N(4)-Hf(1)	117.4(3)
C(39)-N(5)-C(40)	119.5(5)
C(38)-N(6)-C(46)	115.7(4)
C(38)-N(6)-Hf(1)	116.0(3)
C(46)-N(6)-Hf(1)	125.3(3)
N(1)-C(1)-C(2)	122.3(5)
N(1)-C(1)-C(7)	114.1(4)
C(2)-C(1)-C(7)	123.6(5)
C(3)-C(2)-C(1)	132.8(6)
C(4)-C(3)-C(2)	130.3(6)
C(5)-C(4)-C(3)	124.9(5)
C(4)-C(5)-C(6)	130.5(6)
C(5)-C(6)-C(7)	133.0(6)
N(2)-C(7)-C(6)	122.0(5)
N(2)-C(7)-C(1)	114.6(5)
C(6)-C(7)-C(1)	123.5(5)
N(2)-C(8)-C(9)	112.4(4)
C(8)-C(9)-C(10)	112.3(5)
N(3)-C(10)-C(9)	112.1(4)
N(3)-C(11)-C(12)	121.9(5)
N(3)-C(11)-C(17)	113.9(5)
C(12)-C(11)-C(17)	124.1(5)
C(13)-C(12)-C(11)	131.9(6)
C(14)-C(13)-C(12)	129.2(6)
C(15)-C(14)-C(13)	125.8(6)
C(14)-C(15)-C(16)	131.3(7)
C(15)-C(16)-C(17)	131.3(6)
N(4)-C(17)-C(16)	121.8(5)
N(4)-C(17)-C(11)	113.2(5)

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C(16)-C(17)-C(11)	124.8(5)
N(4)-C(18)-C(19)	114.2(4)
C(18)-C(19)-C(20)	116.3(5)
C(21)-C(20)-C(19)	115.4(5)
C(22)-C(21)-C(20)	117.2(5)
N(1)-C(22)-C(21)	115.6(5)
O(1)-C(23)-C(38)	106.9(4)
O(1)-C(23)-C(24)	110.3(4)
C(38)-C(23)-C(24)	111.8(4)
O(1)-C(23)-C(31)	110.0(4)
C(38)-C(23)-C(31)	111.6(4)
C(24)-C(23)-C(31)	106.3(4)
C(25)-C(24)-C(23)	117.5(4)
C(30)-C(25)-C(26)	117.9(5)
C(30)-C(25)-C(24)	121.8(5)
C(26)-C(25)-C(24)	120.2(5)
C(25)-C(26)-C(27)	121.1(6)
C(28)-C(27)-C(26)	119.5(6)
C(29)-C(28)-C(27)	120.3(6)
C(28)-C(29)-C(30)	119.9(6)
C(25)-C(30)-C(29)	121.2(6)
C(32)-C(31)-C(23)	118.6(4)
C(37)-C(32)-C(33)	116.9(5)
C(37)-C(32)-C(31)	122.0(5)
C(33)-C(32)-C(31)	120.9(5)
C(34)-C(33)-C(32)	121.4(6)
C(33)-C(34)-C(35)	121.0(6)
C(36)-C(35)-C(34)	118.0(6)
C(35)-C(36)-C(37)	121.6(6)
C(32)-C(37)-C(36)	121.2(6)
C(39)-C(38)-N(6)	126.8(5)
C(39)-C(38)-C(23)	119.0(4)
N(6)-C(38)-C(23)	114.0(4)
N(5)-C(39)-C(38)	163.5(6)
N(5)-C(40)-C(45)	109.9(5)
N(5)-C(40)-C(41)	107.7(4)
C(45)-C(40)-C(41)	111.5(5)
C(40)-C(41)-C(42)	112.9(5)
C(43)-C(42)-C(41)	111.7(5)
C(44)-C(43)-C(42)	110.6(5)
C(43)-C(44)-C(45)	110.7(6)
C(40)-C(45)-C(44)	112.4(5)
N(6)-C(46)-C(47)	115.7(4)
N(6)-C(46)-C(51)	112.8(4)
C(47)-C(46)-C(51)	111.2(4)
C(46)-C(47)-C(48)	110.2(5)
C(49)-C(48)-C(47)	111.2(5)
C(50)-C(49)-C(48)	111.2(5)
C(49)-C(50)-C(51)	110.7(5)
C(46)-C(51)-C(50)	109.9(5)
Cl(2)-C(52)-Cl(1)	114.4(5)

Symmetry transformations used to generate equivalent atoms:

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Table S-XVIII.Anisotropic displacement parameters (A^2 x 10^3) for 4.
The anisotropic displacement factor exponent takes the form:
-2 pi^2 [h^2 a*^2 U11 + ... + 2 h k a* b* U12]

	U11	U22	U33	U23	U13	U12
Hf(1)	25(1)	20(1)	15(1)	0(1)	-2(1)	-1(1)
O(1)	30(2)	21(2)	15(2)	2(2)	-6(2)	-4(2)
N(1)	25(2)	25(2)	19(2)	4(2)	-1(2)	-4(2)
N(2)	26(2)	24(2)	17(2)	4(2)	0(2)	2(2)
N(3)	25(2)	24(2)	26(3)	3(2)	-9(2)	-2(2)
N(4)	27(2)	20(2)	23(2)	4(2)	-3(2)	0(2)
N(5)	32(3)	22(3)	26(2)	2(2)	-1(2)	-3(2)
N(6)	24(2)	16(2)	18(2)	-3(2)	-7(2)	-4(2)
C(1)	29(3)	23(3)	16(3)	1(2)	1(2)	-2(2)
C(2)	31(3)	28(3)	26(3)	-4(2)	0(2)	-4(3)
C(3)	45(4)	31(3)	25(3)	-6(3)	3(3)	-10(3)
C(4)	61(4)	18(3)	20(3)	-2(2)	-10(3)	-12(3)
C(5)	65(5)	23(3)	25(3)	-3(3)	-19(3)	10(3)
C(6)	42(3)	27(3)	16(3)	-8(2)	-11(2)	9(3)
C(7)	33(3)	25(3)	9(2)	3(2)	4(2)	0(2)
C(8)	25(3)	33(3)	26(3)	2(2)	3(2)	9(2)
C(9)	21(3)	28(3)	37(3)	6(3)	-5(2)	-2(2)
C(10)	32(3)	27(3)	28(3)	2(2)	-10(2)	-1(2)
C(11)	30(3)	21(3)	19(3)	0(2)	-5(2)	-7(2)
C(12)	47(4)	39(4)	20(3)	4(3)	-11(3)	-8(3)
C(13)	59(4)	47(4)	29(3)	14(3)	-14(3)	-21(4)
C(14)	76(5)	46(4)	13(3)	-2(3)	-1(3)	-18(4)
C(15)	63(5)	40(4)	21(3)	-1(3)	11(3)	-8(3)
C(16)	48(4)	32(3)	17(3)	-2(2)	8(3)	0(3)
C(17)	28(3)	22(3)	21(3)	5(2)	0(2)	-9(2)
C(18)	34(3)	24(3)	22(3)	1(2)	2(2)	8(2)
C(19)	29(3)	34(3)	38(3)	0(3)	7(3)	8(3)
C(20)	31(3)	38(3)	36(3)	8(3)	9(3)	2(3)
C(21)	22(3)	34(3)	53(4)	1(3)	-1(3)	0(3)
C(22)	25(3)	29(3)	31(3)	1(2)	-7(2)	-1(2)
C(23)	30(3)	19(3)	16(3)	5(2)	-4(2)	-1(2)
C(24)	32(3)	23(3)	19(3)	2(2)	-2(2)	3(2)
C(25)	36(3)	20(3)	23(3)	6(2)	-2(2)	8(2)
C(26)	39(4)	26(3)	29(3)	0(3)	-5(3)	4(3)
C(27)	34(4)	47(4)	51(4)	12(3)	1(3)	10(3)
C(28)	48(4)	44(4)	36(4)	6(3)	10(3)	17(3)
C(29)	50(4)	28(3)	31(3)	2(3)	2(3)	14(3)
C(30)	37(3)	28(3)	29(3)	7(3)	0(3)	2(3)
C(31)	33(3)	22(3)	15(3)	2(2)	-2(2)	-2(2)
C(32)	33(3)	28(3)	15(3)	1(2)	2(2)	2(2)
C(33)	33(3)	28(3)	34(3)	-2(3)	-2(3)	-2(3)
C(34)	50(4)	26(3)	44(4)	1(3)	6(3)	10(3)
C(35)	37(4)	46(4)	35(3)	3(3)	7(3)	10(3)
C(36)	29(3)	47(4)	50(4)	0(3)	-6(3)	2(3)
C(37)	40(4)	30(3)	39(4)	-6(3)	-4(3)	1(3)
C(38)	20(3)	19(3)	16(3)	3(2)	3(2)	-2(2)
C(39)	29(3)	31(3)	13(3)	2(2)	-5(2)	2(3)
C(40)	27(3)	23(3)	28(3)	5(2)	2(2)	3(2)
C(41)	39(4)	36(3)	28(3)	-2(3)	1(3)	-14(3)
C(42)	53(4)	46(4)	41(4)	-6(3)	9(3)	-22(3)
C(43)	44(4)	42(4)	63(5)	24(4)	18(3)	5(3)
C(44)	59(4)	54(4)	25(3)	10(3)	7(3)	-3(3)
C(45)	55(4)	46(4)	25(3)	0(3)	2(3)	-19(3)
C(46)	32(3)	19(3)	15(3)	-1(2)	-6(2)	-7(2)
C(47)	31(3)	27(3)	30(3)	0(3)	-7(3)	-2(2)
C(48)	36(4)	42(4)	42(4)	6(3)	-16(3)	-12(3)

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C(49)	58(5)	46(4)	38(4)	-4(3)	-25(3)	-19(3)
C(50)	62(5)	41(4)	20(3)	-8(3)	-9(3)	-7(3)
C(51)	36(3)	26(3)	17(3)	-2(2)	-2(2)	-6(2)
Cl(1)	54(1)	67(1)	54(1)	11(1)	-4(1)	4(1)
Cl(2)	107(2)	156(3)	99(2)	-41(2)	-30(2)	72(2)
C(52)	70(6)	70(6)	73(6)	14(5)	-10(5)	10(5)

Table S-XIX. Hydrogen coordinates (x 10^4) and isotropic displacement parameters (A^2 x 10^3) for 4.

	x	y	z	U(eq)
H(2A)	-3209(5)	-6507(5)	-3699(3)	37
H(3A)	-3343(6)	-8217(5)	-4073(4)	44
H(4A)	-1962(6)	-9356(5)	-4069(3)	42
H(5B)	-43(6)	-8936(5)	-3822(4)	47
H(6A)	924(6)	-7395(4)	-3470(3)	36
H(8A)	2140(5)	-5881(5)	-3196(3)	34
H(8B)	1718(5)	-6350(5)	-2443(3)	34
H(9A)	2419(5)	-4169(5)	-2329(4)	35
H(9B)	3248(5)	-4969(5)	-2055(4)	35
H(10A)	1760(5)	-5212(5)	-1056(3)	36
H(10B)	2446(5)	-4070(5)	-871(3)	36
H(12A)	1295(6)	-4282(5)	172(3)	44
H(13A)	1012(6)	-3452(6)	1442(4)	54
H(14A)	-176(7)	-2157(6)	1915(4)	57
H(15A)	-1526(6)	-1584(5)	1212(4)	52
H(16A)	-2032(6)	-2066(5)	-77(3)	40
H(18A)	-2225(5)	-2532(4)	-2133(3)	33
H(18B)	-2131(5)	-1833(4)	-1232(3)	33
H(19A)	-4068(5)	-2652(5)	-1131(4)	42
H(20A)	-2895(6)	-4569(5)	-1429(4)	42
H(20B)	-4282(6)	-4520(5)	-1284(4)	42
H(21A)	-4039(5)	-5555(5)	-2569(4)	45
H(21B)	-4595(5)	-4509(5)	-2672(4)	45
H(22A)	-3167(5)	-4964(5)	-3659(4)	35
H(22B)	-2758(5)	-3856(5)	-3082(4)	35
H(24A)	-182(5)	-1095(4)	-4048(3)	30
H(24B)	-1066(5)	-2079(4)	-4446(3)	30
H(26A)	-3023(5)	-2234(5)	-3936(4)	39
H(27A)	-4534(6)	-1528(5)	-3066(4)	52
H(28A)	-4056(6)	-114(5)	-1960(4)	51
H(29A)	-2082(6)	575(5)	-1705(4)	44
H(30A)	-572(5)	-159(5)	-2549(3)	37
H(31A)	411(5)	-3284(4)	-4872(3)	29
H(31B)	1325(5)	-2280(4)	-4621(3)	29
H(33A)	1039(5)	-5013(5)	-4885(4)	40
H(34A)	2569(6)	-6099(5)	-4824(4)	49
H(35A)	4454(6)	-5379(5)	-4286(4)	48
H(36A)	4745(6)	-3558(5)	-3787(4)	52
H(37A)	3226(6)	-2457(5)	-3841(4)	46
H(40A)	3553(5)	-24(4)	-3156(3)	31
H(41A)	3475(6)	1546(5)	-2189(4)	44
H(41B)	2543(6)	1976(5)	-2719(4)	44
H(42A)	4389(7)	2863(6)	-2837(4)	61
H(42B)	5015(7)	1783(6)	-3070(4)	61
H(43A)	3342(6)	2462(5)	-4094(5)	57
H(43B)	4725(6)	2386(5)	-4266(5)	57
H(44A)	4499(7)	539(6)	-4470(4)	56
H(44B)	3612(7)	906(6)	-5069(4)	56
H(45A)	1997(6)	734(6)	-4204(4)	53
H(45B)	2600(6)	-358(6)	-4423(4)	53
H(46A)	1822(5)	-2452(4)	-1228(3)	28
H(47A)	3212(5)	-1078(5)	-2018(4)	37
H(47B)	3524(5)	-2249(5)	-2009(4)	37
H(48A)	3801(6)	-1784(5)	-575(4)	50
H(48B)	4631(6)	-984(5)	-955(4)	50
H(49A)	3584(6)	-49(6)	193(4)	61
H(49B)	3263(6)	336(6)	-606(4)	61

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H(50A)	1471(6)	54(5)	71(4)	53
H(50B)	1769(6)	-1120(5)	79(4)	53
H(51A)	364(5)	-1195(4)	-996(3)	34
H(51B)	1222(5)	-409(4)	-1373(3)	34
H(52A)	-3923(8)	4087(7)	-491(6)	85
H(52B)	-4473(8)	3272(7)	6(6)	85

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Table S-XX. Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters (A^2 x 10^3) for 5. U(eq) is defined as one third of the trace of the orthogonalized Uij tensor.

	x	y	z	U(eq)
Hf(1)	4971(1)	9239(1)	1249(1)	62(1)
O(1)	4200(5)	8747(10)	1017(3)	67(3)
N(1)	5299(8)	9287(13)	714(4)	78(4)
N(2)	5459(7)	11093(15)	1129(4)	85(5)
N(3)	5610(6)	9305(14)	1692(3)	74(4)
N(4)	5503(7)	7517(12)	1287(3)	66(4)
N(5)	4094(7)	11375(14)	1694(4)	73(4)
N(6)	4280(6)	8114(12)	1625(3)	59(3)
C(1)	5609(7)	10258(15)	584(4)	62(4)
C(2)	5805(13)	10256(26)	257(6)	141(11)
C(3)	6128(12)	11138(26)	72(7)	132(10)
C(4)	6358(12)	12175(26)	155(7)	120(9)
C(5)	6261(21)	12787(37)	438(9)	274(29)
C(6)	6018(23)	12325(35)	738(8)	312(33)
C(7)	5724(10)	11225(19)	828(5)	87(6)
C(8)	5562(13)	12031(23)	1388(6)	123(9)
C(9)	6060(15)	11538(33)	1711(11)	231(22)
C(10)	5658(10)	10418(20)	1922(6)	103(7)
C(11)	6056(9)	8448(18)	1718(4)	77(5)
C(12)	6567(10)	8528(22)	1959(6)	113(8)
C(13)	7050(14)	7794(34)	2039(7)	157(13)
C(14)	7192(15)	6716(35)	1968(9)	145(12)
C(15)	6869(15)	5949(32)	1777(9)	150(12)
C(16)	6292(14)	6193(25)	1538(8)	151(11)
C(17)	5980(10)	7358(19)	1507(5)	82(6)
C(18)	5362(9)	6486(17)	1060(4)	78(5)
C(19)	5813(16)	5995(28)	800(7)	166(14)
C(20)	5975(21)	7070(25)	602(10)	213(19)
C(21)	5568(22)	7633(49)	332(9)	233(22)
C(22)	5110(14)	8264(23)	505(6)	125(11)
C(23)	4378(7)	10679(14)	1537(4)	54(4)
C(24)	3625(7)	8289(14)	1145(4)	58(4)
C(25)	3135(8)	9381(15)	1143(4)	63(4)
C(26)	3047(8)	10014(16)	810(4)	65(4)
C(27)	2581(9)	9656(16)	584(4)	70(5)
C(28)	2499(10)	10263(20)	292(4)	80(5)
C(29)	2860(9)	11230(19)	189(4)	78(5)
C(30)	3322(9)	11614(16)	419(4)	76(5)
C(31)	3409(8)	11002(13)	717(4)	58(4)
C(32)	3359(9)	7225(16)	913(4)	75(5)
C(33)	3726(8)	6027(14)	886(4)	65(5)
C(34)	3710(9)	5150(17)	1146(5)	81(6)
C(35)	4029(11)	4035(20)	1122(6)	102(7)
C(36)	4339(11)	3756(18)	826(6)	93(7)
C(37)	4355(10)	4609(18)	564(5)	85(6)
C(38)	4062(11)	5726(16)	600(5)	90(6)
C(39)	3750(9)	7906(12)	1493(4)	63(5)
C(40)	3195(8)	7377(16)	1684(4)	74(5)
C(41)	2780(9)	8327(16)	1882(4)	62(4)
C(42)	3000(9)	9358(17)	2043(4)	75(5)
C(43)	2610(10)	10118(17)	2230(5)	79(5)
C(44)	1979(11)	9850(20)	2252(5)	91(6)
C(45)	1735(10)	8787(21)	2076(5)	86(6)
C(46)	2149(10)	8083(16)	1895(4)	76(5)
C(47)	4374(8)	7929(15)	1985(4)	65(4)
C(48)	4795(9)	6836(16)	2052(4)	63(5)

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C(49)	5224(8)	6906(16)	2308(5)	73(5)
C(50)	5593(9)	5881(18)	2398(4)	77(5)
C(51)	5502(10)	4757(19)	2243(5)	91(7)
C(52)	5047(10)	4637(22)	1982(6)	105(8)
C(53)	4720(9)	5676(17)	1901(5)	78(5)
Cl(1)	7799(7)	4066(15)	1078(4)	155(6)
Cl(2)	7237(9)	5620(18)	612(5)	185(7)
C(510)	7713(13)	4996(27)	658(7)	48(7)

Table S-XXI. Bond lengths [A] and angles [deg] for 5.

Hf(1)-O(1)	1.979(11)
Hf(1)-N(4)	2.181(14)
Hf(1)-N(1)	2.25(2)
Hf(1)-N(3)	2.248(14)
Hf(1)-N(2)	2.30(2)
Hf(1)-C(23)	2.31(2)
Hf(1)-N(6)	2.438(13)
O(1)-C(24)	1.43(2)
N(1)-C(1)	1.34(2)
N(1)-C(22)	1.44(3)
N(2)-C(7)	1.34(2)
N(2)-C(8)	1.46(2)
N(3)-C(11)	1.34(2)
N(3)-C(10)	1.51(2)
N(4)-C(17)	1.37(2)
N(4)-C(18)	1.46(2)
N(5)-C(23)	1.15(2)
N(6)-C(39)	1.28(2)
N(6)-C(47)	1.46(2)
C(1)-C(2)	1.37(2)
C(1)-C(7)	1.45(2)
C(2)-C(3)	1.39(3)
C(3)-C(4)	1.26(3)
C(4)-C(5)	1.32(3)
C(5)-C(6)	1.40(4)
C(6)-C(7)	1.39(3)
C(8)-C(9)	1.76(4)
C(9)-C(10)	1.70(4)
C(11)-C(17)	1.45(3)
C(11)-C(12)	1.47(2)
C(12)-C(13)	1.35(3)
C(13)-C(14)	1.23(3)
C(14)-C(15)	1.32(4)
C(15)-C(16)	1.59(4)
C(16)-C(17)	1.42(3)
C(18)-C(19)	1.52(3)
C(19)-C(20)	1.44(4)
C(20)-C(21)	1.51(5)
C(21)-C(22)	1.38(5)
C(24)-C(39)	1.47(2)
C(24)-C(32)	1.58(2)
C(24)-C(25)	1.58(2)
C(25)-C(26)	1.50(2)
C(26)-C(31)	1.37(2)
C(26)-C(27)	1.40(2)
C(27)-C(28)	1.35(2)
C(28)-C(29)	1.36(3)
C(29)-C(30)	1.42(2)
C(30)-C(31)	1.37(2)
C(32)-C(33)	1.51(2)
C(33)-C(38)	1.39(3)
C(33)-C(34)	1.40(2)
C(34)-C(35)	1.38(3)
C(35)-C(36)	1.39(3)
C(36)-C(37)	1.39(2)
C(37)-C(38)	1.36(2)
C(39)-C(40)	1.53(2)
C(40)-C(41)	1.57(2)
C(41)-C(42)	1.36(2)
C(41)-C(46)	1.39(2)
C(42)-C(43)	1.39(2)
C(43)-C(44)	1.40(3)

C(44)-C(45)	1.44(3)
C(45)-C(46)	1.38(2)
C(47)-C(48)	1.51(2)
C(48)-C(49)	1.38(2)
C(48)-C(53)	1.39(2)
C(49)-C(50)	1.40(2)
C(50)-C(51)	1.37(2)
C(51)-C(52)	1.44(3)
C(52)-C(53)	1.36(2)
Cl(1)-C(510)	1.96(3)
Cl(2)-C(510)	1.24(3)

O(1)-Hf(1)-N(4)	104.6(5)
O(1)-Hf(1)-N(1)	80.1(5)
N(4)-Hf(1)-N(1)	85.4(5)
O(1)-Hf(1)-N(3)	153.5(5)
N(4)-Hf(1)-N(3)	69.3(5)
N(1)-Hf(1)-N(3)	123.5(6)
O(1)-Hf(1)-N(2)	121.4(5)
N(4)-Hf(1)-N(2)	120.1(5)
N(1)-Hf(1)-N(2)	68.8(5)
N(3)-Hf(1)-N(2)	81.6(6)
O(1)-Hf(1)-C(23)	86.8(5)
N(4)-Hf(1)-C(23)	145.3(5)
N(1)-Hf(1)-C(23)	129.2(5)
N(3)-Hf(1)-C(23)	85.9(5)
N(2)-Hf(1)-C(23)	77.4(5)
O(1)-Hf(1)-N(6)	68.9(4)
N(4)-Hf(1)-N(6)	82.1(5)
N(1)-Hf(1)-N(6)	142.2(5)
N(3)-Hf(1)-N(6)	84.6(5)
N(2)-Hf(1)-N(6)	146.8(5)
C(23)-Hf(1)-N(6)	71.5(5)
C(24)-O(1)-Hf(1)	131.0(9)
C(1)-N(1)-C(22)	120(2)
C(1)-N(1)-Hf(1)	122.9(12)
C(22)-N(1)-Hf(1)	116.3(14)
C(7)-N(2)-C(8)	120(2)
C(7)-N(2)-Hf(1)	118.4(13)
C(8)-N(2)-Hf(1)	121.2(13)
C(11)-N(3)-C(10)	116(2)
C(11)-N(3)-Hf(1)	118.9(12)
C(10)-N(3)-Hf(1)	123.1(12)
C(17)-N(4)-C(18)	117(2)
C(17)-N(4)-Hf(1)	123.4(12)
C(18)-N(4)-Hf(1)	119.0(11)
C(39)-N(6)-C(47)	120.3(14)
C(39)-N(6)-Hf(1)	112.4(10)
C(47)-N(6)-Hf(1)	125.9(10)
N(1)-C(1)-C(2)	121(2)
N(1)-C(1)-C(7)	112(2)
C(2)-C(1)-C(7)	126(2)
C(1)-C(2)-C(3)	131(3)
C(4)-C(3)-C(2)	131(3)
C(3)-C(4)-C(5)	127(3)
C(4)-C(5)-C(6)	128(3)
C(7)-C(6)-C(5)	134(3)
N(2)-C(7)-C(6)	121(2)
N(2)-C(7)-C(1)	117(2)
C(6)-C(7)-C(1)	121(2)
N(2)-C(8)-C(9)	114(2)
C(10)-C(9)-C(8)	105(2)
N(3)-C(10)-C(9)	107(2)
N(3)-C(11)-C(17)	115(2)
N(3)-C(11)-C(12)	124(2)
C(17)-C(11)-C(12)	121(2)

C(13)-C(12)-C(11)	135(2)
C(14)-C(13)-C(12)	133(3)
C(13)-C(14)-C(15)	126(4)
C(14)-C(15)-C(16)	131(3)
C(17)-C(16)-C(15)	124(2)
N(4)-C(17)-C(16)	121(2)
N(4)-C(17)-C(11)	111(2)
C(16)-C(17)-C(11)	127(2)
N(4)-C(18)-C(19)	123(2)
C(20)-C(19)-C(18)	105(3)
C(19)-C(20)-C(21)	125(4)
C(22)-C(21)-C(20)	105(3)
C(21)-C(22)-N(1)	117(3)
N(5)-C(23)-Hf(1)	177.0(14)
O(1)-C(24)-C(39)	105.9(13)
O(1)-C(24)-C(32)	110.7(13)
C(39)-C(24)-C(32)	114.8(12)
O(1)-C(24)-C(25)	109.2(12)
C(39)-C(24)-C(25)	109.5(13)
C(32)-C(24)-C(25)	106.6(13)
C(26)-C(25)-C(24)	115.0(13)
C(31)-C(26)-C(27)	117(2)
C(31)-C(26)-C(25)	121(2)
C(27)-C(26)-C(25)	122(2)
C(28)-C(27)-C(26)	121(2)
C(27)-C(28)-C(29)	123(2)
C(28)-C(29)-C(30)	116(2)
C(31)-C(30)-C(29)	121(2)
C(26)-C(31)-C(30)	122(2)
C(33)-C(32)-C(24)	117.5(14)
C(38)-C(33)-C(34)	118(2)
C(38)-C(33)-C(32)	122(2)
C(34)-C(33)-C(32)	120(2)
C(35)-C(34)-C(33)	121(2)
C(34)-C(35)-C(36)	119(2)
C(37)-C(36)-C(35)	121(2)
C(38)-C(37)-C(36)	119(2)
C(37)-C(38)-C(33)	122(2)
N(6)-C(39)-C(24)	120(2)
N(6)-C(39)-C(40)	124.2(14)
C(24)-C(39)-C(40)	115(2)
C(39)-C(40)-C(41)	117.3(13)
C(42)-C(41)-C(46)	118(2)
C(42)-C(41)-C(40)	124(2)
C(46)-C(41)-C(40)	117(2)
C(41)-C(42)-C(43)	121(2)
C(44)-C(43)-C(42)	120(2)
C(43)-C(44)-C(45)	119(2)
C(46)-C(45)-C(44)	117(2)
C(45)-C(46)-C(41)	124(2)
N(6)-C(47)-C(48)	111.2(13)
C(49)-C(48)-C(53)	116(2)
C(49)-C(48)-C(47)	120(2)
C(53)-C(48)-C(47)	123(2)
C(48)-C(49)-C(50)	122(2)
C(51)-C(50)-C(49)	119(2)
C(50)-C(51)-C(52)	120(2)
C(53)-C(52)-C(51)	117(2)
C(52)-C(53)-C(48)	125(2)
Cl(2)-C(510)-Cl(1)	119(2)

Symmetry transformations used to generate equivalent atoms:

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Table S-XII. Anisotropic displacement parameters (A^2 x 10^3) for 5. The anisotropic displacement factor exponent takes the form: -2 pi^2 [h^2 a*^2 U11 + ... + 2 h k a* b* U12]

	U11	U22	U33	U23	U13	U12
Hf(1)	73(1)	57(1)	55(1)	3(1)	4(1)	-6(1)
O(1)	71(8)	70(7)	59(7)	17(6)	10(6)	14(6)
N(1)	115(12)	46(9)	73(10)	8(8)	9(9)	-4(8)
N(2)	86(11)	84(11)	86(12)	-13(10)	12(9)	-8(9)
N(3)	73(10)	87(11)	62(9)	-22(8)	11(7)	7(8)
N(4)	82(10)	67(9)	49(8)	-19(7)	5(8)	-6(7)
N(5)	87(11)	74(10)	58(9)	-4(8)	4(8)	-18(9)
N(6)	60(9)	63(9)	55(9)	-3(7)	-2(7)	-6(7)
C(1)	61(11)	47(10)	79(12)	-4(9)	0(9)	10(8)
C(2)	195(28)	145(23)	83(16)	-13(16)	77(18)	-19(21)
C(3)	131(22)	110(21)	155(25)	36(19)	59(18)	-36(17)
C(4)	148(23)	92(18)	120(22)	42(17)	49(18)	-2(18)
C(5)	434(67)	223(41)	165(32)	-77(31)	151(39)	-232(45)
C(6)	566(77)	228(39)	141(27)	-60(27)	148(39)	-312(50)
C(7)	121(17)	73(13)	66(13)	9(11)	10(12)	-5(12)
C(8)	141(24)	130(21)	99(17)	-59(16)	55(16)	-41(18)
C(9)	138(28)	192(33)	363(56)	-218(39)	17(34)	0(26)
C(10)	86(15)	101(16)	121(18)	-52(15)	-28(13)	26(13)
C(11)	79(13)	88(14)	65(12)	-7(11)	19(10)	25(11)
C(12)	93(16)	110(18)	134(19)	-42(15)	-64(15)	48(14)
C(13)	166(28)	170(29)	136(24)	-59(23)	-93(21)	67(24)
C(14)	112(24)	149(32)	173(33)	-26(25)	-3(21)	10(22)
C(15)	123(25)	159(31)	168(30)	8(23)	47(21)	90(23)
C(16)	169(28)	101(20)	183(29)	-77(20)	32(23)	31(19)
C(17)	98(15)	83(14)	64(12)	-6(11)	-1(11)	17(12)
C(18)	80(13)	83(13)	72(12)	-22(11)	25(10)	-3(11)
C(19)	226(34)	130(25)	143(24)	-70(21)	89(24)	-61(23)
C(20)	400(62)	62(18)	176(35)	6(21)	-15(39)	-44(28)
C(21)	291(57)	276(55)	131(33)	25(37)	15(35)	-124(45)
C(22)	201(31)	87(16)	87(18)	-34(14)	54(18)	-24(17)
C(23)	73(11)	43(9)	45(9)	-3(8)	7(8)	-11(8)
C(24)	67(11)	38(8)	68(11)	6(8)	-5(8)	2(8)
C(25)	79(12)	66(11)	45(9)	4(8)	2(8)	-18(9)
C(26)	71(12)	72(12)	50(10)	15(9)	2(9)	-1(9)
C(27)	111(15)	54(10)	45(10)	-12(9)	-14(10)	5(10)
C(28)	91(15)	96(14)	54(11)	-9(12)	-9(10)	9(12)
C(29)	88(14)	92(14)	54(11)	15(11)	13(10)	24(12)
C(30)	97(15)	57(11)	72(13)	21(10)	-6(11)	3(10)
C(31)	81(12)	34(8)	58(10)	13(8)	-8(8)	-11(8)
C(32)	121(16)	66(11)	38(10)	14(9)	-11(9)	-1(11)
C(33)	85(12)	38(9)	71(12)	-8(9)	-39(10)	-3(8)
C(34)	113(16)	49(11)	82(13)	13(10)	-34(11)	-3(10)
C(35)	137(20)	75(15)	95(17)	-14(13)	-9(15)	-29(14)
C(36)	142(20)	47(11)	89(15)	-4(12)	-27(14)	-20(12)
C(37)	122(17)	71(13)	63(12)	-6(11)	-16(11)	8(12)
C(38)	142(19)	38(10)	89(15)	-25(10)	-5(14)	-15(11)
C(39)	120(16)	18(8)	53(10)	23(7)	-7(10)	-5(9)
C(40)	107(15)	67(11)	49(10)	-14(9)	15(10)	-26(10)
C(41)	70(12)	59(11)	58(11)	9(9)	-6(9)	-22(9)
C(42)	90(13)	69(13)	66(12)	-2(10)	11(10)	5(11)
C(43)	87(14)	61(12)	88(14)	22(11)	16(11)	-6(11)
C(44)	125(20)	82(15)	65(13)	25(12)	24(12)	7(13)
C(45)	90(14)	100(16)	68(13)	24(12)	26(11)	-2(13)
C(46)	110(17)	52(11)	67(12)	3(9)	-4(11)	14(11)
C(47)	79(12)	55(11)	61(11)	-10(9)	-7(9)	3(9)
C(48)	98(13)	56(11)	33(9)	17(8)	2(8)	5(9)

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C(49)	87(13)	48(11)	83(14)	4(9)	3(11)	-6(9)
C(50)	91(14)	82(14)	58(11)	21(10)	-6(9)	17(11)
C(51)	131(18)	71(13)	73(14)	25(11)	1(12)	48(13)
C(52)	144(22)	89(15)	83(16)	-6(12)	-16(13)	54(14)
C(53)	79(12)	65(12)	88(14)	2(11)	-22(11)	15(10)

Table S-XXIII. Hydrogen coordinates (x 10^4) and isotropic displacement parameters (A^2 x 10^3) for 5.

	x	y	z	U(eq)
H(2A)	5705(13)	9536(26)	139(6)	169
H(3A)	6184(12)	10928(26)	-152(7)	158
H(4A)	6622(12)	12547(26)	1(7)	144
H(5A)	6368(21)	13627(37)	437(9)	329
H(6A)	6064(23)	12877(35)	916(8)	374
H(8A)	5166(13)	12257(23)	1485(6)	148
H(8B)	5734(13)	12773(23)	1285(6)	148
H(9A)	6439(15)	11198(33)	1619(11)	277
H(9B)	6162(15)	12228(33)	1858(11)	277
H(10A)	5873(10)	10192(20)	2127(6)	123
H(10B)	5250(10)	10722(20)	1981(6)	123
H(12A)	6554(10)	9256(22)	2086(6)	135
H(13A)	7336(14)	8185(34)	2178(7)	189
H(14A)	7561(15)	6414(35)	2056(9)	174
H(15A)	7001(15)	5123(32)	1784(9)	180
H(16A)	6149(14)	5529(25)	1409(8)	181
H(18A)	4991(9)	6723(17)	939(4)	94
H(18B)	5251(9)	5781(17)	1200(4)	94
H(19A)	6176(16)	5644(28)	907(7)	199
H(19B)	5621(16)	5358(28)	662(7)	199
H(20A)	6364(21)	6871(25)	494(10)	255
H(20B)	6061(21)	7736(25)	760(10)	255
H(21A)	5802(22)	8201(49)	192(9)	279
H(21B)	5391(22)	6986(49)	192(9)	279
H(22A)	4893(14)	7665(23)	645(6)	150
H(22B)	4815(14)	8579(23)	343(6)	150
H(25A)	2740(8)	9047(15)	1215(4)	76
H(25B)	3260(8)	10003(15)	1306(4)	76
H(27A)	2325(9)	8988(16)	637(4)	84
H(28A)	2178(10)	10007(20)	153(4)	97
H(29A)	2806(9)	11614(19)	-18(4)	94
H(30A)	3572(9)	12294(16)	366(4)	91
H(31A)	3723(8)	11264(13)	859(4)	69
H(32A)	2948(9)	7017(16)	992(4)	90
H(32B)	3314(9)	7565(16)	688(4)	90
H(34A)	3481(9)	5320(17)	1338(5)	97
H(35A)	4036(11)	3480(20)	1301(6)	123
H(36A)	4538(11)	2990(18)	803(6)	111
H(37A)	4562(10)	4419(18)	366(5)	102
H(38A)	4088(11)	6309(16)	428(5)	108
H(40A)	3347(8)	6762(16)	1842(4)	89
H(40B)	2934(8)	6941(16)	1525(4)	89
H(42A)	3418(9)	9558(17)	2027(4)	90
H(43A)	2769(10)	10808(17)	2342(5)	94
H(44A)	1720(11)	10354(20)	2379(5)	109
H(45A)	1317(10)	8586(21)	2084(5)	104
H(46A)	1998(10)	7405(16)	1775(4)	92
H(47A)	3979(8)	7785(15)	2093(4)	78
H(47B)	4554(8)	8678(15)	2082(4)	78
H(49A)	5270(8)	7654(16)	2424(5)	87
H(50A)	5896(9)	5965(18)	2562(4)	92
H(51A)	5736(10)	4068(19)	2306(5)	110
H(52A)	4979(10)	3880(22)	1874(6)	126
H(53A)	4425(9)	5606(17)	1733(5)	93

Table S-XXIV. Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters (A^2 x 10^3) for 6. U(eq) is defined as one third of the trace of the orthogonalized Uij tensor.

	x	y	z	U(eq)
Hf(1)	6278(1)	8695(1)	7175(1)	26(1)
O(1)	5872(2)	10023(7)	6810(3)	29(2)
O(2)	6496(2)	10632(7)	7389(3)	34(2)
N(1)	6860(2)	8194(9)	6995(4)	31(2)
N(2)	6264(2)	7157(9)	6381(4)	32(2)
N(3)	5828(2)	7254(9)	7414(4)	30(2)
N(4)	6370(2)	8173(9)	8193(4)	29(2)
C(1)	6924(3)	7339(12)	6533(5)	31(3)
C(2)	7288(3)	7080(13)	6368(6)	44(3)
C(3)	7421(4)	6309(15)	5888(6)	54(3)
C(4)	7229(4)	5435(15)	5442(6)	54(4)
C(5)	6856(4)	5171(14)	5365(6)	50(3)
C(6)	6575(3)	5701(12)	5680(5)	37(3)
C(7)	6580(3)	6694(11)	6165(5)	32(3)
C(8)	5911(3)	6611(12)	6035(5)	37(3)
C(9)	5561(3)	7249(12)	6280(5)	34(3)
C(10)	5525(3)	6736(11)	6938(5)	31(3)
C(11)	5814(3)	6825(11)	8015(6)	34(3)
C(12)	5549(3)	5861(12)	8176(6)	42(3)
C(13)	5478(3)	5301(14)	8753(6)	52(4)
C(14)	5632(3)	5655(14)	9356(6)	49(4)
C(15)	5931(3)	6568(13)	9517(5)	44(3)
C(16)	6138(3)	7329(12)	9132(5)	35(3)
C(17)	6120(3)	7427(11)	8481(5)	30(3)
C(18)	6698(3)	8808(13)	8596(5)	43(3)
C(19)	7008(3)	7718(12)	8796(5)	37(3)
C(20)	7175(3)	7002(13)	8261(6)	44(3)
C(21)	7399(3)	7946(12)	7872(6)	41(3)
C(22)	7185(3)	8900(12)	7372(5)	34(3)
C(23)	5930(3)	11495(13)	6854(5)	40(3)
C(24)	5548(3)	12258(12)	6908(6)	41(3)
C(25)	5228(3)	11695(14)	6460(6)	44(3)
C(26)	5055(4)	10460(17)	6618(8)	71(5)
C(27)	4764(5)	9779(22)	6231(11)	95(6)
C(28)	4666(6)	10391(32)	5692(10)	124(11)
C(29)	4799(6)	11576(26)	5458(8)	101(9)
C(30)	5111(5)	12320(19)	5878(7)	81(5)
C(31)	6056(3)	12087(13)	6231(5)	42(3)
C(32)	6341(3)	11260(13)	5918(5)	33(3)
C(33)	6225(3)	10096(13)	5552(5)	40(3)
C(34)	6471(4)	9326(15)	5221(6)	54(4)
C(35)	6839(4)	9739(16)	5263(6)	54(4)
C(36)	6956(4)	10893(18)	5625(6)	64(5)
C(37)	6712(3)	11637(14)	5944(6)	51(4)
C(38)	6233(3)	11739(11)	7442(5)	28(3)
C(39)	6050(3)	11480(12)	8039(5)	37(3)
C(40)	6133(3)	12243(13)	8578(6)	45(3)
C(41)	6421(4)	13395(12)	8670(6)	52(4)
C(42)	6662(4)	13345(14)	8128(7)	58(4)
C(43)	6425(4)	13203(13)	7494(6)	50(3)