SUPPLEMENTARY INFORMATION

Cooperative Borane Activation by Tuning Hemilability of Different Os-κ^2-*N,Se*-Chelated Complexes

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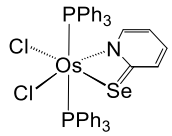
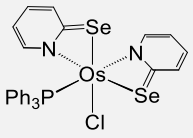
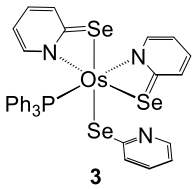
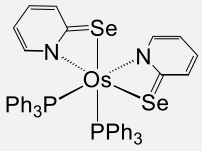
I Supplementary Data

Table S1. g-values of **1-3** determined by EPR spectroscopy (X-band frozen glass EPR spectra) and redox Potentials of **1-4** determined by Cyclic Voltammetry.

Complexes	g-value	Redox Potentials		
		E^1,V	E^2,V	E^3,V
 **1**	g_z = 2.724 g_x = 2.227 g_y = 1.2-1.7	-0.892 (red) -0.562 (ox)	0.547(red) 0.927 (ox)	-
 **2**	g_z= 2.851 g_y= 2.052, g_x =1.4–1.7	-1.089 (red) -0.611 (ox)	0.371 (red) 0.871 (ox)	-
 **3**	g_z = 2.324 g_y= 2.200 g_x =1.928	-1.014 (red) -0.883 (ox)	0.085 (red) 0.211 (ox)	1.047 (red) 1.200 (ox)
 **4**	-	0.043 (red) 0.319 (ox)	0.960 (red) 1.368 (ox)	-

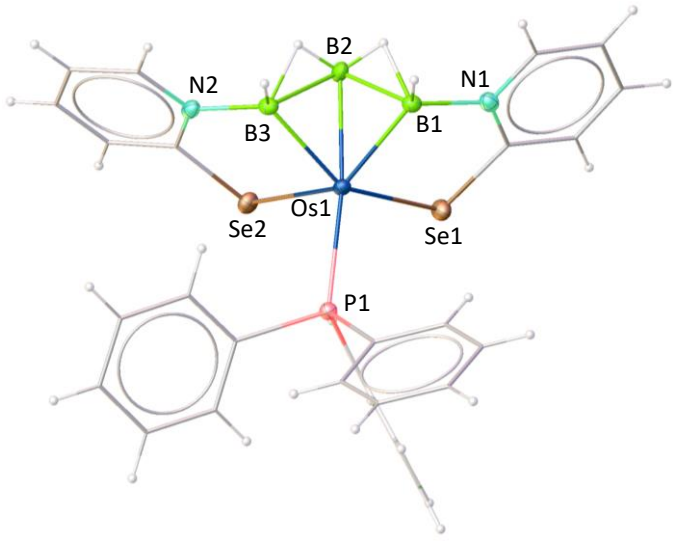


Figure S1. Molecular structure and labelling diagram of **6**. Selected bond lengths (Å) and bond angles (˚) of **6**: B1-B2 1.878(8), B2-B3 1.916(8) Os1-B1 2.133(7), B2-Os1 2.240(6), B3-Os1 2.157(6), B1-B2-B3 93.7(4).

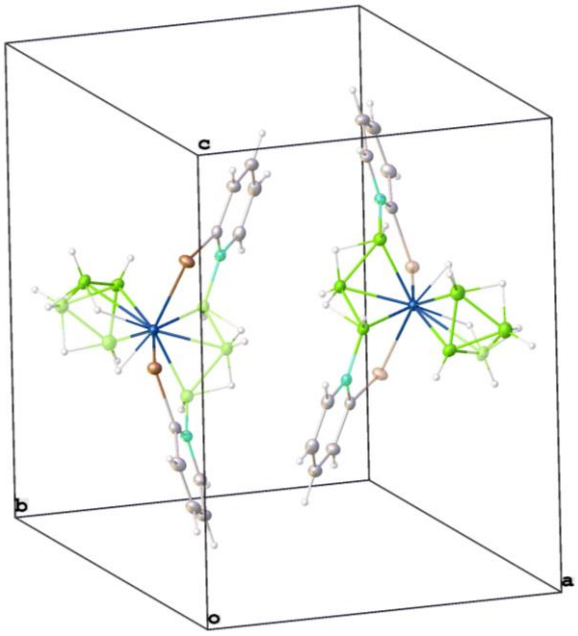


Figure S2. Molecular packing of isomers of **8** in unit cell along b-axis.

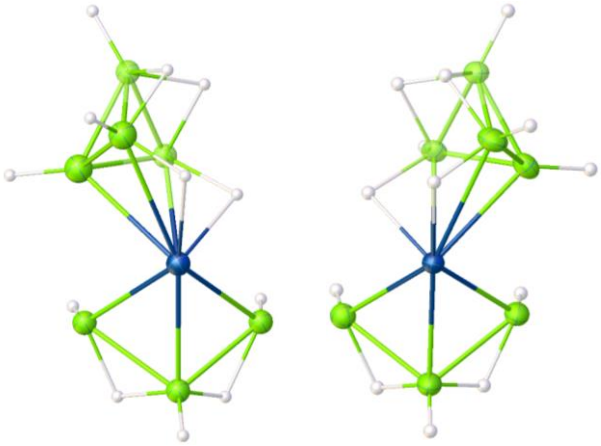


Figure S3. Two isomeric forms of **8**.

II Experimental Details

General Procedures and Instrumentation

All manipulations were carried out using standard Schlenk line and glove box techniques under inert atmosphere of dry argon. Solvents such as toluene, hexane and THF were distilled through Na/benzophenoneketyl and dichloromethane was dried over calcium hydride prior to use under argon. Chloroform-d was degassed by three freeze-thaw cycles, dried over calcium hydride for 12 h, and stored over 4 Å molecular sieves in a Young's ampoule under argon. $[Os(PPh_3)_3Cl_2]$, $[(3,5\text{-}(CF_3)_2C_6H_3)BH_2{\cdot}SMe_2]$ and $[(3,5\text{-}(CF_3)_2C_6H_3)_2BH{\cdot}SMe_2]$ were synthesized according to the reported literature procedure[1-3] and $[BH_3{\cdot}SMe_2]$ were obtained commercially (Sigma Aldrich) and used as received. The preparative TLC was performed with Merck 105554 TLC silica gel 60 F254 and thickness of layer 250 µm on aluminum sheets with 20x20 cm size. The external reference for the ^{11}B NMR spectroscopy, $[Bu_4N][(B_3H_8)]$ was synthesized according to the literature method.[4] The ^{1}H, $^{11}B\{^{1}H\}$, $^{19}F\{^{1}H\}$, $^{31}P\{^{1}H\}$, $^{13}C\{^{1}H\}$, and ^{77}Se NMR spectra were recorded on Bruker 400 and 500 MHz instruments. The residual solvent protons were used as reference (δ, ppm, C_6D_6; 7.16, $CDCl_3$; 7.26, C_7D_8; 7.09 ppm), while a sealed tube containing $[Bu_4N][(B_3H_8)]$ in C_6D_6 (δ_B, ppm, -30.07) was used as an external reference for $^{11}B\{^{1}H\}$ NMR spectra. ^{1}H decoupled $^{11}B\{^{1}H\}$ spectra of all complexes were processed with a backward linear prediction algorithm to remove the broad $^{11}B\{^{1}H\}$ background signal of the NMR tube.[5] ^{1}H NMR Evans measurements were performed at 25.8 °C on a 500 MHz spectrometer. A sealed capillary containing 2.0 mM $^{t}BuOH$ in $CDCl_3$ was placed inside an NMR tube containing only the sample solution (**1**: 12.127 mM, **2**: 12.453 mM in $CDCl_3$; no $^{t}BuOH$ added to the bulk), note that diamagnetic corrections were applied using Pascal constants.[6] EPR measurements were conducted using a JEOL model JES FA200 X-band (9.5 GHz) electron spin resonance spectrometer. Samples of the paramagnetic complexes were prepared in a dichloromethane (for **1** and **2**) or toluene (for **3**) solution, loaded into quartz capillary tubes, and analyzed at 77 K. Mass spectra were carried out using Qtof Micro YA263 HRMS instrument and Bruker MicroTOF-II mass spectrometer in ESI ionization mode. All cyclic voltammetry (CV) experiments were performed at room temperature using a Metrohm Autolab PGSTAT 204 potentiostat. A conventional three-electrode setup was employed, consisting of a 1 mm diameter glassy carbon disc as the working electrode, an Ag/AgCl reference electrode (3.0 M KCl, connected by a Vycor tip), and a platinum wire counter electrode. All potentials are referenced to the ferrocenium/ferrocene ($FeCp_2^{+/0}$) couple for measurements conducted in organic solvent. Cyclic voltammograms of complexes **1**, **2**, and **3** were recorded in dichloromethane containing 0.1 M $(nBu_4N)PF_6$) at a scan rate of 100 mV/s. UV-vis absorption spectra were gained from Jasco V-650 spectrometer. Infrared spectra were obtained on a Jasco FT/IR–1400 spectrometer.

II.1 Synthesis and Characterizations

Syntheses of 1, 2, 3 and 4: In a flame-dried Schlenk tube, $[Os(PPh_3)_3Cl_2]$, (0.600 g, 0.573 mmol) and K[L] (L = 2-selanopyridine, (0.562 g, 2.863 mmol) was taken in 1:5 stoichiometric ratio. After adding 30 mL of THF the reaction was thermalized for 4 h at 60 °C during which the colour of the mixture changed from green to brown. The solvent was removed using vacuum and the residue was extracted in hexane/CH_2Cl_2 (40:60 v/v) and passed through celite. The products were further isolated by performing chromatographic work-up using silica-gel TLC plates by eluting in 100% toluene. This enabled us to isolate pale green **1** (0.90 g, 15%, R_f= 0.6), blueish green **2** (0.210 g, 35%, R_f= 0.4), dark green **3** (0.110 g, 18%, R_f= 0.1) and orange **4** (0.120 g, 20%, R_f= 0.8).

Modified Syntheses of 1, 2 and 3: In a flame-dried Schlenk tube, $[Os(PPh_3)_3Cl_2]$ (0.600 g, 0.573 mmol) and K[L] (0.562 g, 2.863 mmol) were taken in a 1:5 molar ratio. To this, 30 mL of THF was added at –78 °C, and the reaction mixture was allowed to warm to room temperature and stirred for 12 h under an open-air atmosphere. The solution gradually changed to a greenish-brown color, affording complexes **1** (0.108 g, 18%), **2** (0.240 g, 40%), and **3** (0.110 g, 18%), along with trace amounts of **4**. Although the Os(III) complexes (**1**-**3**) formed in comparatively higher yields than the Os(II) complex **4**, the overall product yield was modest, with substantial decomposition observed under aerobic conditions relative to the reaction carried out under inert atmosphere.

Modified Syntheses of 1 and 2: In a flame-dried Schlenk tube, $[Os(PPh_3)_3Cl_2]$, (0.600 g, 0.573 mmol) and K[L] (0.562 g, 2.863 mmol) was taken in 1:5 ratio. 30 mL of CCl_4 solvent was added and it was further thermalized for 12 h at 60 °C. The colour of the reaction mixture changed from green to blueish green yielding **1** and **2** with an enhanced yield of 30% (0.180 g) and 65% (0.390 g) respectively with a trace amount of **3** and **4**.

Modified Syntheses of 3 and 4: In a flame-dried Schlenk tube, $[Os(PPh_3)_3Cl_2]$, (0.600 g, 0.573 mmol) and K[L] (0.562 g, 2.863 mmol) was taken in 1:5 stoichiometric ratio. 30 mL of THF solvent was added to it at -78 °C, after attaining room temperature the reaction was stirred for 4 h under inert atmosphere. The colour of the reaction mixture changed from green to orange yielding **3** and **4** with an enhanced yield of 30% (0.180 g) and 61% (0.365 g) respectively with a trace amount of **1** and **2**.

1: MS (ESI^+): *m/z* calculated for $[M+NH_4]^+$: 962.0648, found: 962.0597; ^1H NMR (500 MHz, $CDCl_3$, 22 °C): δ = 17.20 (br), 13.73 (br), 7.32 (br), 6.80 (br), 5.81 (br), 2.05 (br), -0.93 (br), -1.75 (br), -8.33 (br), -10.96 (br), -16.61 (br) ppm.

2: MS (ESI^+): *m/z* calculated for $[M]^+$: 802.9227, found: 802.9211; ^1H NMR (500 MHz, $CDCl_3$, 22 °C): δ = 26.54 (br), 10.41 (br), 6.82 (br), 6.66 (br), 5.65 (br), 5.48 (br), 3.42 (br), 2.53 (br), -2.50 (br), -13.33 (br), -21.34 (br) ppm.

3: MS (ESI^+): *m/z* calculated for $[M]^+$: 923.9056, found: 923.9076; ^1H NMR (500 MHz, $CDCl_3$, 22 °C): δ = 24.42 (br), 17.71 (br), 14.66 (br), 14.10 (br), 13.42 (br), 11.96 (br), 6.31 (br), 5.92 (br), -1.26 (br), -4.62 (br), -6.63 (br), -11.13 (br), -12.91 (br), -17.97 (br) ppm.

4: MS (ESI^+): *m/z* calculated for $[M]^+$: 1030.0461, found: 1030.0479; ^1H NMR (500 MHz, $CDCl_3$, 22 °C): δ = 7.93 (d, J = 5.5 Hz, 2H, L), 7.19 (t, J = 8.0 Hz, 12H, $Ar_{(ph)}$), 7.07 (t, J = 7.1 Hz, 6H, $Ar_{(ph)}$), 6.95 (t, J = 7.4 Hz, 12H, $Ar_{(ph)}$), 6.73 (t, J = 7.5 Hz, 2H, L), 6.36 (d, J = 7.9 Hz, 2H, L), 6.19 (t, J = 6.4 Hz, 2H, $Ar_{(ph)}$) ppm; $^{31}P\{^1H\}$ NMR (202 MHz, $CDCl_3$, 22 °C): δ = -6.3 (s, 2P) ppm; $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$, 22 °C): δ = 176.6 (s, C=Se), 148.1 (s, C-N), 138.9, 138.8, 138.5, 138.4, 134.5, 133.5, 130.3, 128.5, 127.2, 117.3 (s, C=C, $Ar_{(Ph/L)}$) ppm.

Syntheses of 5 and **6**: In a flame-dried Schlenk tube, $[OsCl(PPh_3)(\kappa^2\text{-}N,Se\text{-}L)_2]$, **2** (100 mg, 0.125 mmol) in 15 mL dry toluene was treated with stoichiometric amount of $[BH_3{\cdot}SMe_2]$ (0.1 mL, 0.498 mmol) was thermalized for 4 h at 60 °C. During course of the reaction the colour of the reaction changed from blueish green to orange. The volatile components were removed under vacuum and the remaining residue was extracted into hexane/CH_2Cl_2 (80:20 v/v) and passed through Celite. After removal of the solvent, the residue was subjected to chromatographic work-up by using TLC plates. Elution with

hexane/CH_2Cl_2 (60:40 v/v) mixture yielded red solid **5** (15 mg, 15%, Rf= 0.7) and orange solid **6** (35 mg, 35%, Rf= 0.3).

5: MS (ESI^+): *m/z* calculated for $[M\text{-}2H]^+$: 818.0568, found: 818.0868; $^{11}B\{^1H\}$ NMR (160 MHz, C_6D_6, 22 °C): δ = 59.9 (br, 1B), 14.2 (br, 1B), 4.6 (br, 1B), -15.2(br, 1B) ppm; ^1H NMR (500 MHz, C_6D_6, 22 °C): δ = 8.05 (d, J = 5.8 Hz, 1H, L), 7.82 (d, J = 8.2 Hz, 1H, L), 7.64 (d, J = 8.8 Hz, 1H, L), 7.62 (d, J = 8.6 Hz, 1H, L), 7.71-7.67, 7.58-6.91 (m, 15H, $Ar_{(Ph)}$), 6.25 (t, J = 7.2 Hz, 1H, L), 6.11 (t, J = 7.5 Hz, 1H, L), 5.80 (t, J = 6.5 Hz, 1H, L), 5.46 (t, J = 6.7 Hz, 1H, L), 4.47(br, B-H_t), 3.39(br, B-H_t), -7.65 (br, Os-*H*-B), -11.61 (d, 1H, Os-*H*-B), -11.66 (d, 1H, Os-*H*-B) ppm; $^{31}P\{^1H\}$ NMR (202 MHz, C_6D_6, 22 °C): δ = 27.9 (s, 1P) ppm; $^{13}C\{^1H\}$ NMR (100 MHz, C_6D_6, 22 °C): δ = 196.0 (s, C=Se), 179.0 (s, C-Se), 172.5, 159.1 (s, C-N), 149.7, 145.2, 144.4, 138.7, 137.0, 136.6, 134.8, 133.5, 132.4, 131.5, 130.6, 130.1, 128.7, 127.8, 125.6, 121.0, 116.5 (s, C=C, $Ar_{(Ph/L)}$) ppm ; IR (dicholoromethane, cm^{-1}): $\tilde{\nu}$ = 2507 (B-H), 2452 (B-H-B, Os-H-B), 1436 (C=Se).

6: MS (ESI^+): *m/z* calculated for $[M\text{-}H]^+$: 805.0311, found: 805.0319; $^{11}B\{^1H\}$ NMR (160 MHz, $CDCl_3$, 22 °C): δ = 17.0 (br, 1B), -39.0 (br, 2B) ppm; ^1H NMR (500 MHz, $CDCl_3$, 22 °C): δ = 8.40 (d, J = 5.8 Hz, 2H, L), 7.70 (d, J = 8.1 Hz, 2H, L), 7.34-7.21 (m, 15H, $Ar_{(ph)}$), 7.13 (t, J = 7.1 Hz, 2H, L), 6.84 (t, J = 6.3 Hz, 2H, L), 4.10 (br, 3H, B-H_t), −1.41 (br, 2H, B-*H*-B), −13.40 (d, 2H, Os-*H*) ppm; $^{31}P\{^1H\}$ NMR (202 MHz, $CDCl_3$, 22 °C): δ = 16.1 (s, 1P) ppm; ^{77}Se NMR (95 MHz, $CDCl_3$, 22 °C): δ = 234.8 (s, 2Se) ppm; $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$, 22 °C): δ = 175.8 (s, C=Se), 147.0 (s, C-N), 137.2, 136.8, 133.8, 133.7, 131.7, 129.4, 127.7, 127.6, 116.3 (s, C=C, $Ar_{(Ph/L)}$) ppm ; IR (dicholoromethane, cm^{-1}): $\tilde{\nu}$ = 2499 (B-H), 2431 (B-H-B), 1534 (C=Se).

Syntheses of 7: In a flame-dried Schlenk tube, $[OsCl(PPh_3)(\kappa^2\text{-}N,Se\text{-}L)_2]$ **2** (100 mg, 0.125 mmol) in 15 mL dry toluene was treated with stoichiometric amount of $[(3,5\text{-}(CF_3)_2C_6H_3)BH_2{\cdot}SMe_2]$ (71.8 mg, 0.249 mmol) and thermalized for 4 h at 60 °C. The volatile components were removed under vacuum and the remaining residue was extracted into hexane/CH_2Cl_2 (60:40 v/v) and passed through Celite. After removal of the solvent, the residue was subjected to chromatographic work-up by using TLC plates. Elution with hexane/CH_2Cl_2 (50:50 v/v) mixture yielded yellow solid **7** (12 mg, 12%, Rf= 0.4).

7: $^{11}B\{^1H\}$ NMR (160 MHz, C_6D_6, 22 °C): δ = 27.8 (br, 2B) ppm, ^1H NMR (500 MHz, C_6D_6, 22 °C): δ = 8.39-6.07 (m, 29H, H_{Ar}), -9.72 (t, 2H, Os-*H*-B) ppm; $^{31}P\{^1H\}$ NMR (202 MHz, C_6D_6, 22 °C): δ = 22.3 (s, 1P) ppm; ^{19}F NMR (470 MHz, C_6D_6, 22 °C): δ −62.2 (s, 6F) ppm; IR (n-Hexane, cm^{-1}): $\tilde{\nu}$ = 2360 (Os-H-B), 1454 (C=Se).

Syntheses of 8: In a flame-dried Schlenk tube, $[(PPh_3)(H)_2Os(\eta^5\text{-}B_3H_5(C_5H_4NSe)_2)]$ **6** (100 mg, 0.124 mmol) in 15 mL dry toluene was treated with excess $[BH_3{\cdot}SMe_2]$ (0.5 mL) and thermalized for 12 h at 90 °C. The colour of the mixture changed from orange to pale yellow. The volatile components were removed under vacuum and the remaining residue was extracted into hexane/CH_2Cl_2 (60:40 v/v) and passed through Celite. After removal of the solvent, the residue was subjected to chromatographic work-up by using TLC plates. Elution with hexane/CH_2Cl_2 (60:40 v/v) mixture yielded yellow solid **8** (30 mg, 30%, Rf = 0.5).

8: MS (ESI^+): *m/z* calculated for $[M]^+$: 593.0259, found: 593.0291; $^{11}B\{^1H\}$ NMR (160 MHz, $CDCl_3$, 22 °C): δ = 12.2 (br), 10.4 (br), 8.9 (br), -2.6(br), -7.2(br), -10.3(br), -11.5 (br), -21.3(br), 22.4 (br), -28.1(br) ppm; ^1H NMR (500 MHz, $CDCl_3$, 22 °C): δ 8.32 (t, J = 6.2 Hz, L), 7.86 (d, J = 8.4 Hz, L), 7.84 (d, J = 8.8 Hz, L), 7.63 (q, J = 5.3 Hz, L), 7.36 (t, J = 7.6 Hz, L), 7.23 (t, J = 7.1 Hz, L), 7.00 (t, J = 6.5 Hz, L), 6.87 (t, J =

6.4 Hz, L), 3.74 (br, B-H_t), 2.66 (br, B-H_t), -0.54(br, B-*H*-B), -1.29 (br, B-*H*-B), -1.44 (br, B-*H*-B), -2.79 (br, B-*H*-B) ppm, -4.19 (br, Os-*H*-B), -9.83 (br, Os-*H*-B) ppm; ^{77}Se NMR (95 MHz, $CDCl_3$, 22 °C): δ = 125.0 ppm (s, 2Se); $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$, 22 °C): δ = 174.7, 172.6 (s, C=Se), 147.2, 146.9 (s, C-N), 136.3–116.9 (s, C=C, $Ar_{(Ph/L)}$) ppm; IR (dicholoromethane, cm^{-1}): $\tilde{\nu}$ = 2541 (B-H), 2482 (B-H-B, Os-H-B), 1536 (C=Se).

Syntheses of 10: In a flame-dried Schlenk tube, $[Os(PPh_3)_2(\kappa^2\text{-}N,Se\text{-}(C_5H_4NSe))_2]$ **4** (500 mg, 0.485 mmol) in 15 mL dry toluene was treated with stoichiometric amount of $[BH_3{\cdot}SMe_2]$ (0.1 mL, 0.970 mmol) and thermalized for 4 h at 60 °C. The colour of the mixture changed from orange to red. The volatile components were removed under vacuum and the remaining residue was extracted into hexane/toluene (60:40 v/v) and passed through Celite. After removal of the solvent, the residue was subjected to chromatographic work-up by using TLC plates. Elution with hexane/CH_2Cl_2 (50:50 v/v) mixture yielded red solid **10** (20 mg, 20%, Rf = 0.6).

10: MS (ESI^+): *m/z* calculated for $[M]^+$: 978.0715, found: 978.0793; $^{11}B\{^1H\}$ NMR (160 MHz, C_6D_6, 22 °C): δ = 0.6 (br, 1B), -21.0 (br, 1B) ppm; ^{1}H NMR (500 MHz, C_6D_6, 22 °C): δ 7.58 (m, 1H, L), 7.51 (m, 6H, $Ar_{(ph)}$), 7.44 (t, J = 8.4 Hz, 7H, $Ar_{(ph)}$), 7.13 (d, J =7.4 Hz, 1H, L), 7.06-6.91 (m, 17H, $Ar_{(ph)}$), 6.05 (t, J = 7.5 Hz, 1H, L), 5.73 (t, J = 8.0 Hz, 1H, L), 2.90, 2.11 (br,B-H_t), -11.17 (br, 1H, Os-*H*-B), -14.14 (br,1H, Os-*H*-B) ppm; $^{31}P\{^1H\}$ NMR (202 MHz, C_6D_6, 22 °C): δ = 13.6 (d, 1P), 6.1 (s, 1P) ppm; $^{13}C\{^1H\}$ NMR (100 MHz, C_6D_6, 22 °C): δ = 175.9 (s, C=Se), 147.1 (s, C-N), 140.6, 140.2, 136.1, 135.7, 134.7, 134.6, 134.5, 134.4, 132.7, 132.1, 129.4, 129.2, 128.6, 127.7, 127.6, 127.5, 127.4, 125.7, 116.0(s, C=C, $Ar_{(Ph/L)}$) ppm; IR (n-hexane, cm^{-1}): $\tilde{\nu}$ = 2381 (B-H),2342 (Os-H-B) 1431 (C=Se).

Syntheses of 11: In a flame-dried Schlenk tube, $[Os(PPh_3)_2(\kappa^2\text{-}N,S\text{-}C_7H_4NS_2)_2]$ $\mathbf{4^{mbz}}$ (100 mg, 0.095 mmol) in 15 mL dry toluene was treated with stoichiometric amount of $[(3,5\text{-}(CF_3)_2C_6H_3)_2BH{\cdot}SMe_2]$ (47.75 mg, 0.095 mmol). The reaction was stirred at room temperature for 2hr. The volatile components were removed under vacuum and the remaining residue was extracted into hexane and passed through Celite. After removal of the solvent, the residue was subjected to chromatographic work-up by using TLC plates. Elution with hexane/CH_2Cl_2 (80:20 v/v) mixture yielded orange crystalline solid **11** (15 mg, 15%, Rf = 0.8).

11: MS (ESI^+): *m/z* calculated for $[M]^+$: 1486.1462, found: 1486.1342; $^{11}B\{^1H\}$ NMR (160 MHz, C_6D_6, 22 °C): δ = 2.2 (br, 1B) ppm; ^{1}H NMR (500 MHz, C_6D_6, 22 °C): δ = 8.97-6.14 (m, 44H, H_{Ar}), -5.51 (br,1H, Os-*H*-B) ppm; $^{31}P\{^1H\}$ NMR (202 MHz, C_6D_6, 22 °C): δ = -5.5 (s, 1P) ppm, δ = -16.8 (s, 1P) ppm; ^{19}F NMR (470 MHz, C_6D_6, 22 °C) : δ = -62.2 (s, 6F), -62.6 (s, 6F) ppm; IR (n-hexane, cm^{-1}): $\tilde{\nu}$ = 2360 (Os-H-B), 1356 (C=S).

II.2 Spectroscopic details

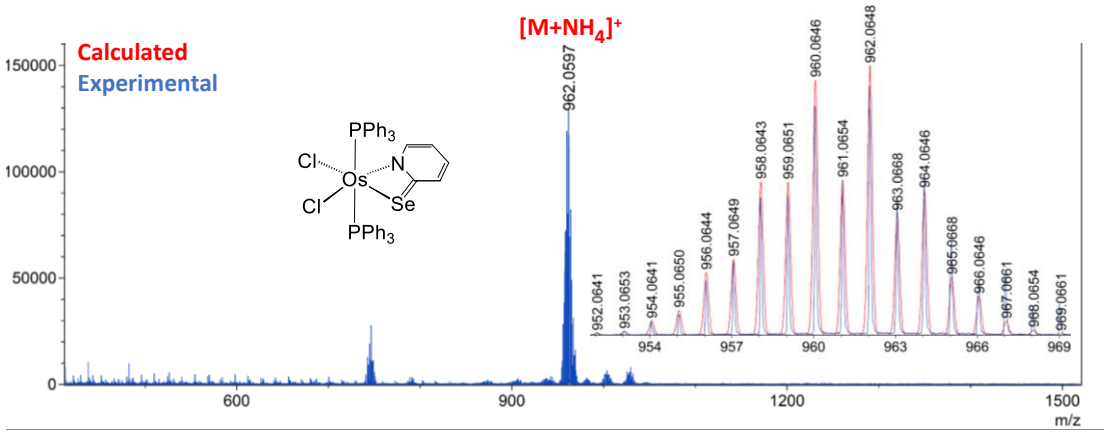


Figure S4. ESI-MS spectrum of **1**.

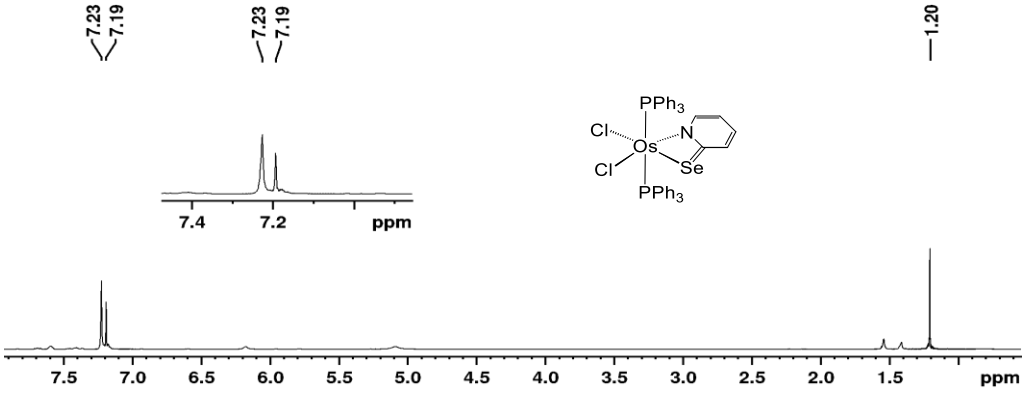


Figure S5. Evans NMR spectrum of **1**

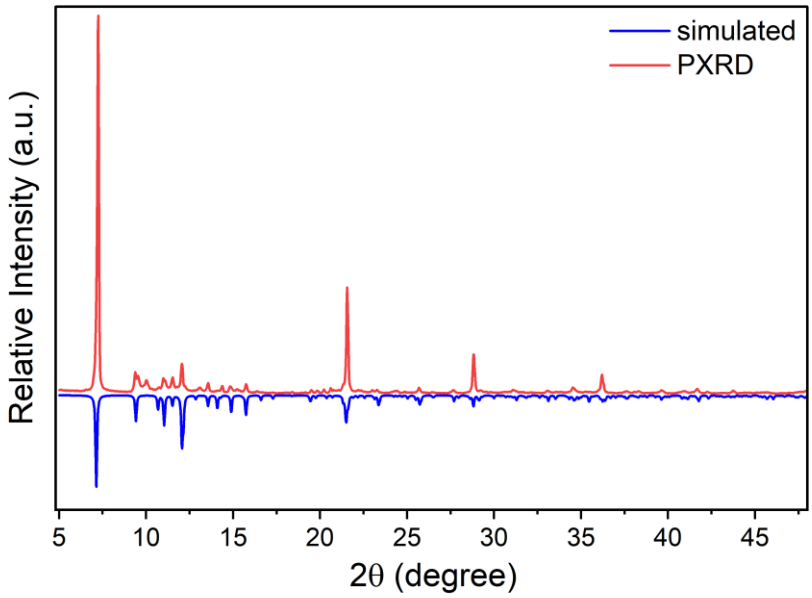


Figure S6. Comparison of powder X-ray diffraction (PXRD) patterns with the simulated patterns from single-crystal X-ray diffraction (SCXRD) for **1**.

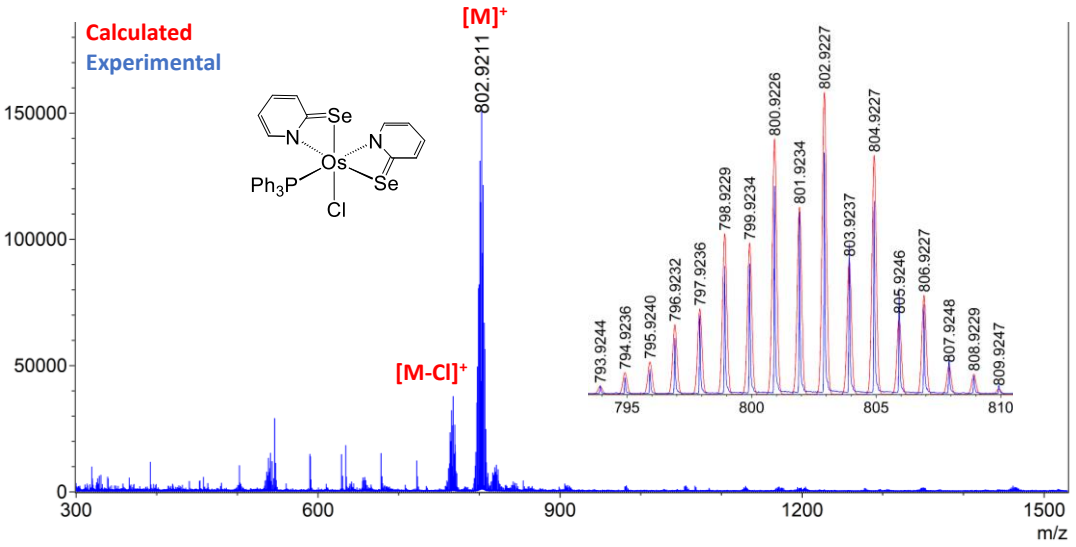


Figure S7. ESI-MS spectrum of **2**.

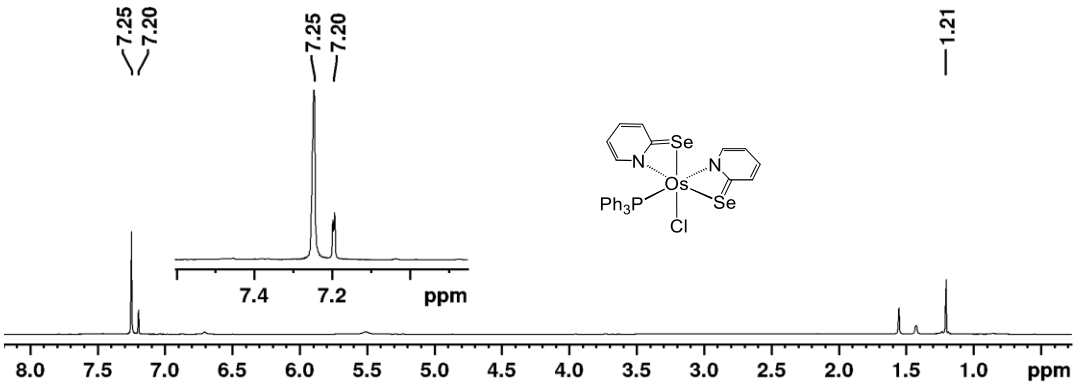


Figure S8. Evans NMR spectrum of **2**

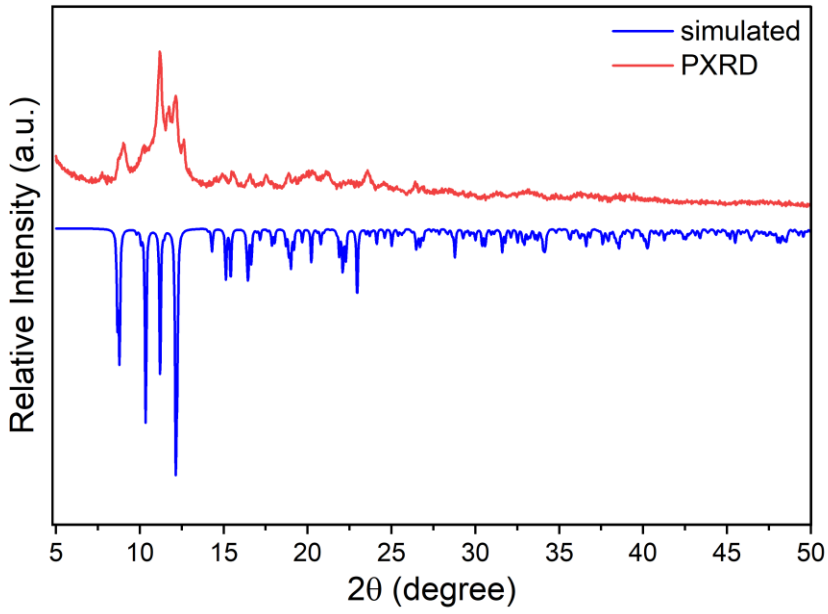


Figure S9. Comparison of powder X-ray diffraction (PXRD) patterns with the simulated patterns from single-crystal X-ray diffraction (SCXRD) for **2**.

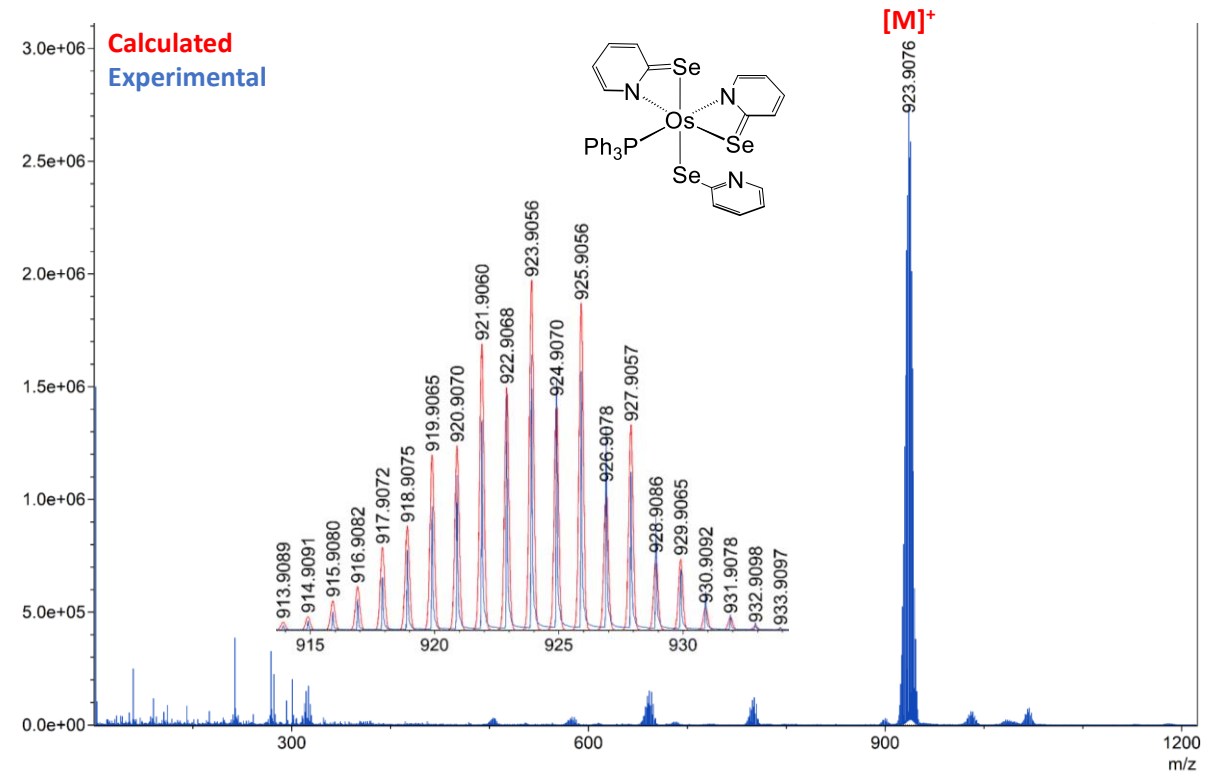


Figure S10. ESI-MS spectrum of **3**.

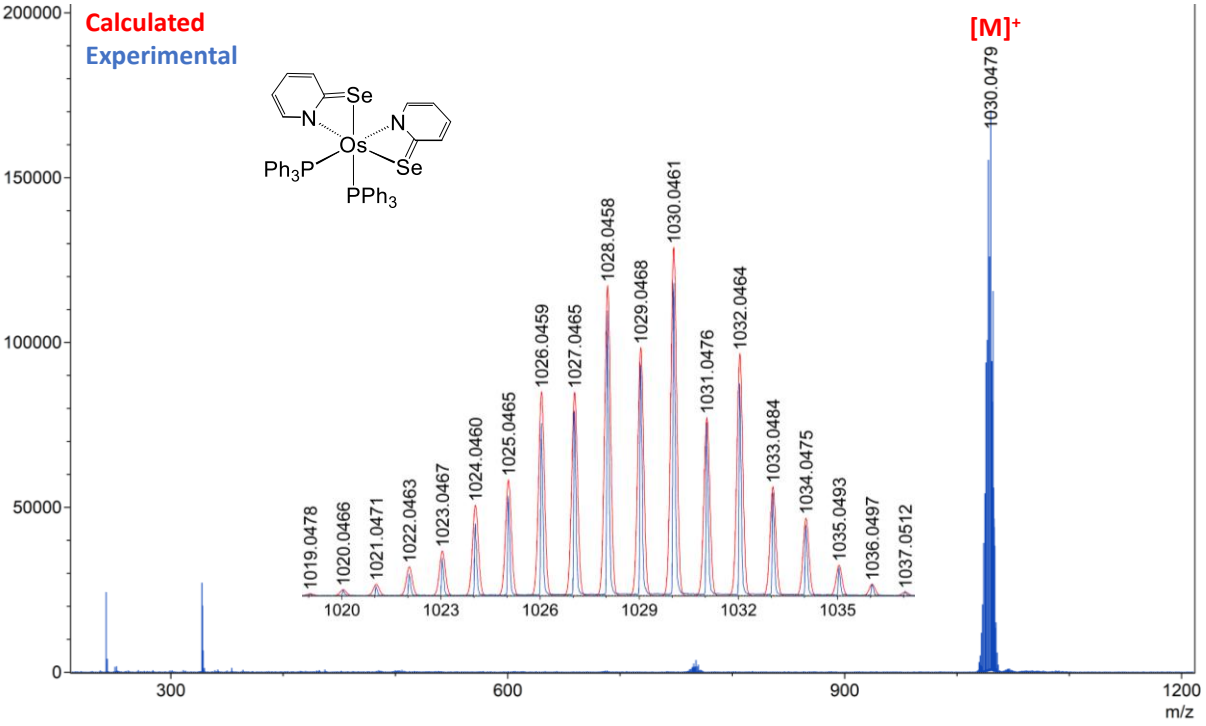


Figure S11. ESI-MS spectrum of **4**.

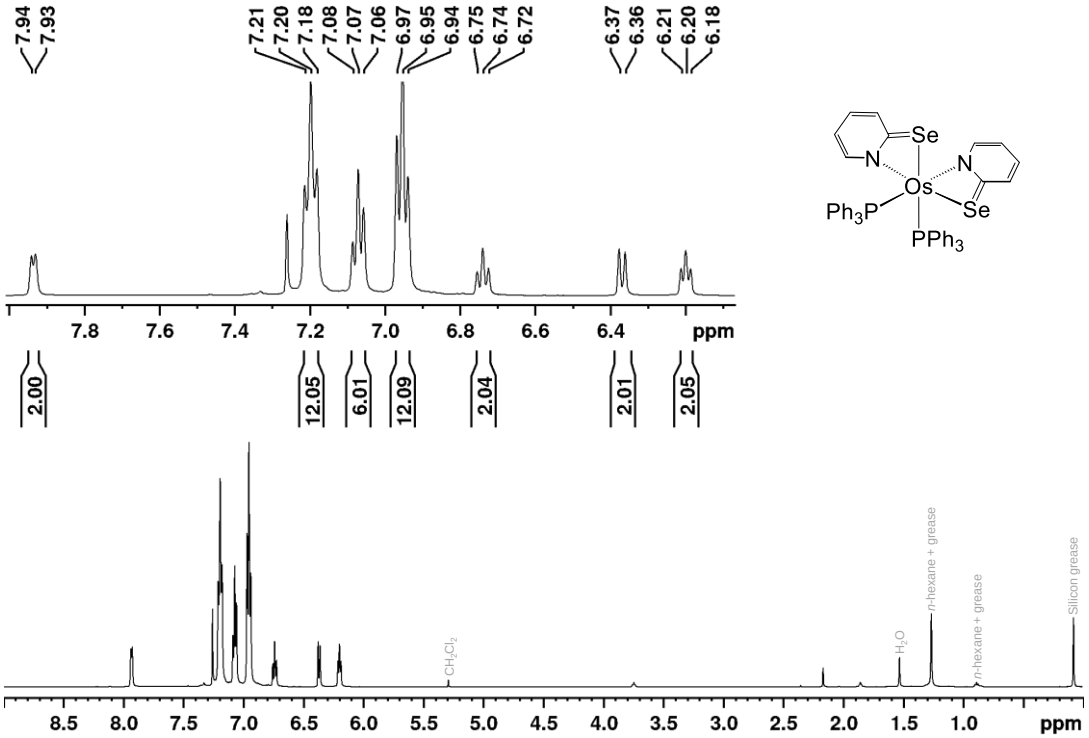


Figure S12. ^{1}H NMR spectrum of **4** in $CDCl_3$.

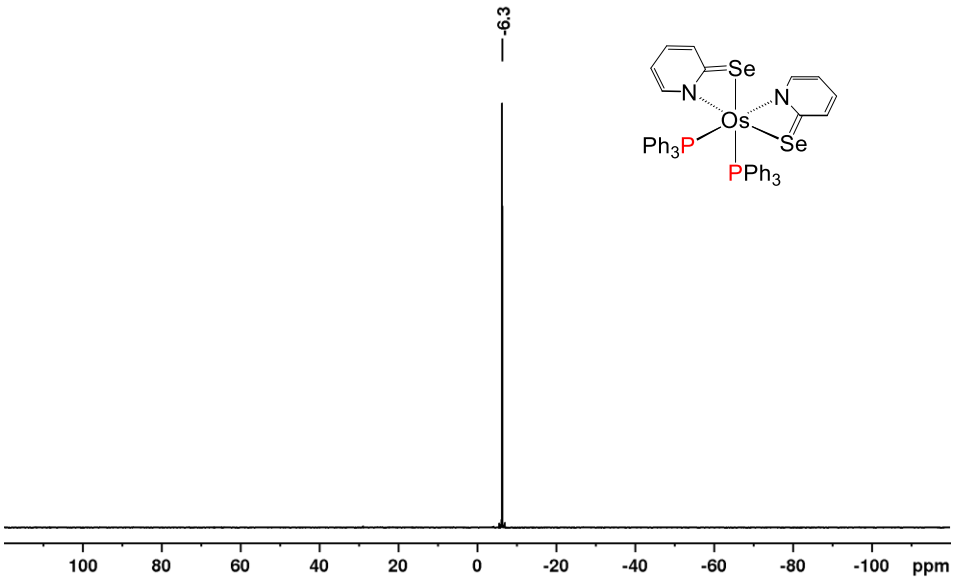


Figure S13. $^{31}P\{^{1}H\}$ NMR spectrum of **4** in $CDCl_3$.

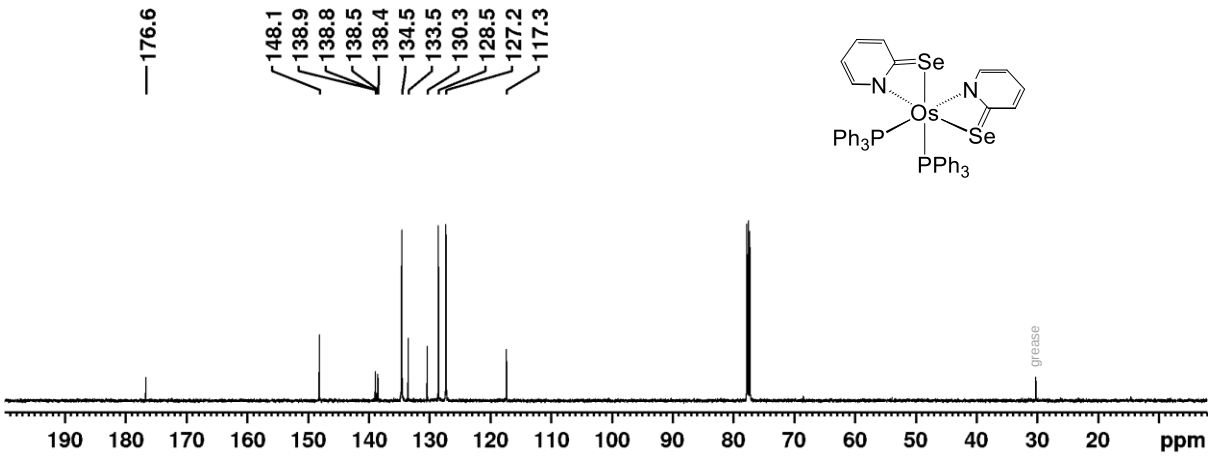
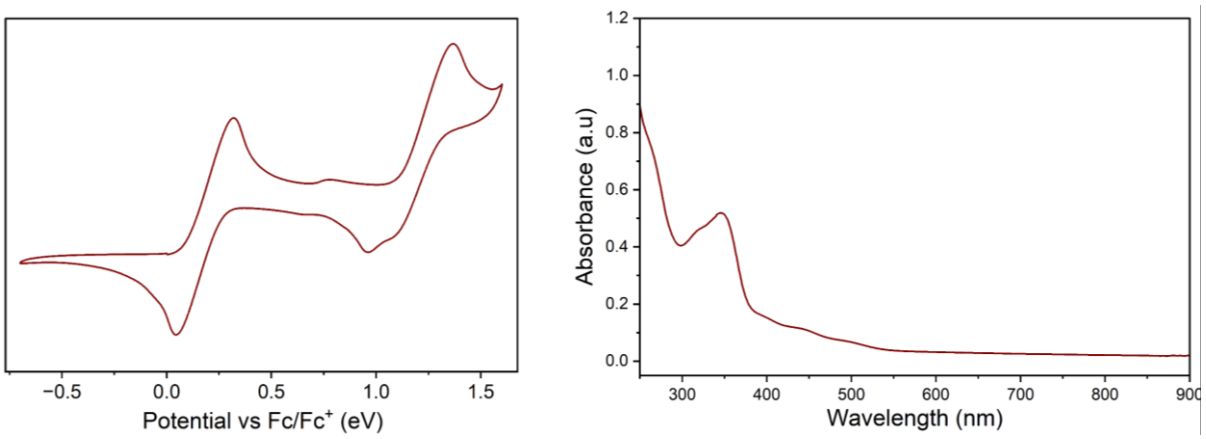


Figure S14. $^{13}C\{^{1}H\}$ NMR spectrum of **4** in $CDCl_3$.



(a) (b)

Figure S15. Cyclic voltammetry and UV-Vis spectrum of **4**.

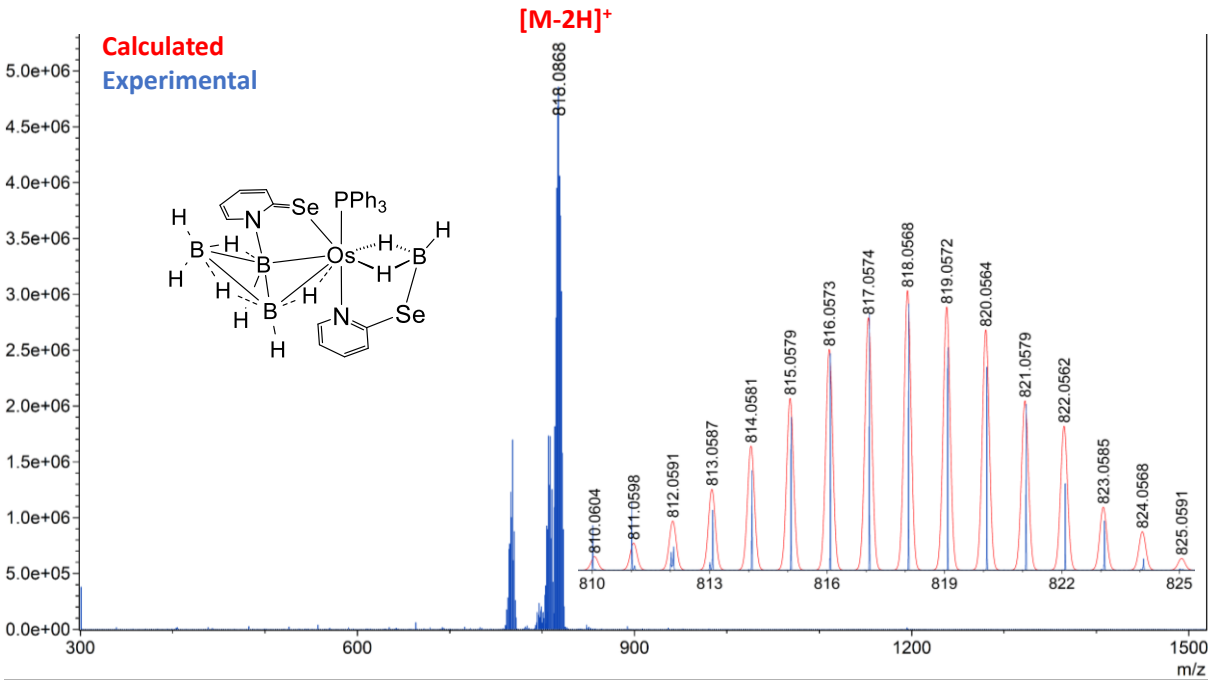


Figure S16. ESI-MS spectrum of **5**.

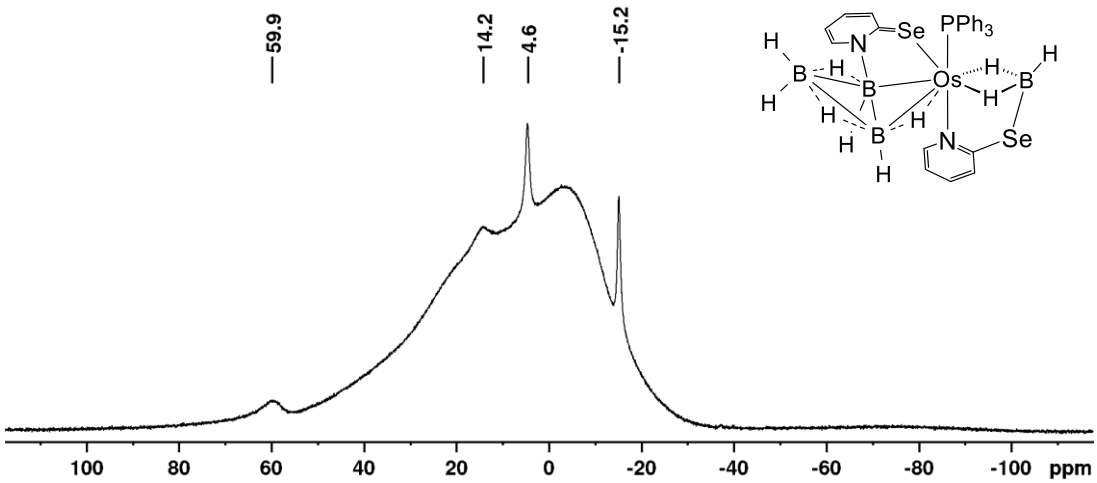


Figure S17. $^{11}B\{^{1}H\}$ NMR spectrum of **5** in C_6D_6.

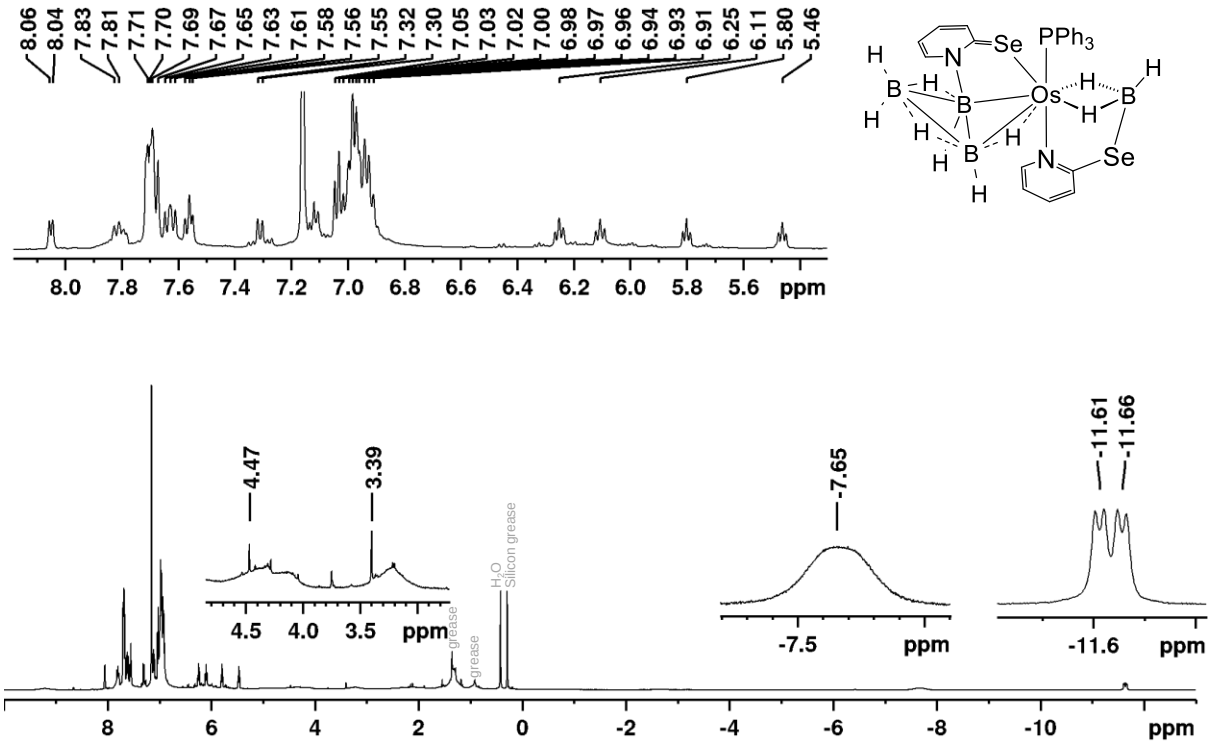


Figure S18. ^{1}H NMR spectrum of **5** in C_6D_6.

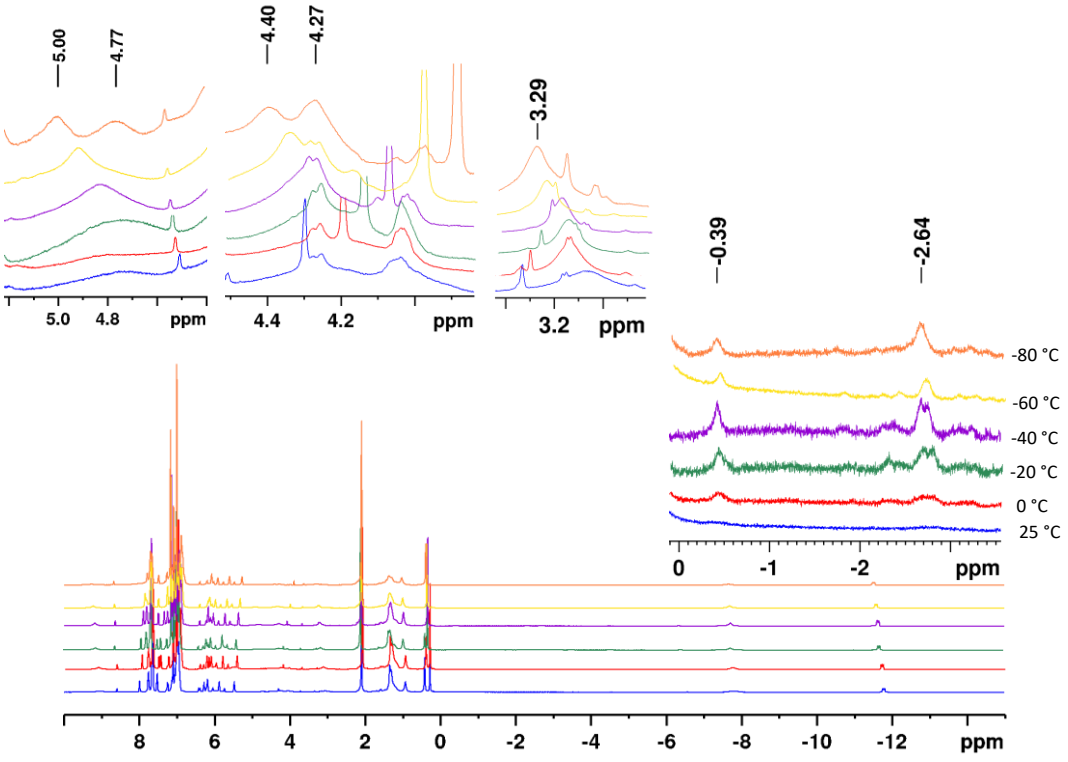


Figure S19. ^{1}H VT-NMR spectra of **5** in toluene-D_8.

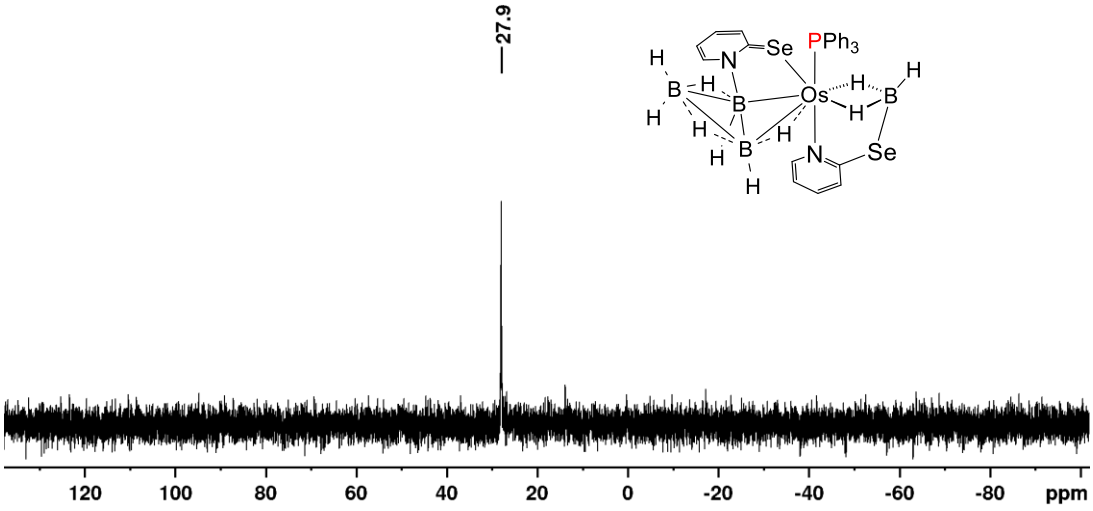


Figure S20. $^{31}P\{^{1}H\}$ NMR spectrum of **5** in C_6D_6.

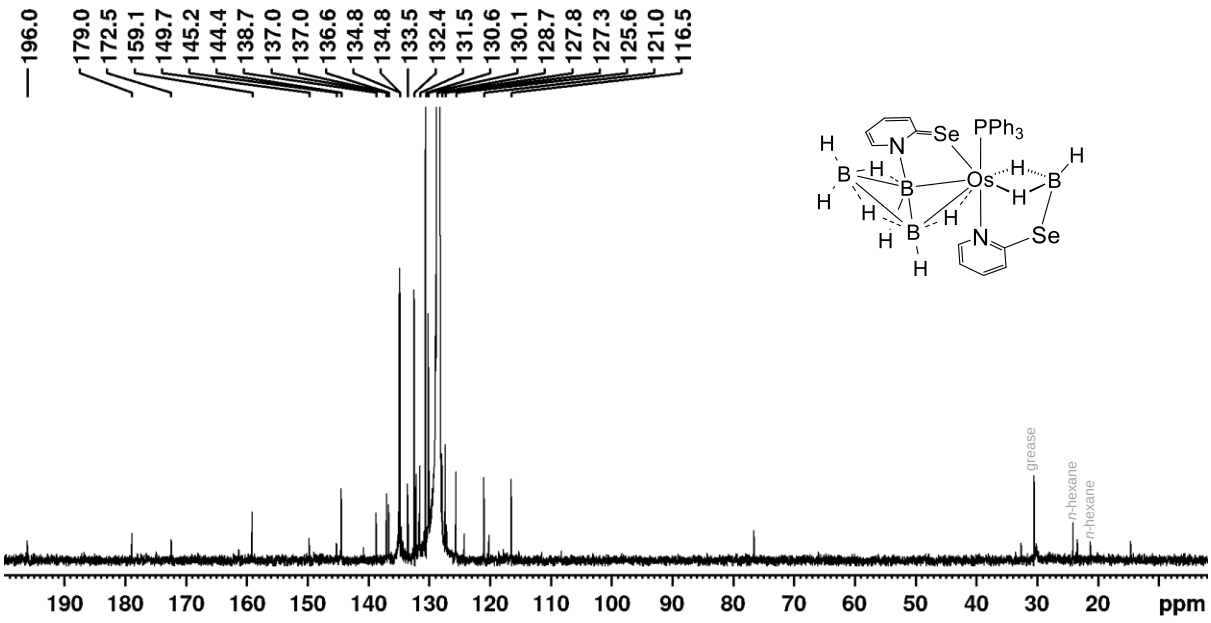


Figure S21. $^{13}C\{^{1}H\}$ NMR spectrum of **5** in C_6D_6.

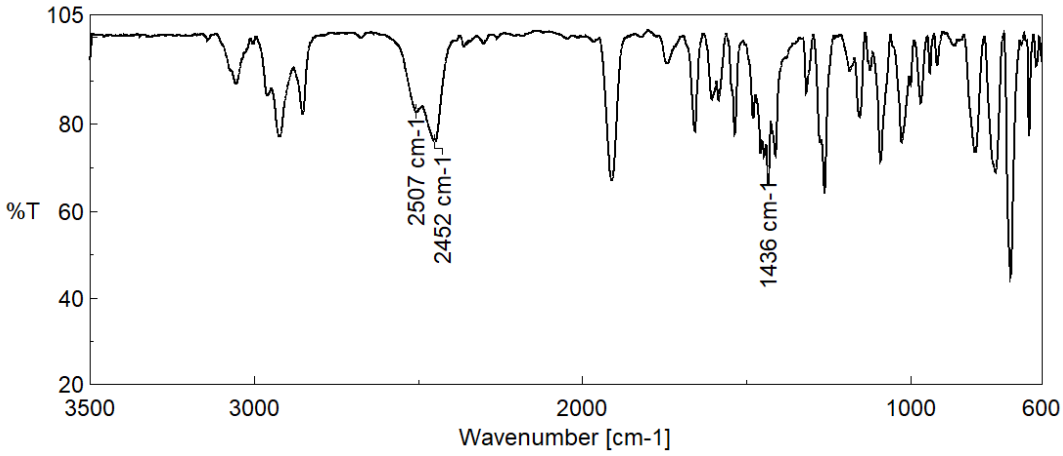


Figure S22. IR spectrum of **5**.

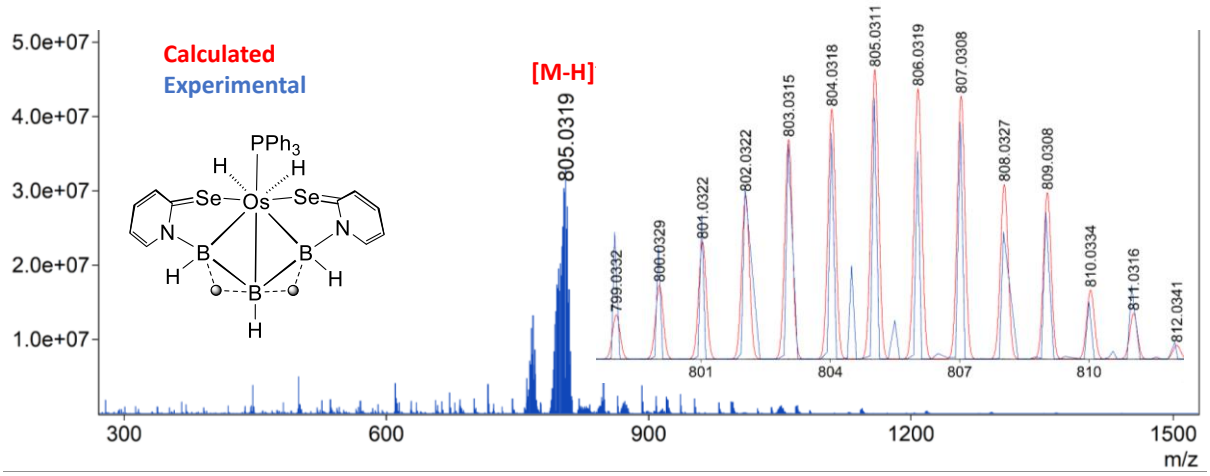


Figure S23. ESI-MS spectrum of **6**.

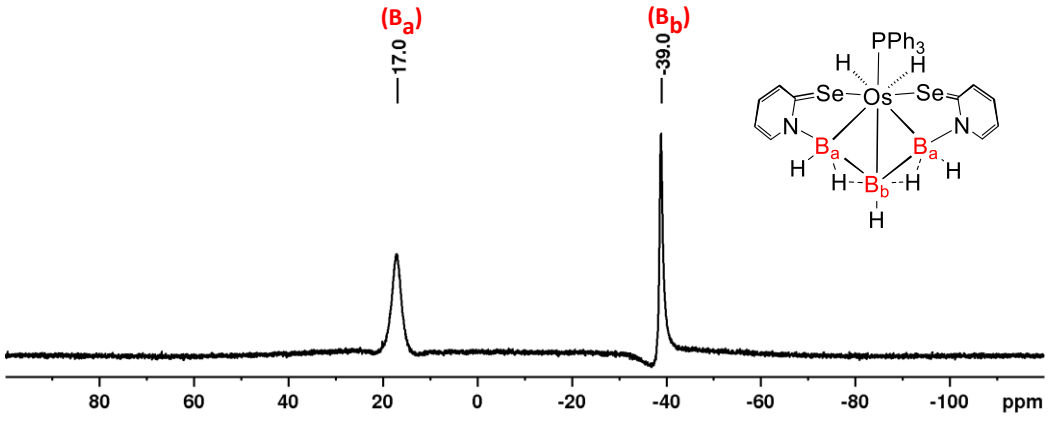


Figure S24. $^{11}B\{^{1}H\}$ NMR spectrum of **6** in $CDCl_3$.

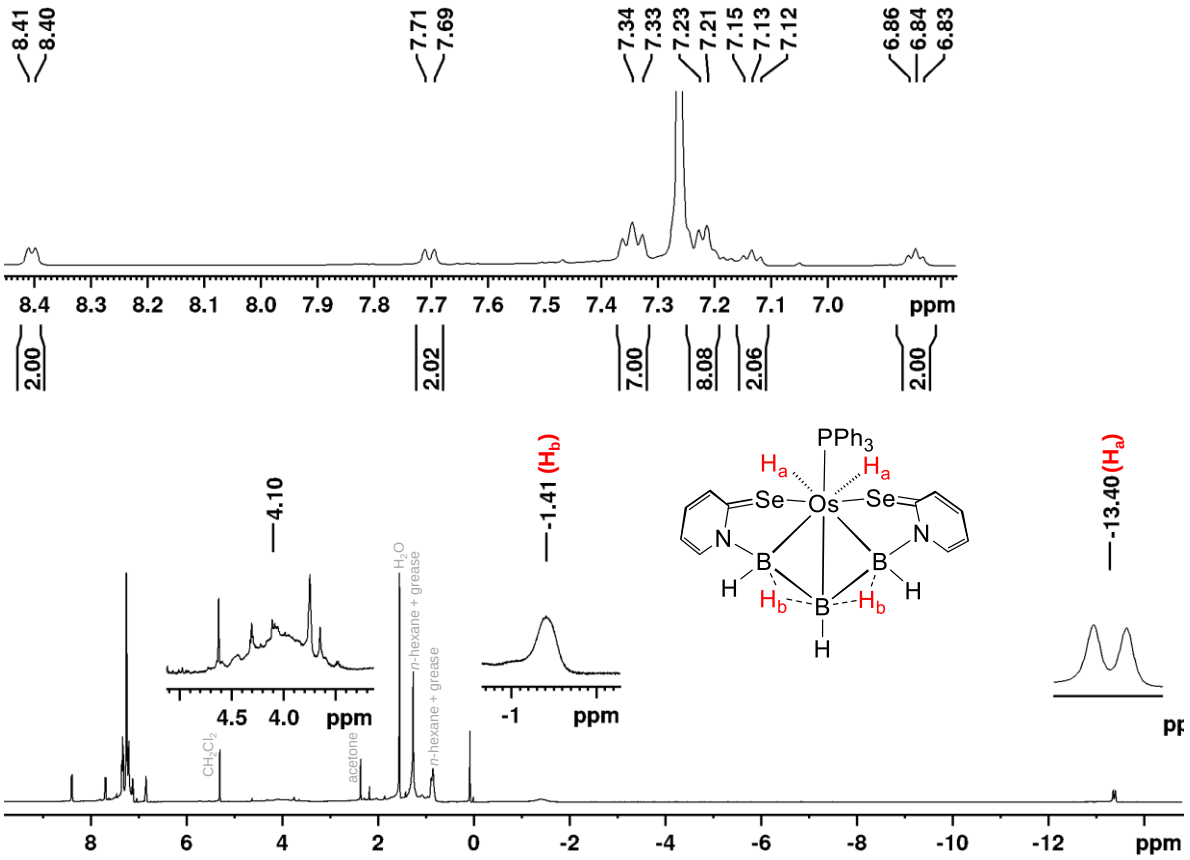


Figure S25. ^{1}H NMR spectrum of **6** in $CDCl_3$.

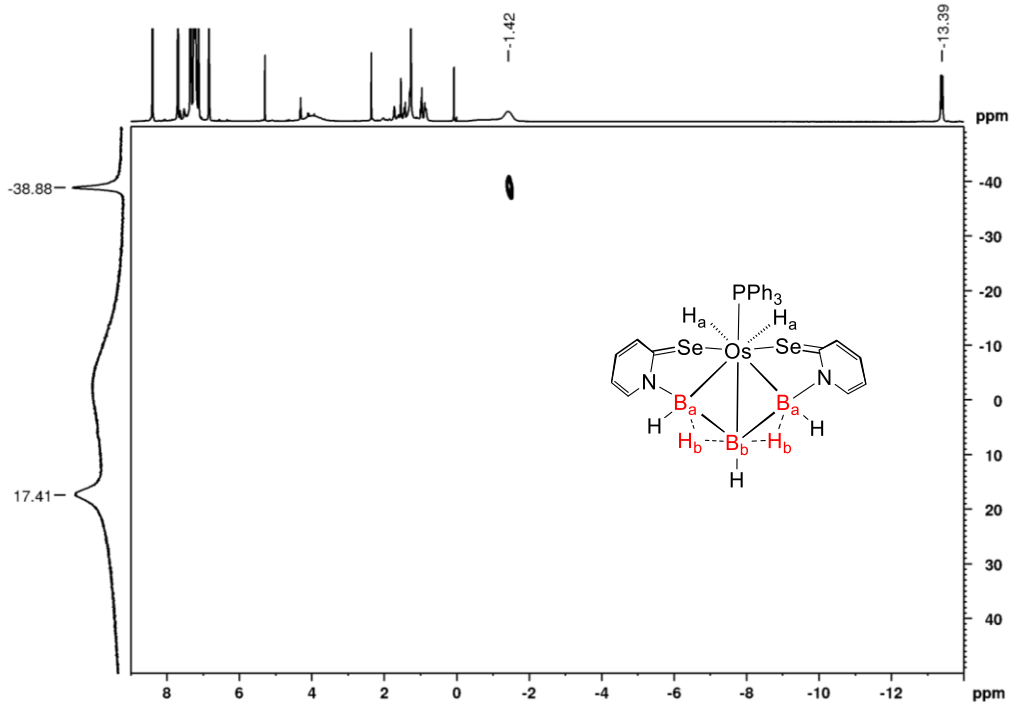


Figure S26. ^{1}H(^{11}B)/^{11}B(^{1}H) HSQC spectrum of **6** in $CDCl_3$.

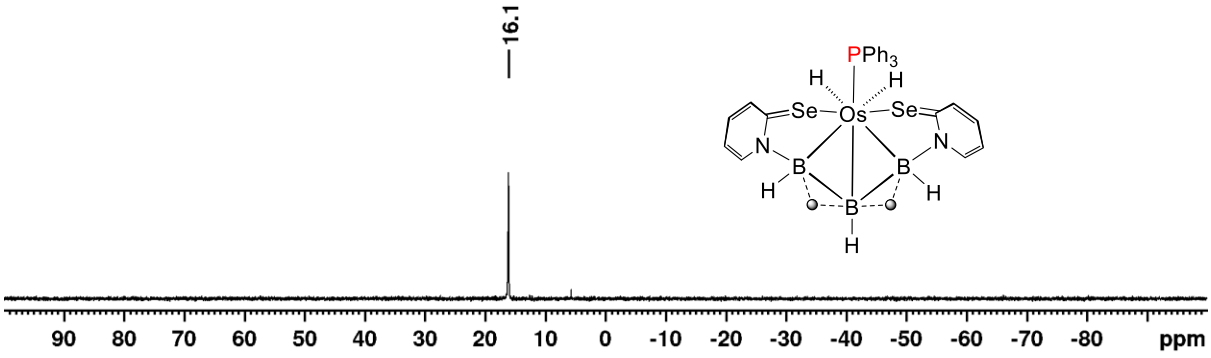


Figure S27. $^{31}P\{^{1}H\}$ NMR spectrum of **6** in $CDCl_3$.

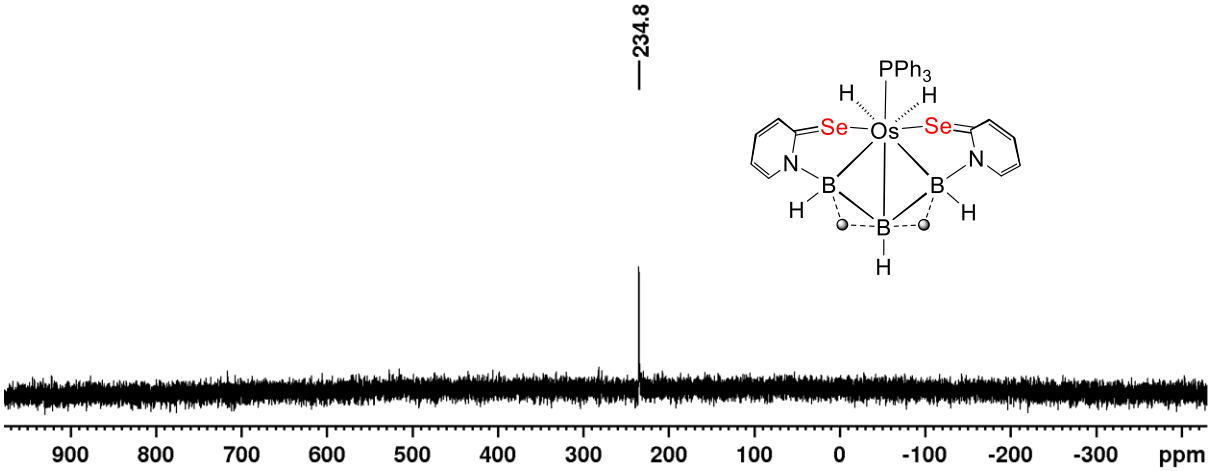


Figure S28. ^{77}Se NMR spectrum of **6** in $CDCl_3$.

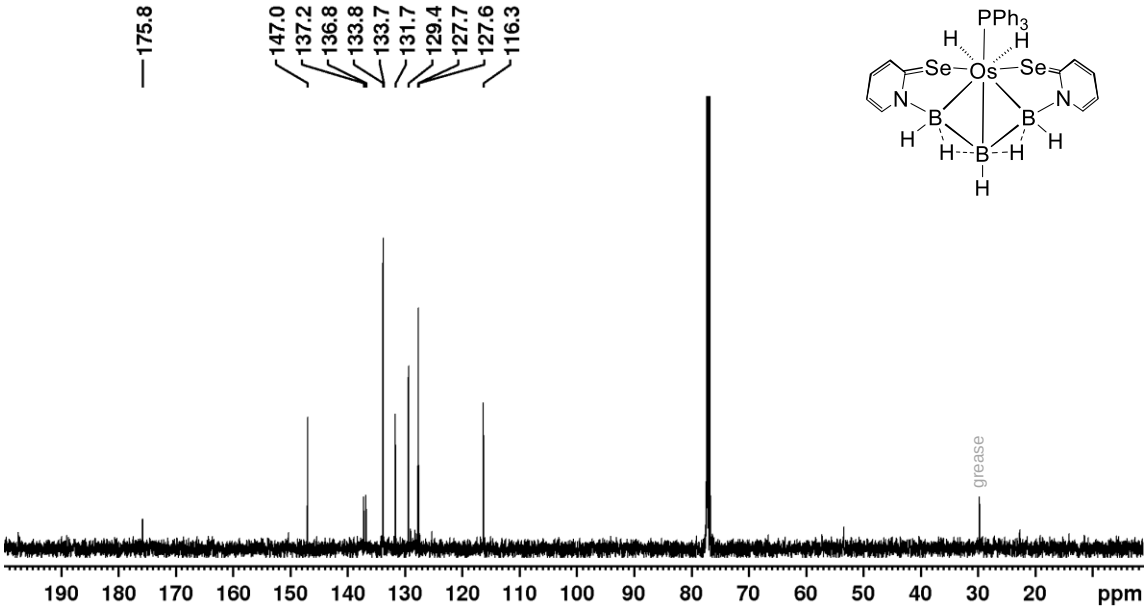


Figure S29. $^{13}C\{^{1}H\}$ NMR spectrum of **6** in $CDCl_3$.

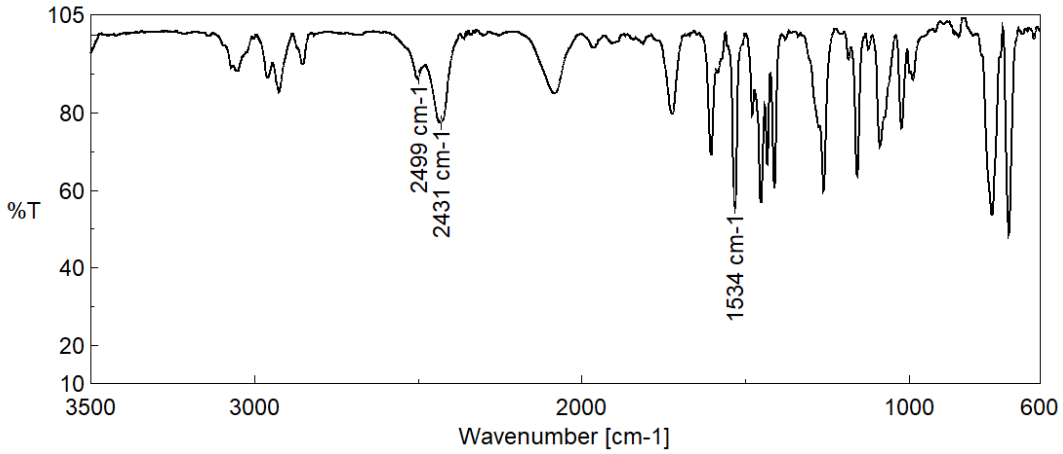


Figure S30. IR spectrum of **6**.

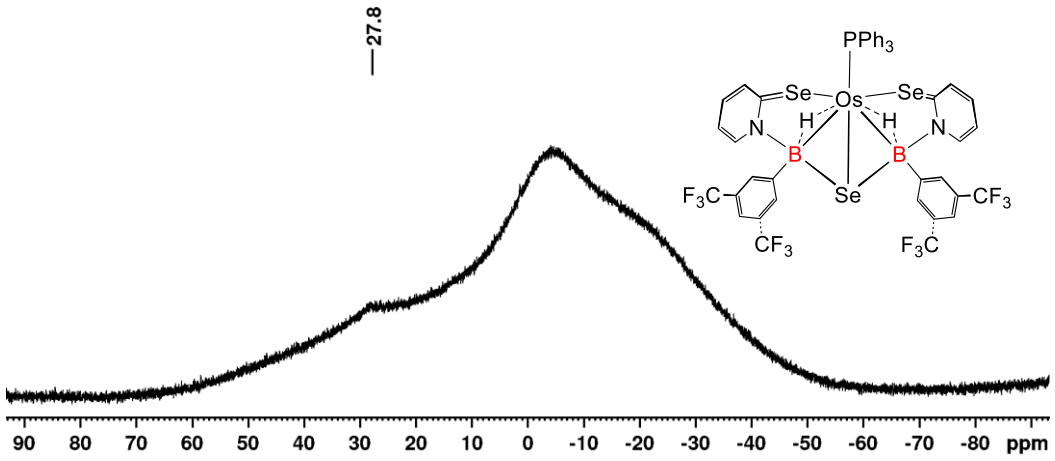


Figure S31. $^{11}B\{^{1}H\}$ NMR spectrum of **7** in C_6D_6.

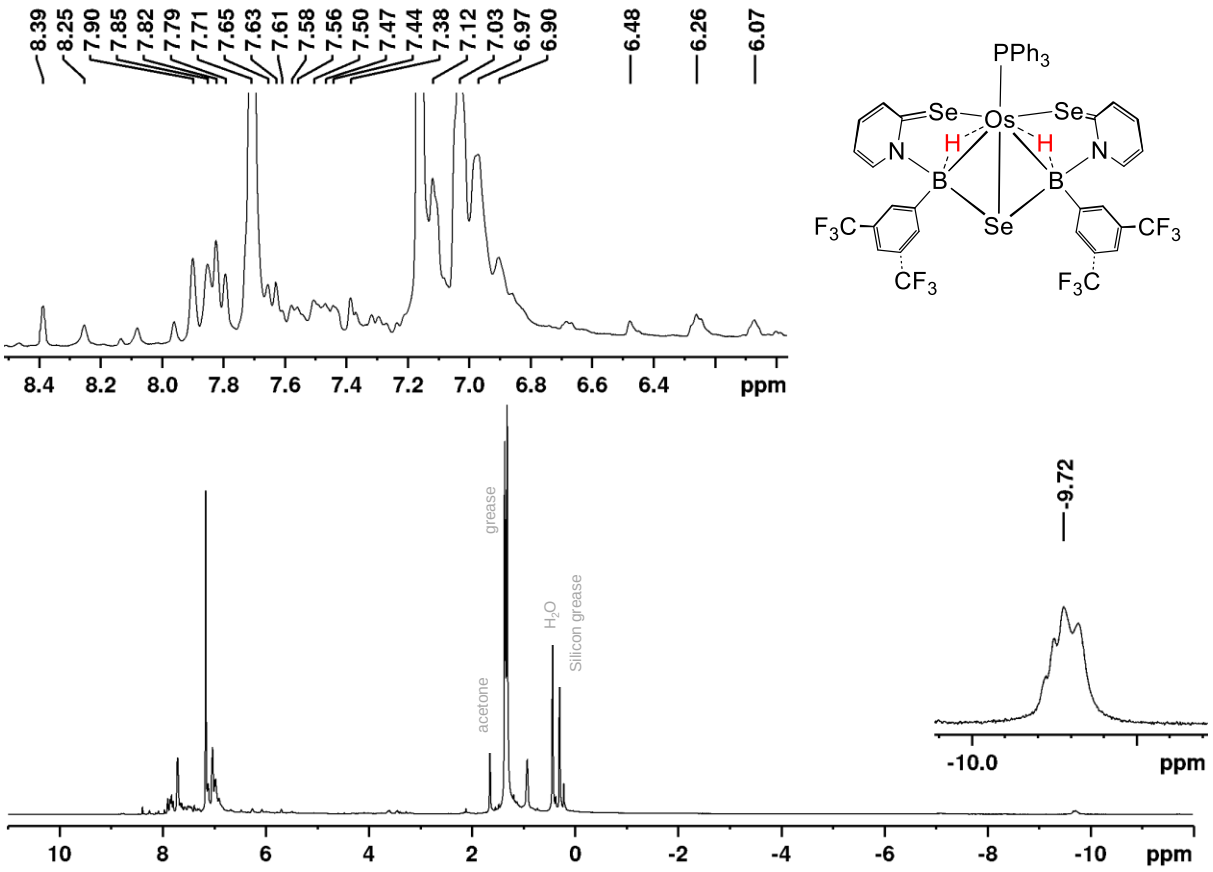


Figure S32. ^{1}H NMR spectrum of **7** in C_6D_6.

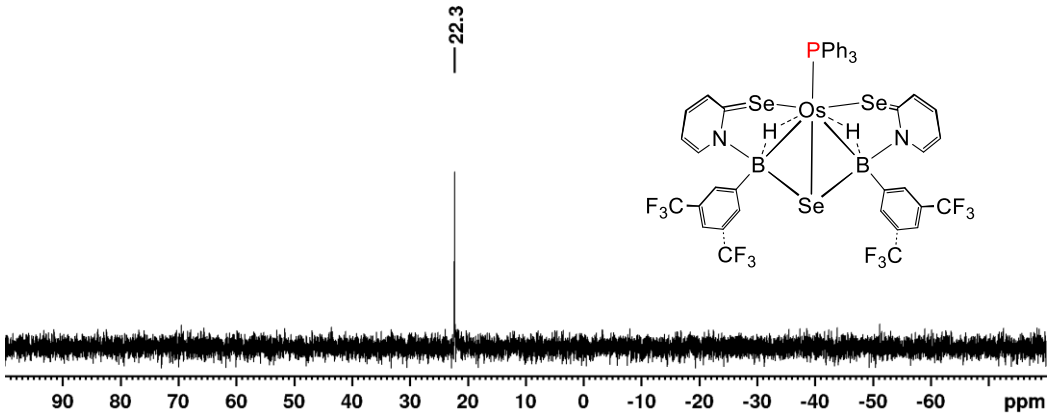


Figure S33. $^{31}P\{^{1}H\}$ NMR spectrum of **7** in C_6D_6.

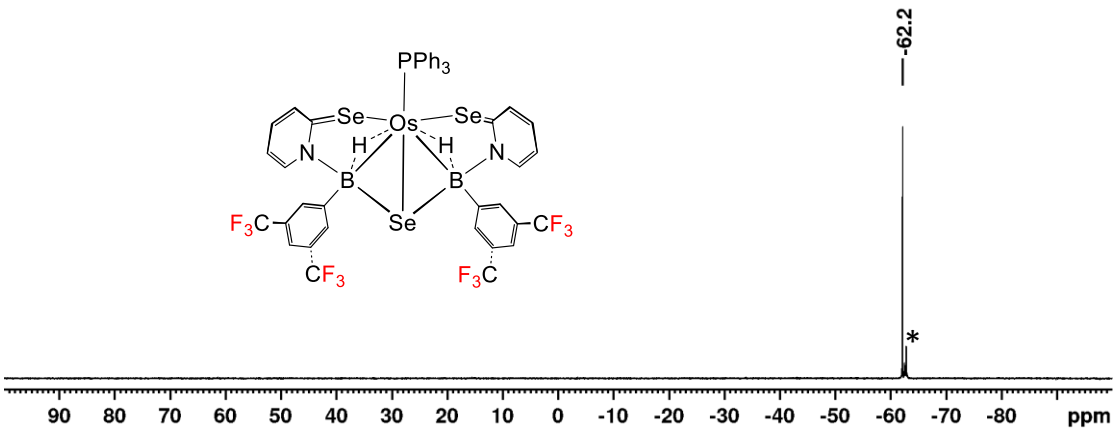


Figure S34. $^{19}F\{^{1}H\}$ NMR spectrum of **7** in C_6D_6. (* = inseparable impurity of BH_2R; R = $(CF_3)_2C_6H_3$)

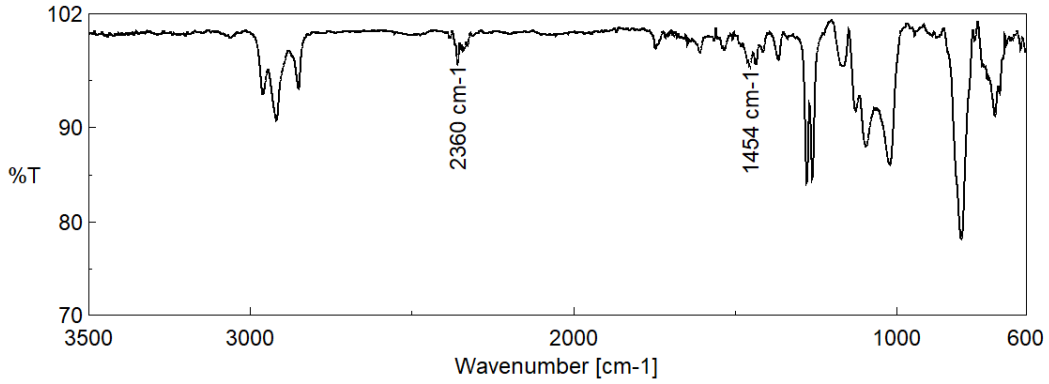


Figure S35. IR spectrum of **7**.

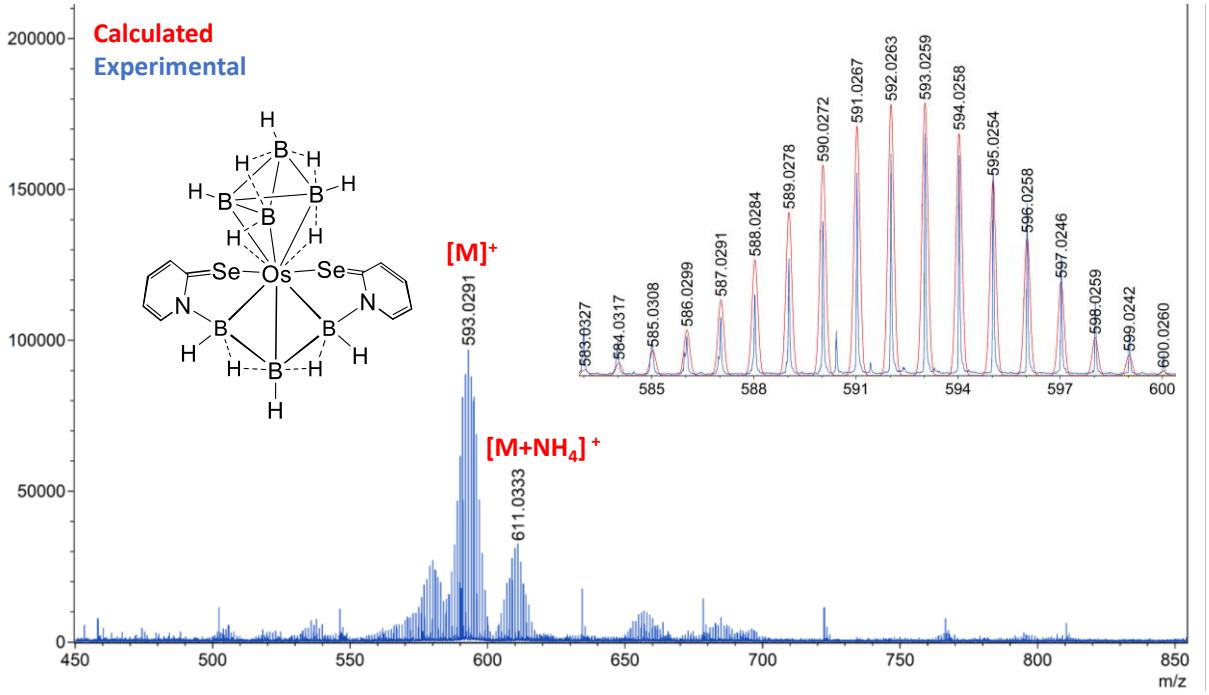


Figure S36. ESI-MS spectrum of **8**.

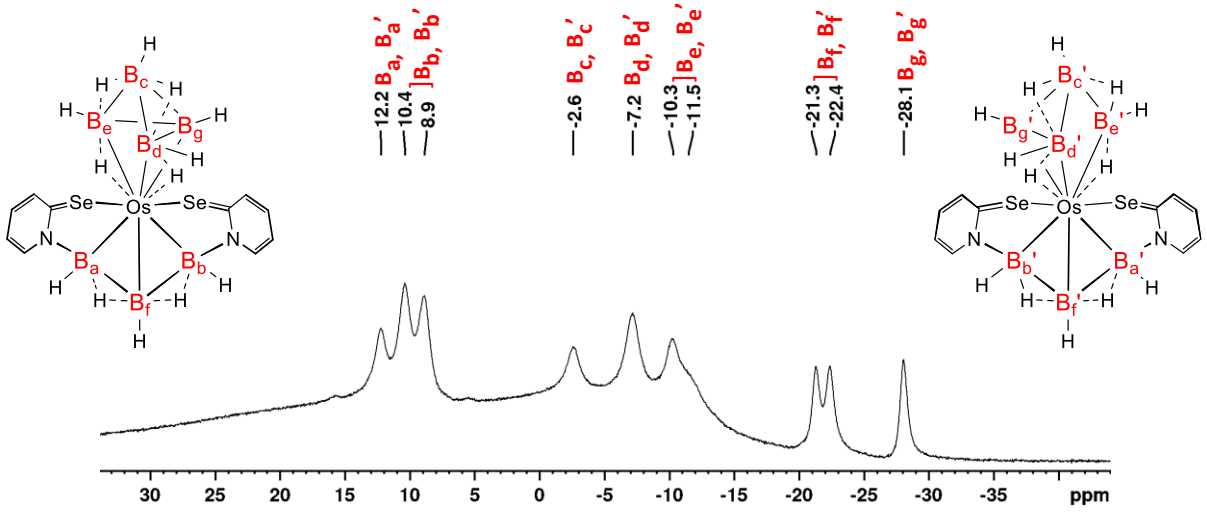


Figure S37. $^{11}B\{^{1}H\}$ NMR spectrum of **8** in $CDCl_3$.

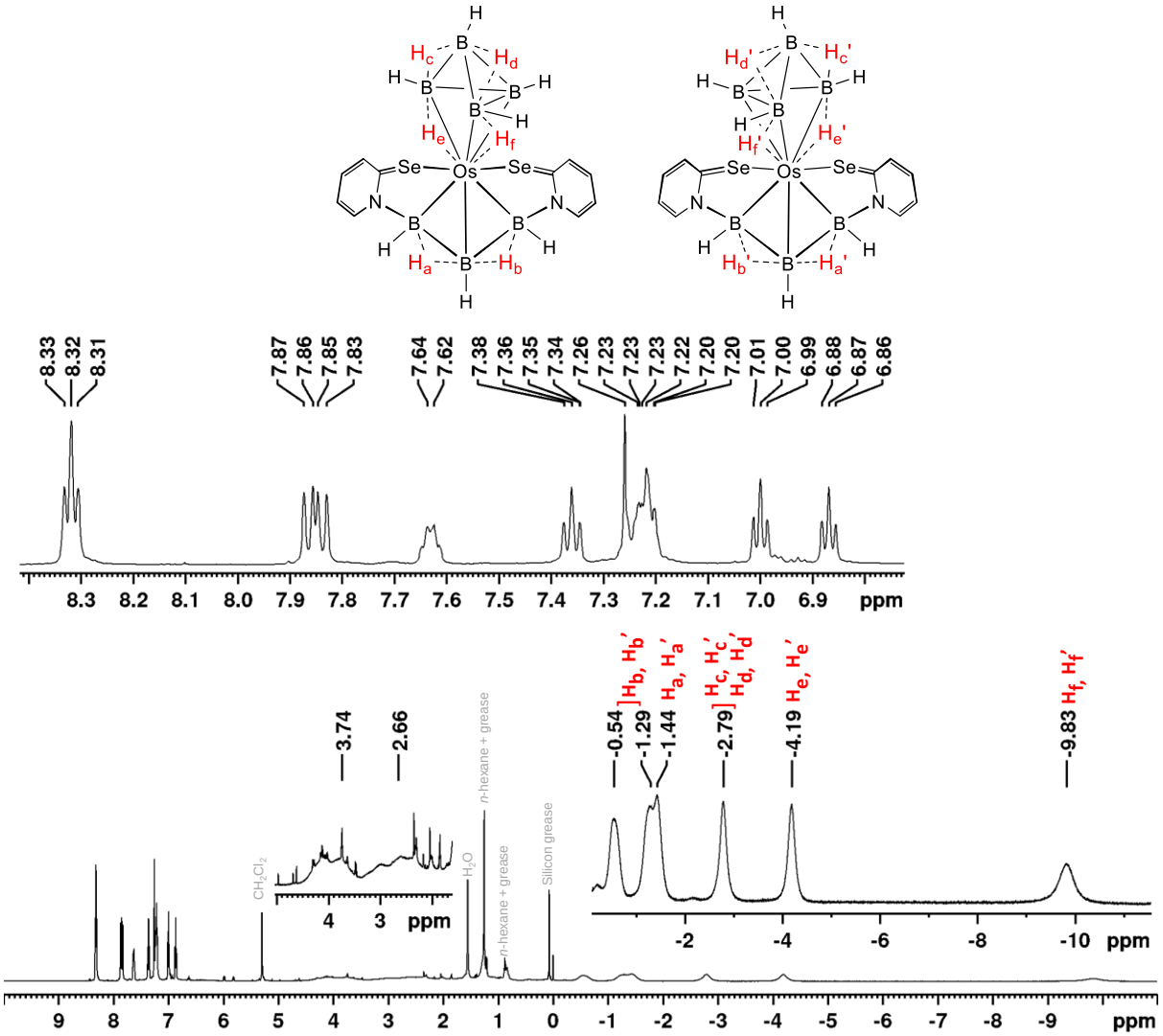


Figure S38. ^{1}H NMR spectrum of **8** in $CDCl_3$.

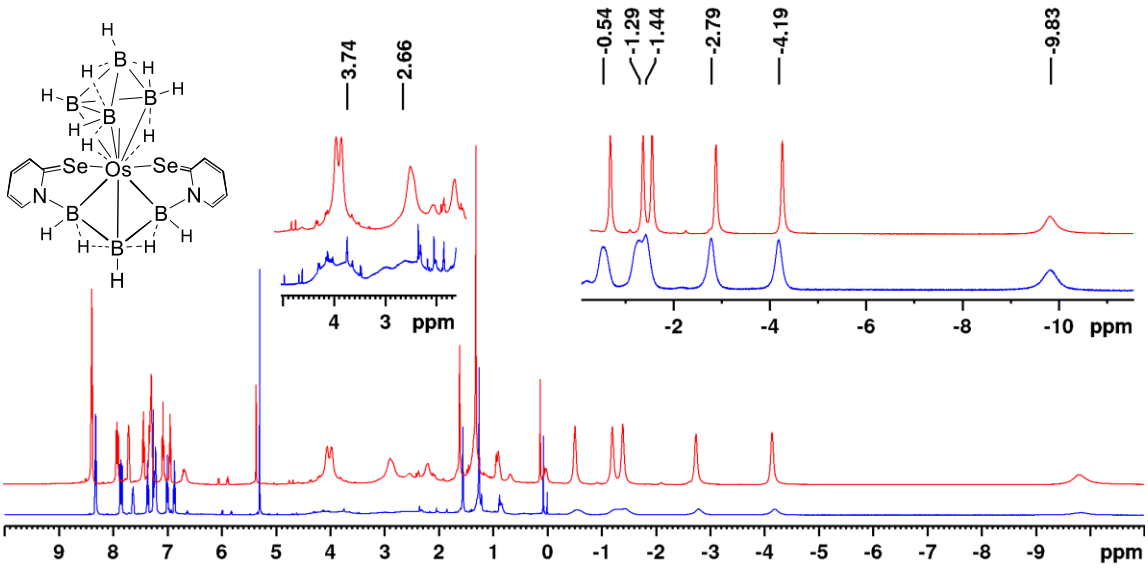


Figure S39. Stacked ^{1}H (blue) and $^{1}H\{^{11}B\}$ (red) NMR spectrum of **8** in $CDCl_3$.

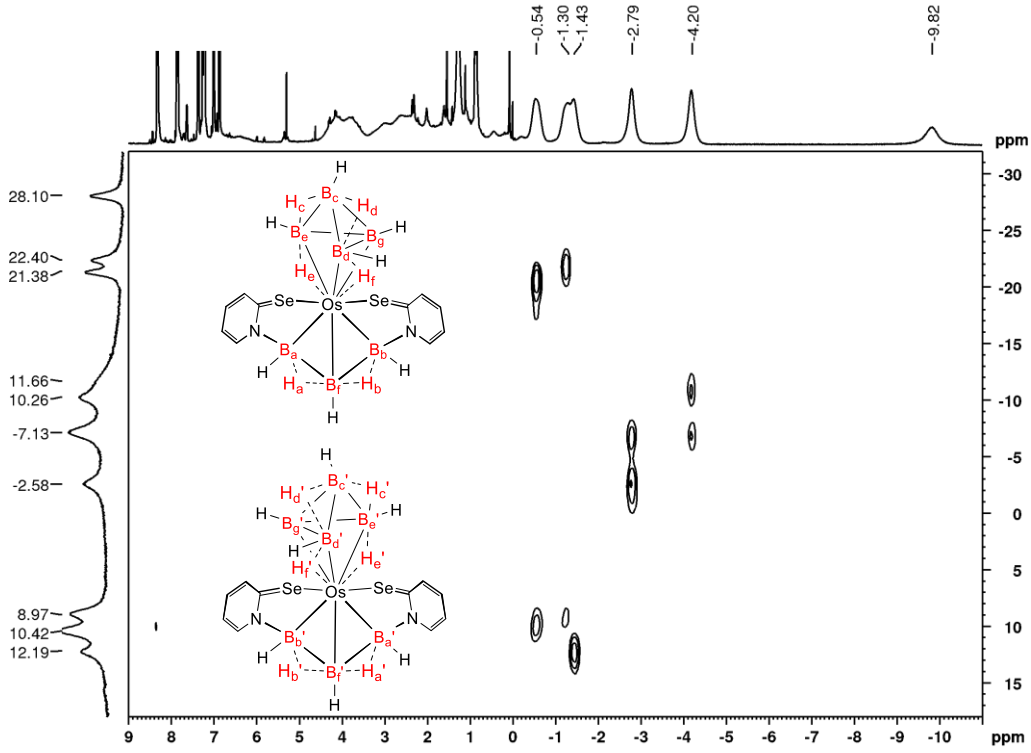


Figure S40. $^{1}H(^{11}B)/^{11}B(^{1}H)$ HSQC spectrum of **8** in $CDCl_3$.

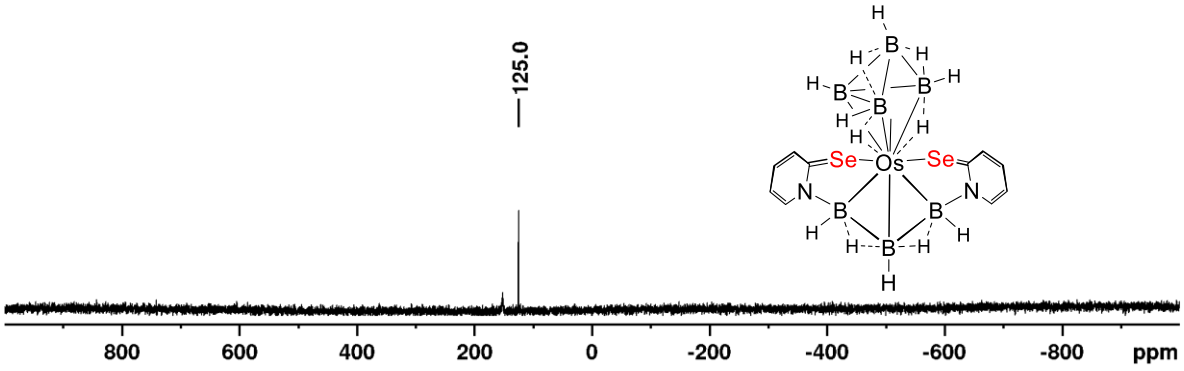


Figure S41. ^{77}Se NMR spectrum of **8** in $CDCl_3$.

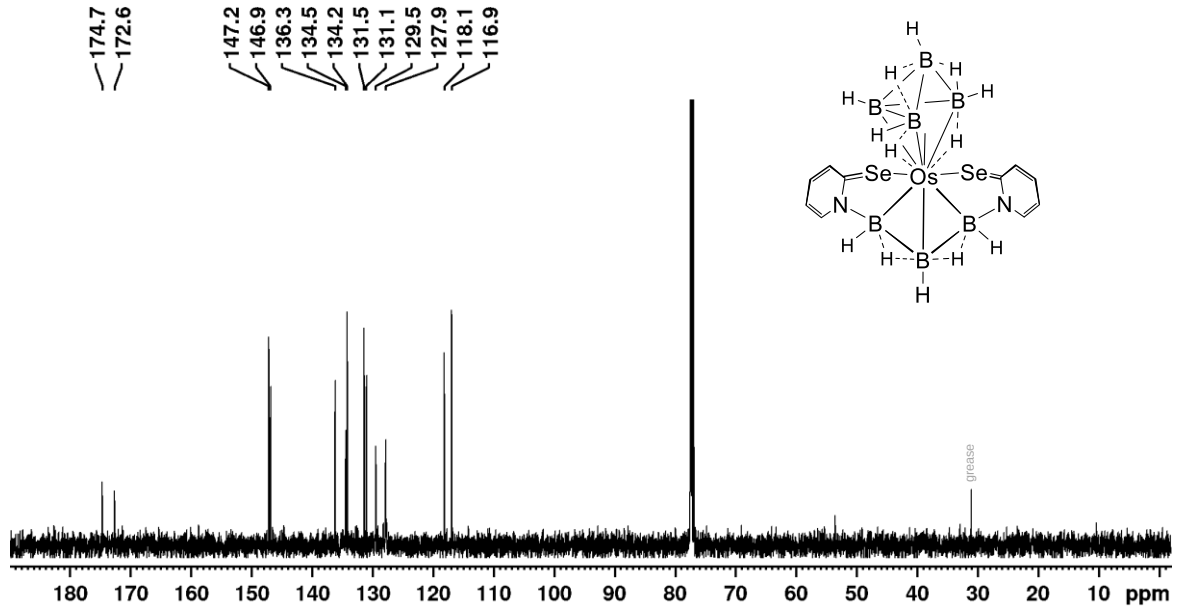


Figure S42. $^{13}C\{^1H\}$ NMR spectrum of **8** in $CDCl_3$.

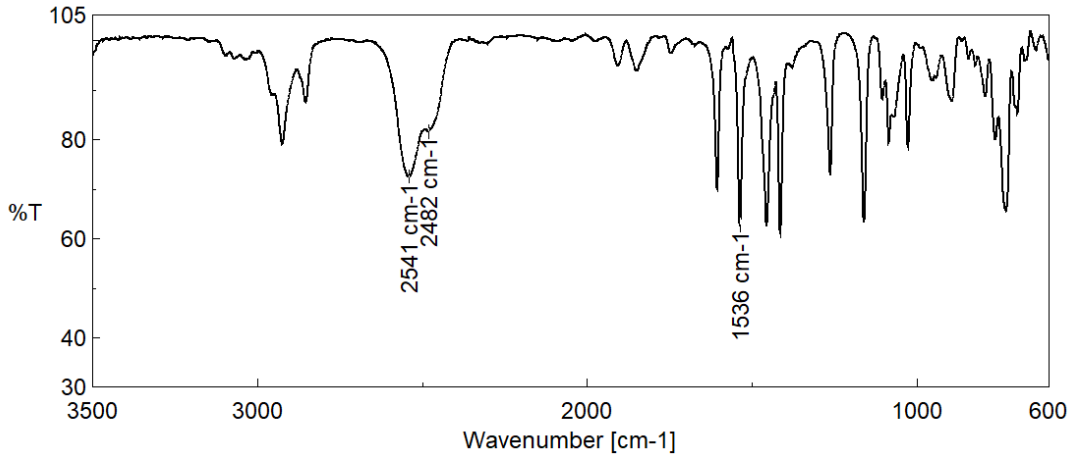


Figure S43. IR spectrum of **8**.

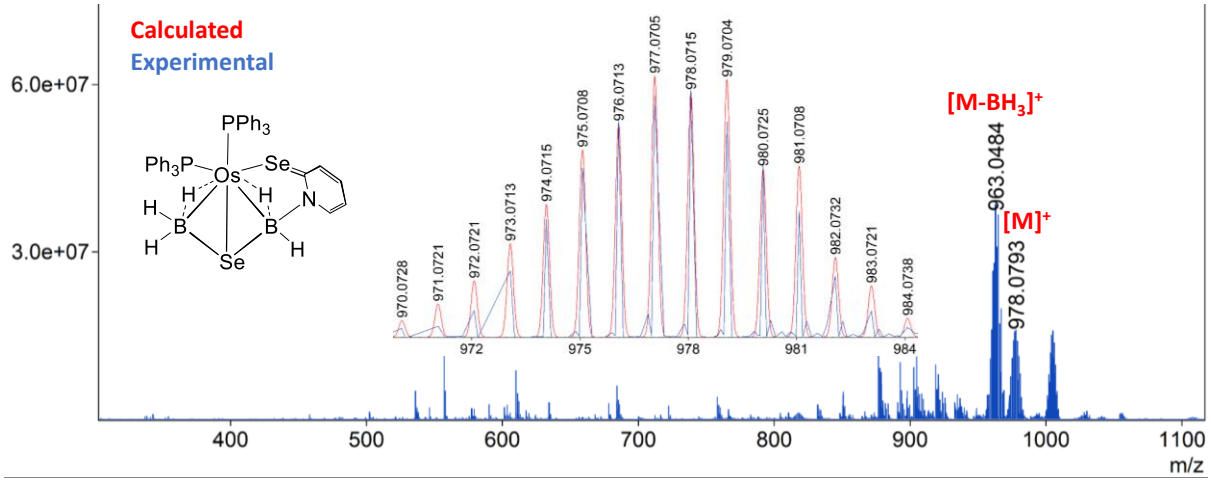


Figure S44. ESI-MS spectrum of **10**.

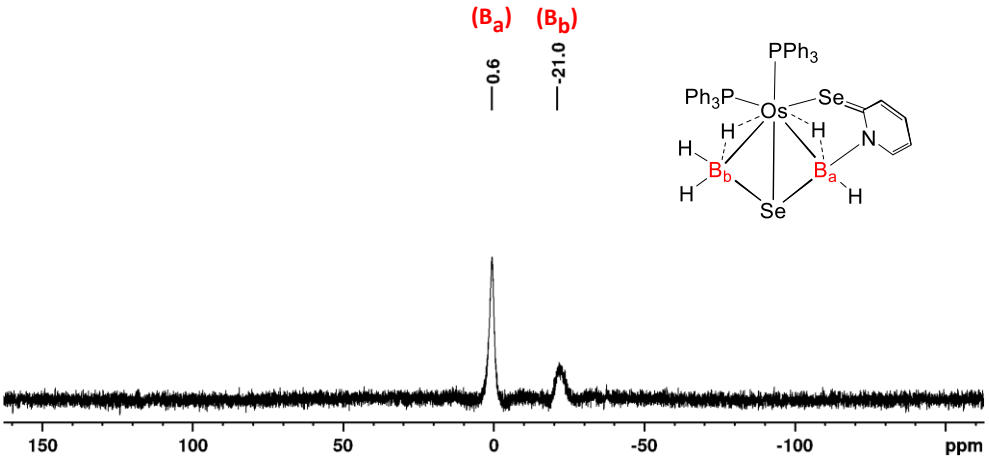


Figure S45. $^{11}B\{^{1}H\}$ NMR spectrum of **10** in C_6D_6.

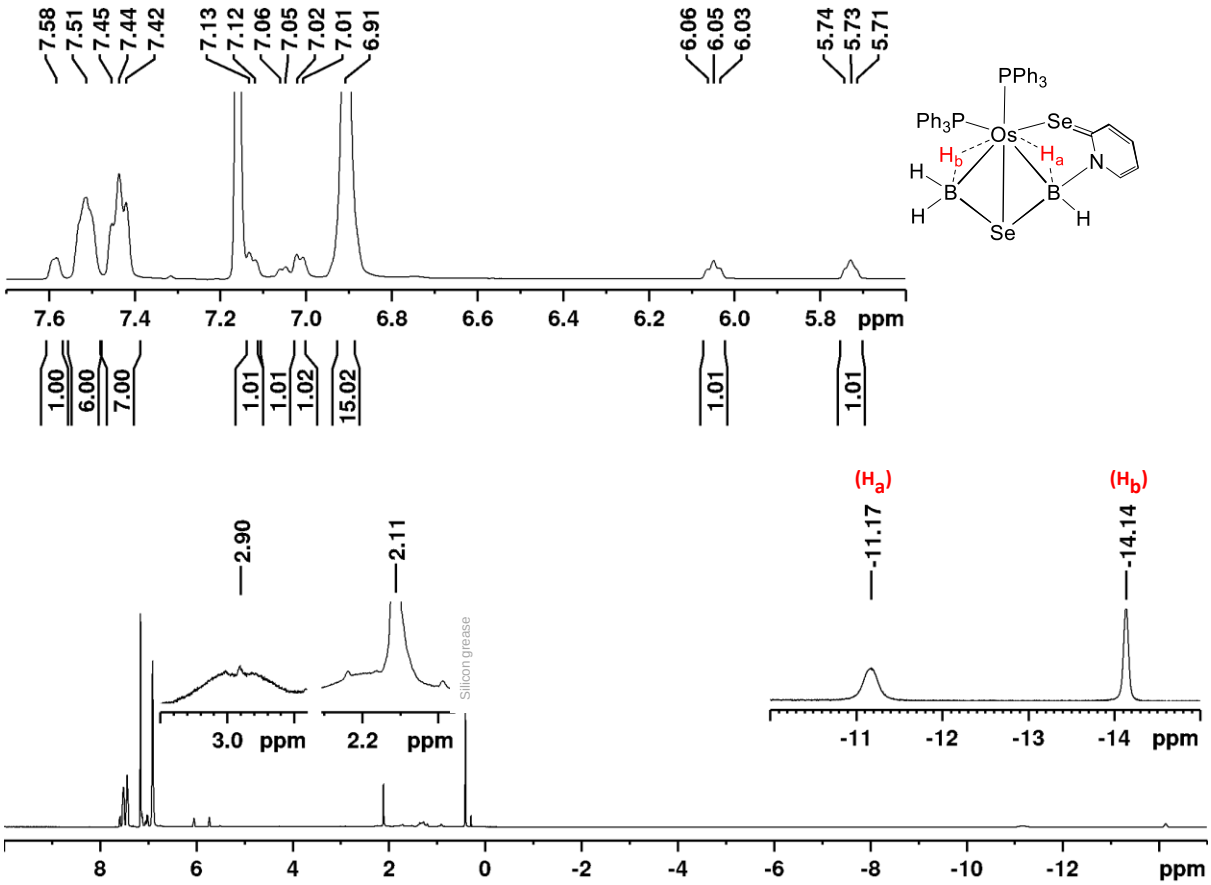


Figure S46. ^{1}H NMR spectrum of **10** in C_6D_6.

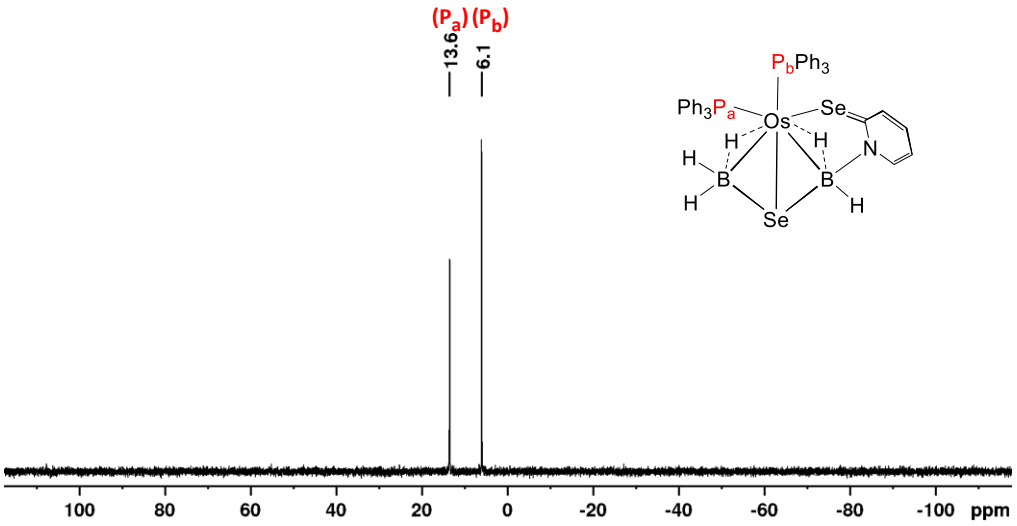


Figure S47. $^{31}P\{^{1}H\}$ NMR spectrum of **10** in C_6D_6.

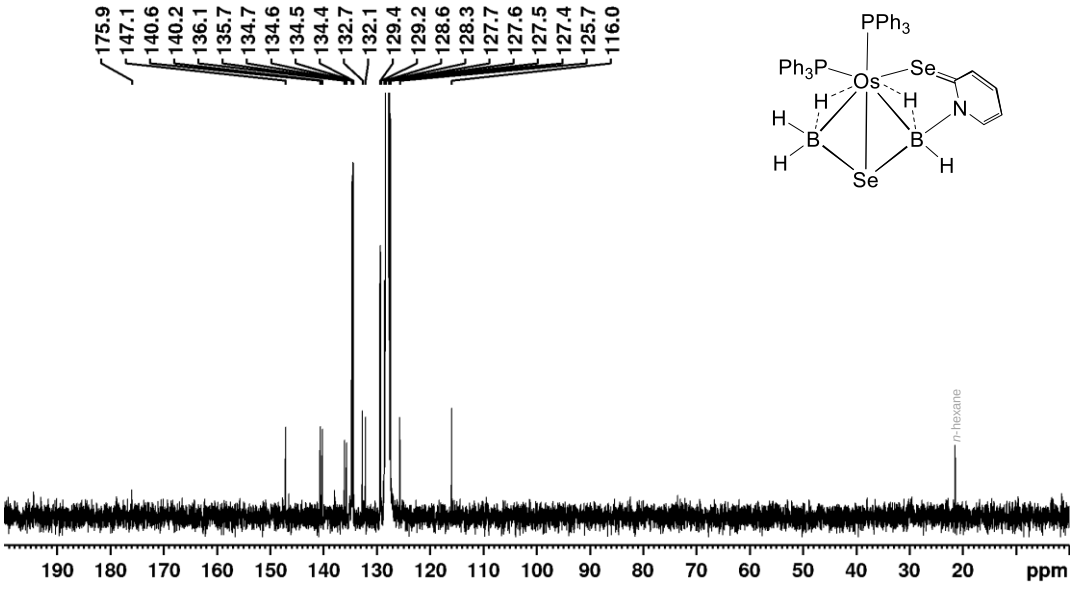


Figure S48. $^{13}C\{^{1}H\}$ NMR spectrum of **10** in C_6D_6.

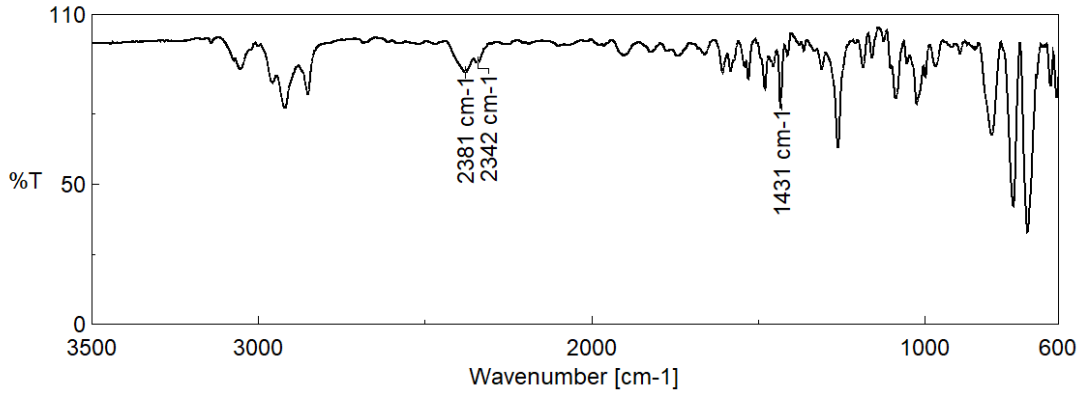


Figure S49. IR spectrum of **10**.

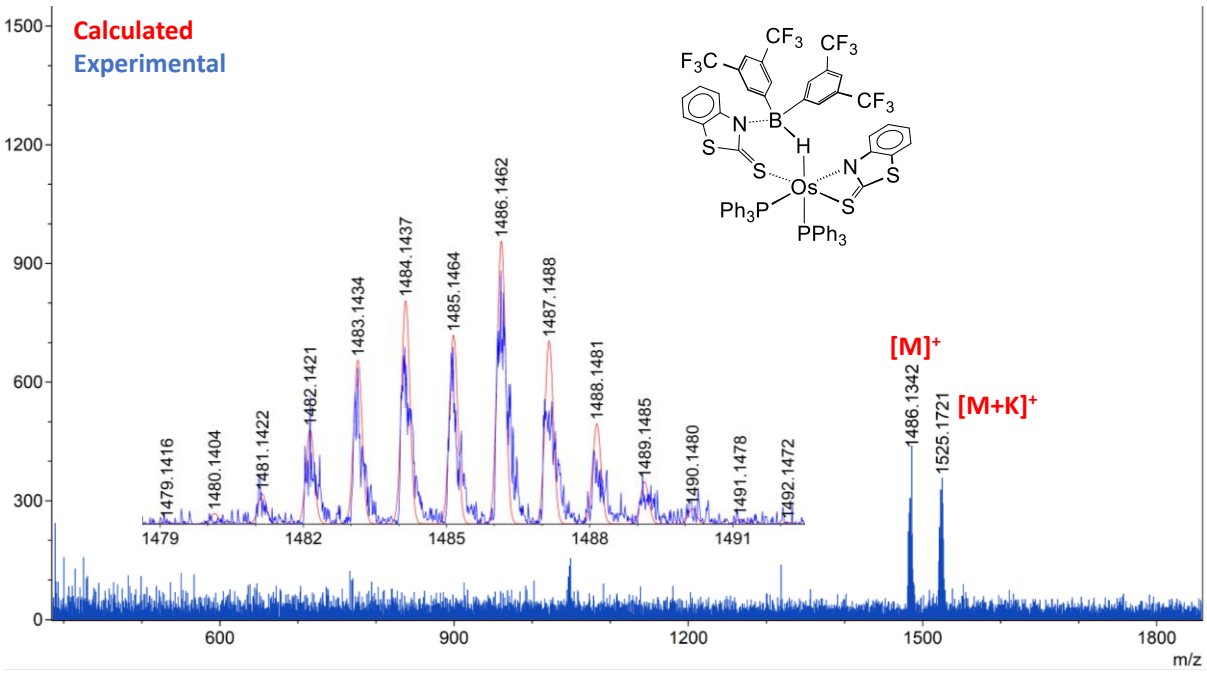


Figure S50. ESI-MS spectrum of **11**.

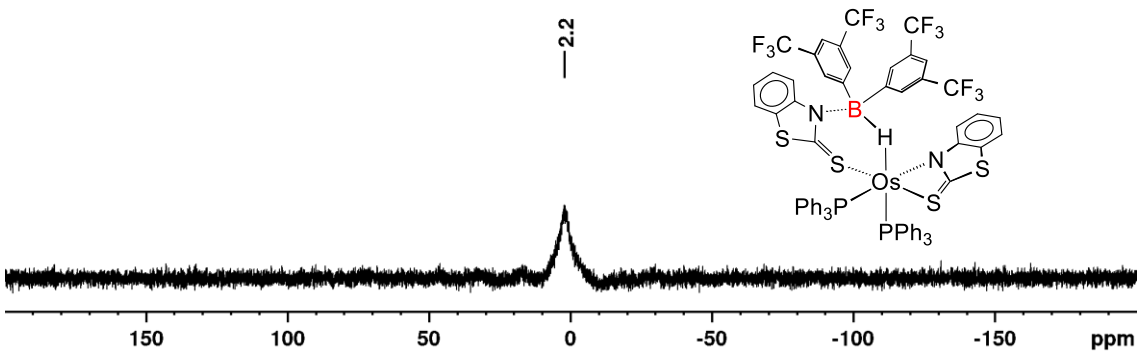


Figure S51. $^{11}B\{^{1}H\}$ NMR spectrum of **11** in C_6D_6.

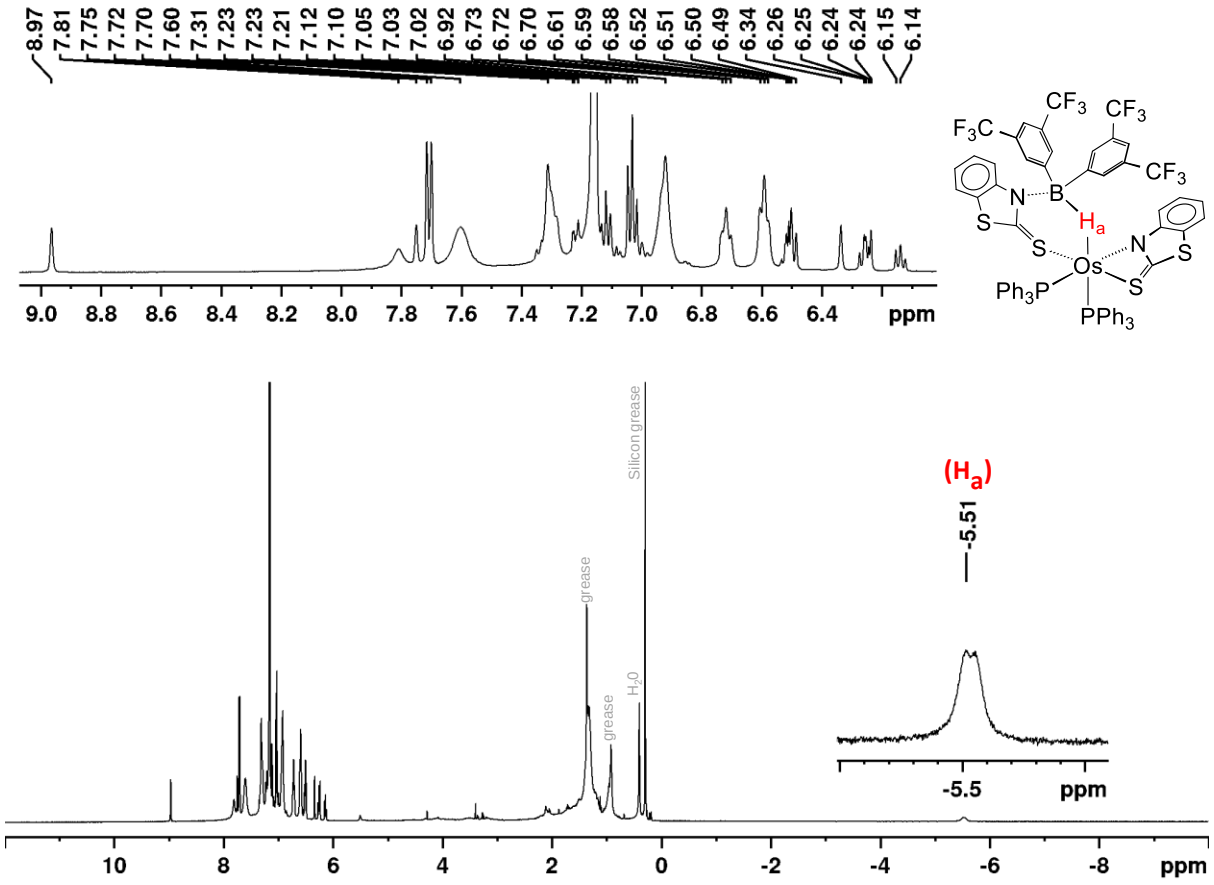


Figure S52. ^{1}H NMR spectrum of **11** in C_6D_6.

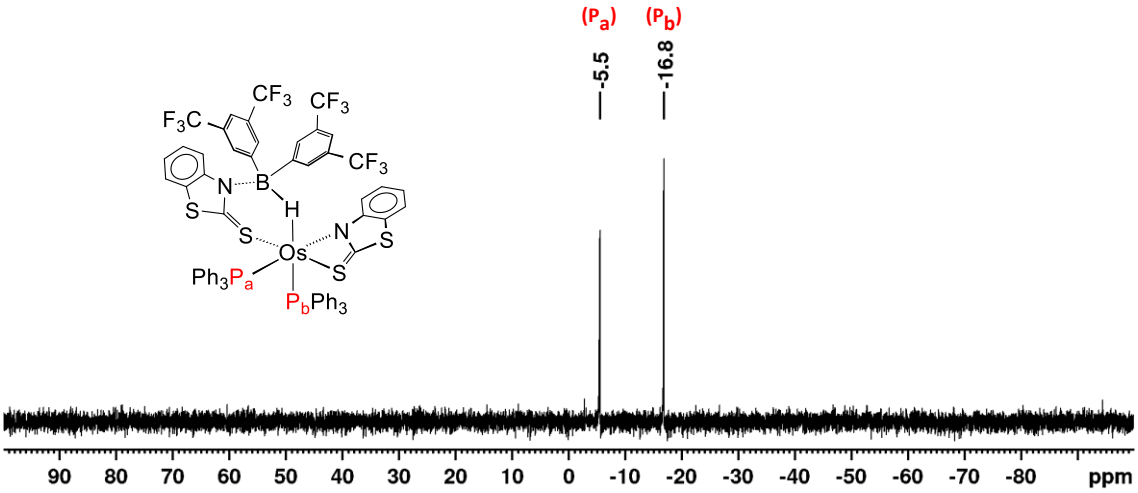


Figure S53. $^{31}P\{^{1}H\}$ NMR spectrum of **11** in C_6D_6.

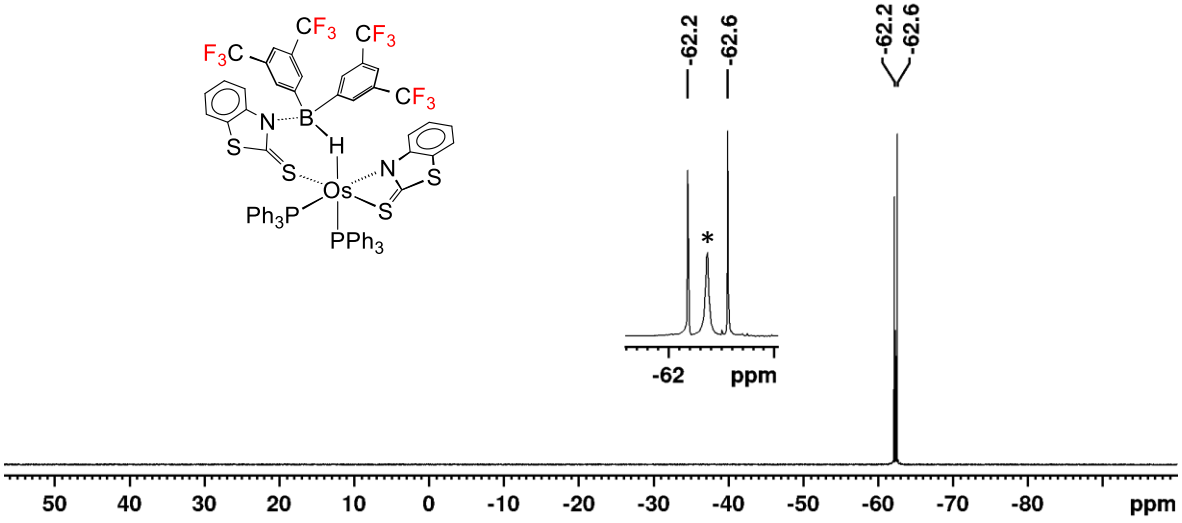


Figure S54. $^{19}F\{^{1}H\}$ NMR spectrum of **11** in C_6D_6.(* = inseparable impurity of BHR_2; R = $(CF_3)_2C_6H_3$)

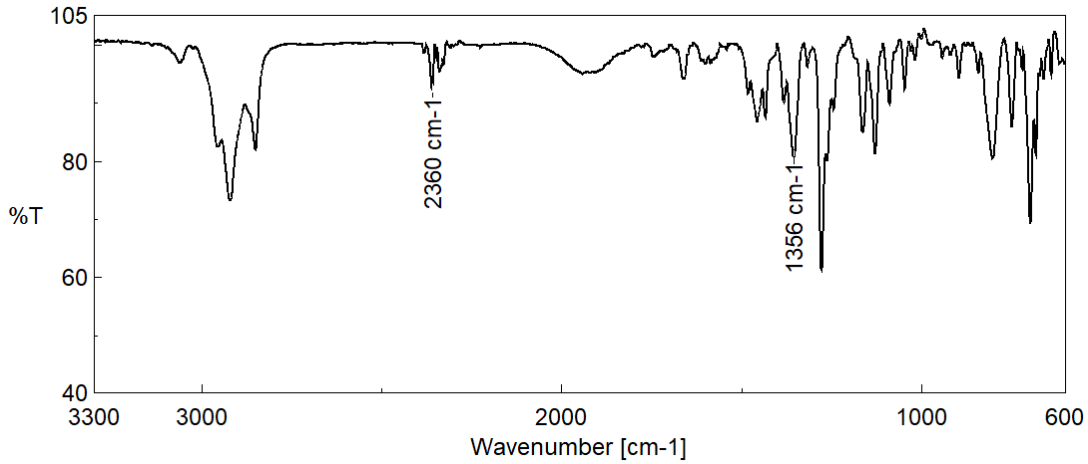


Figure S55. IR spectrum of **11**.

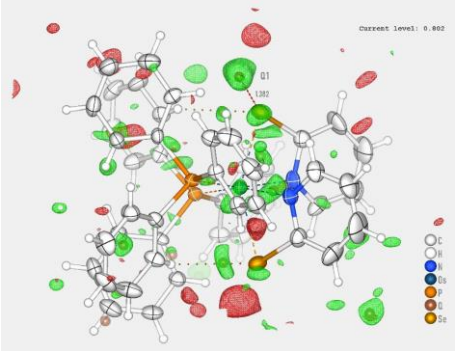
II.3 X-ray Analysis Details

Suitable X-ray quality crystals of complexes **1**, **2**, **4**, **5**, **6**, **7**, and **8** were obtained by slow diffusion of a hexane–CH_2Cl_2 solution at –4 °C, while crystals of complexes **10** and **11** were grown from hexane. Crystal data of **6** was obtained and integrated using a Bruker APEX-II CCD diffractometer with graphite monochromated Mo-Kα (λ = 0.71073 Å) radiation at 296(2) K. All other crystal data were obtained and integrated using a D8 VENTURE Bruker AXS diffractometer with PHOTON II detector with graphite monochromated Mo-Kα (λ = 0.71073 Å) radiation at 156(2) K for **1**, 200(2) K for **2**, 269(2) K for **4**, 150(2) K for **5**, 296(2) K for **6**, 150(2) K for **7**, 150(2) K for **8**, 301(2) K for **10**, 150(2) K for **11**. All the structures were solved using SHELXT-2018 and SHELXS-97[4] and refined using SHELXL−2018, SHELXL-2014 and SHELXL-2019.[7] Note that for compounds **6, 7** and **11** highly disordered lattice solvent molecules were removed using the SQUEEZE procedure in PLATON. Using Olex2 all the molecular structures were drawn.[8] Powder X-ray diffraction (XRD) patterns were recorded on a Bruker D8 Advanced Powder X-ray diffractometer using Cu Kα (*λ* = 1.5418 Å) radiation. The monophasic nature of each of the three compounds **1** and **2** was verified by comparing their powder XRD patterns with those simulated, using the MERCURY program,[9] on the basis of their single-crystal X-ray structures. Crystallographic data have been deposited with the Cambridge Crystallographic Data Center as supplementary publication no CCDC- 2481059 (**1**), 2481057 (**2**), 2466501 (**4**), 2481058 (**5**), 2466499 (**6**), 2466503 (**7**), 2466500 (**8**), 2472949 (**10**), 2472945 (**11**). These data can be obtained free of charge from The Cambridge Crystallographic Data Centre via www.ccdc.cam.ac.uk/data_request/cif.

Crystal data for **1***:* $C_{41}H_{34}Cl_2NOsP_2Se$, M_r= 942.69, monoclinic, space group $P2_1/n$, a = 9.5636(5) Å, b = 15.3610(8) Å, c = 25.2268(15) Å, α = 90 °, β = 100.93 °, γ = 90 °, V = 3638.8(3) Å^3, Z = 4, ρ_{calcd} = 1.721 g/cm^3, μ = 4.771 mm^{-1}, F(000) = 1844, R_1 = 0.0336, wR_2 = 0.0724, 7566 independent reflections [2θ ≤ 52.7 °] and 867 parameters.

Crystal data for **2***:*$C_{28}H_{23}ClN_2OsPSe_2$, M_r= 802.02 , monoclinic, space group $P2_1/n$, a = 12.1253(9) Å, b = 18.0329(10) Å, c = 12.3810(9) Å, α = 90 °, β = 91.515(3) °, γ = 90 °, V = 2706.2(3) Å^3, Z =4, ρ_{calcd} = 1.968 g/cm^3, μ = 7.580 mm^{-1}, F(000) = 1524, R_1 = 0.0228, wR_2 = 0.0517, 6722 independent reflections [2θ ≤ 56.6 °] and 316 parameters.

Crystal data for **4***:* $C_{46}H_{38}N_2OsP_2Se_2$, M_r= 1028.84, monoclinic, space group $P2_1/c$, a= 19.9615(18) Å, b = 17.6882(17) Å, c= 11.2486(11) Å, α = 90 °, β = 95.367(3) °, γ = 90 °, V= 3954.3(6) Å^3, Z= 4, ρ_{calcd} = 1.728 g/cm^3, μ = 5.182 mm^{-1}, F(000) = 2008, R_1 = 0.0729, wR_2 = 0.1605, 105792 independent reflections [2θ ≤ 52.88 °] and 480 parameters.



Note: The complex **4** was having non-merohedral twinning. The twin matrix is: (1 0 0.332; 0 -1 0; 0 0 -1) and program PLATON was used to generate the twin matrix and make HKLF5 format data. The

heaviest peak at 1.38 angstrom from Se1 (as shown in above image) measured 7.9 e/Å^3 and gave an A-level alert in the checkcif. After the twinning was resolved and HKLF5 data generated, the difference peaks have considerably reduced in height. But such techniques won't ideally clean the effects of non-merohedral twinning. Some effects of insufficient twin separation are still shown as elevated difference peaks.

Crystal data for **5***:* $C_{28}H_{33}B_4N_2OsPSe_2$, M_r= 819.89, orthorhombic, space group Pna21, a = 15.3411(5) Å, b = 9.6807(3) Å, c = 20.0705(5) Å, α = 90 °, β = 90 °, γ = 90 °, V = 2980.72(15) Å^3, Z = 4, ρ_{calcd} = 1.827 g/cm^3, μ = 6.795 mm^{-1}, F(000) =1576, R_1 = 0.0218, wR_2 = 0.0449, 5802 independent reflections [2θ ≤ 52.00 °] and 373 parameters.

Crystal data for **6***:* $C_{28}H_{28}B_3N_2OsPSe_2$,0.5[$CH_2Cl_2$], M_r= 846.604, triclinic, space group P-1, a = 11.1170(3) Å, b = 15.9224(5) Å, c = 17.8651(5) Å, α = 78.0360(1) °, β = 81.905(1) °, γ = 84.906 (1) °, V = 3056.97(15) Å^3, Z = 4, ρ_{calcd} = 1.839 g/cm^3, μ = 6.713 mm^{-1}, F(000) = 1618.9 , R_1 = 0.0282, wR_2 = 0.0712, 10760 independent reflections [2θ ≤ 50.00 °] and 707 parameters.

Crystal data for **7***:* $C_{44}H_{31}B_2F_{12}N_2OsPSe_3$, C_7H_8, M_r= 1387.51, monoclinic, space group $P2_1/n$, a = 10.8616(7) Å, b = 30.8022(19) Å, c = 14.9297(10) Å, α = 90 °, β = 97.324(2) °, γ = 90 °, V = 4954.1(6) Å^3, Z =4, ρ_{calcd} = 1.860 g/cm^3, μ = 4.895 mm^{-1}, F(000) = 2680.0, R_1 = 0.0506, wR_2 = 0. 0.1149, 12305 independent reflections [2θ ≤ 56.58 °] and 779 parameters.

Crystal data for **8***:* $C_{10}H_{21}B_7N_2OsSe_2$, M_r= 593.08 , monoclinic, space group $P2_1/c$, a = 12.7281(8) Å, b = 9.4036(6) Å, c = 15.1331(9) Å, α = 90 °, β = 101.480(2) °, γ = 90 °, V = 1775.04(19) Å^3, Z =4, ρ_{calcd} = 2.219 g/cm^3, μ =11.275 mm^{-1}, F(000) = 1096, R_1= 0.0136, wR_2 = 0.0333, 3628 independent reflections [2θ ≤ 52.7780 °] and 247 parameters.

Crystal data for **10***:* $C_{41}H_{39}B_2NOsP_2Se_2$,$C_7H_8$ M_r= 1069.58, monoclinic, space group $P2_1/n$, a = 11.0801(15) Å, b = 11.7542(12) Å, c = 34.230(4) Å, α = 90 °, β = 90.696(5) °, γ = 90 °, V = 4457.7(9) Å^3, Z =4, ρ_{calcd} = 1.594 g/cm^3, μ = 4.599 mm^{-1}, F(000) = 2104, R_1 = 0.0308, wR_2 = 0.0785, 9111 independent reflections [2θ ≤ 52.7760 °] and 512 parameters.

Crystal data for **11***:* $C_{66}H_{45}BF_{12}N_2OsP_2S_4$,0.5[$C_7H_8$], M_r= 1531.29, triclinic, space group P-1, a = 20.9226(9) Å, b = 24.4708(12) Å, c = 24.7349(12) Å, α = 73.662(2) °, β = 84.215(2) °, γ = 87.098(2) °, V = 12087.5(10) Å^3, Z =6, ρ_{calcd} = 1.262 g/cm^3, μ = 1.789 mm^{-1}, F(000) = 4578, R_1 = 0.0622, wR_2 = 0.1864, 42528 independent reflections [2θ ≤ 50.00 °] and 2745 parameters.

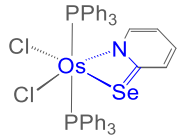
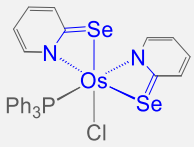
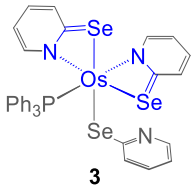
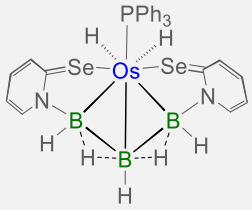
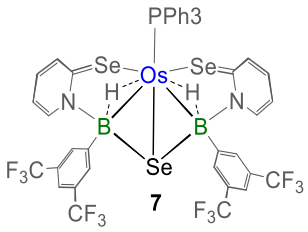
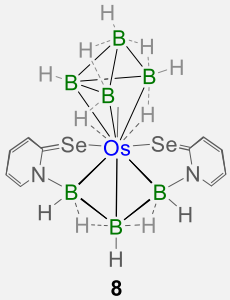
III Computational Details

All molecules were fully optimized with the Gaussian 16 program[10] using the gradient-corrected Becke–Lee–Yang–Parr (B3LYP) functional[11] in conjunction with the 6-31g(d)-LANL2DZ basis set from the EMSL Basis Set Exchange Library.[12] The model complexes were fully optimized starting from X-ray coordinates in the gaseous state (no solvent effect). Frequency calculations were carried out for the verification of the nature of the stationary state and to confirm the absence of any imaginary frequency which eventually confirmed the minima on the potential energy hypersurface for all structures. Furthermore, the gauge including atomic orbital (GIAO)[13] method was employed to compute the ^{11}B chemical shifts. Wiberg bond indices (WBI)[14] was generated from natural bond orbital analysis (NBO).[15] All the optimized geometries and orbital pictures were drawn by Chemcraft[16] visualization programs. Laplacian electronic distribution plots and Two-dimensional electron density and were generated using the Multiwfn package.[17]

Table S2. Selected geometrical parameters and Wiberg Bond Indices (WBI) of **1, 2, 3, 6, 7, 8, 10** and **11**.

1				**2**			
	Expt.	Cal.	WBI		Expt.	Cal.	WBI
Os1-N1	2.046(18)	2.118	0.559	Os1-N1	2.108(3)	2.105	0.593
Os1-Se1	2.5523(19)	2.663	0.781	Os1-N2	2.098(3)	2.135	0.531
Os1-Cl1	2.374(4)	2.404	0.820	Os1-Se1	2.4515(4)	2.533	0.943
Os1-Cl2	2.381(4)	2.428	0.788	Os1-Se2	2.5355(4)	2.642	0.804
Os1-P1	2.387(4)	2.493	0.699	Os1-Cl1	2.3681(9)	2.474	0.798
Os1-P2	2.385(5)	2.488	0.704	Os1-P1	2.2951(9)	2.443	0.784
3				**6**			
	Expt.	Cal.	WBI		Expt.	Cal.	WBI
Os1-N1	-	2.105	0.542	Os1-B1	2.132(7)	2.180	0.620
Os1-N2	-	2.147	0.528	Os1-B2	2.241(6)	2.256	0.477
Os1-Se1	-	2.559	0.899	Os1-B3	2.153(6)	2.221	0.605
Os1-Se2	-	2.680	0.733	B1-B2	1.877(8)	1.811	0.549
Os1-Se3	-	2.523	0.965	B2-B3	1.918(9)	1.848	0.512
Os1-P1	-	2.445	0.766	Os1-Se1	2.5263(5)	2.625	0.754
7				**8**			
	Expt.	Cal.	WBI		Expt.	Cal.	WBI
Os1-B1	2.234(7)	2.308	0.404	Os1-B1	2.416(3)	2.463	0.305
Os1-B2	2.289(7)	2.352	0.372	Os1-B2	2.210(3)	2.224	0.517
B1-Se3	2.093(7)	2.151	0.893	B1-B2	1.713(4)	1.721	0.710
B2-Se3	2.111(7)	2.154	0.881	Os1-B5	2.160(3)	2.165	0.576
Os1-Se1	2.4711(7)	2.548	0.815	Os1-B6	2.181(3)	2.201	0.520
Os1-Se2	2.5617(7)	2.551	0.809	Os1-B7	2.191(3)	2.221	0.503
Os1-Se3	2.5617(7)	2.633	0.579	B5-B6	1.860(4)	1.864	0.464
10				**11**			
	Expt.	Cal.	WBI		Expt.	Cal.	WBI
Os1-B1	2.374(5)	2.425	0.320	Os1-B1	3.001	3.104	0.129
Os1-B2	2.379(4)	2.478	0.318	Os1-S1	2.441(3)	2.508	0.667
B1-Se2	2.062(6)	2.080	0.950	Os1-N1	2.176(8)	2.220	0.432
B2-Se2	2.082(5)	2.087	0.949	Os1-S3	2.339(3)	2.410	0.707
Os1-Se1	2.4951(5)	2.591	0.786	Os1-P1	2.309(3)	2.373	0.833
Os1-Se2	2.5288(5)	2.620	0.652	Os1-P2	2.314(3)	2.394	0.818

Table S3. Calculated natural charge and HOMO-LUMO energy gap of **1, 2, 3, 6, 7, 8, 10** and **11**.

Complexes	Natural Charge (q)				ΔE_{H-L} (eV)
	q_{Os}	q_B	q_N	q_E (E=S, Se)	
 **1**	-0.570	-	-0.461	0.199	2.087
 **2**	-0.494	-	-0.434 (N1) -0.458 (N2)	0.286 (Se1) 0.199 (Se2)	2.060
 **3**	-0.706	-	-0.431 (N1) -0.456 (N2) -0.493 (N3)	0.253 (Se1) 0.174 (Se3) 0.245 (Se5)	2.088
 **6**	-1.831	0.290 (B1) -0.293 (B2) 0.316 (B3)	-0.573 (N1) -0.567 (N2)	0.334 (Se1) 0.364 (Se2)	3.159
 **7**	-1.523	0.419 (B1) 0.426 (B2)	-0.600 (N1) -0.594 (N2)	0.345 (Se1) 0.360 (Se2) 0.230 (Se3)	3.053
 **8**	-1.370	-0.211 (B1) -0.181 (B2) -0.043 (B3) -0.142 (B4) 0.387 (B5) -0.205 (B6) 0.321 (B7)	-0.564 (N1) -0.560 (N2)	0.388 (Se1) 0.328 (Se2)	3.343

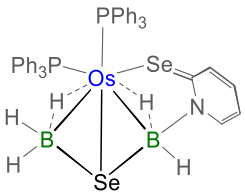
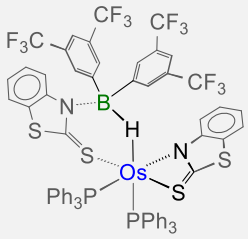
 **10**	-1.507	0.081 (B1) -0.262 (B2)	-0.562 (N1)	0.299 (Se1) 0.301 (Se2)	2.996
 **11**	-0.812	0.566	-0.529 (N1) -0.637 (N2)	0.088 (S1) 0.142 (S3)	3.379

Table S4. TD-DFT calculated energies (excitation energy (cm^{-1}), λ_{calc} (nm)), oscillator strength (*f*), and main composition of the first UV−vis electronic excitations for **1**, **2** and **3**.

Complex	No	Excitation Energy (cm^{-1})	Calc. Wavelength (nm) (*f*) [a]	Main electronic transition (% weight) [b]
1	1	14980	667 (0.030)	β-HOMO-2→ β-LUMO (79) β-HOMO→ β-LUMO (13)
	2	19804	504 (0.04)	β-HOMO-3→ β-LUMO (89)
2	1	16084	621 (0.052)	β-HOMO-3→ β-LUMO (66) β-HOMO-2→ β-LUMO (19)
	2	16316	612 (0.035)	β-HOMO-3→ β-LUMO (17) β-HOMO-2→ β-LUMO (76)
	3	23748	421 (0.012)	α-HOMO→ α-LUMO+1 (11) β-HOMO→ β-LUMO+1 (24) β-HOMO→ β-LUMO+2 (39)
3	1	14271	700 (0.052)	β-HOMO-1→ β-LUMO (91)
	2	19780	505 (0.037)	β-HOMO-5→ β-LUMO+2 (43) β-HOMO-4→ β-LUMO+2 (49)
	3	20130	496 (0.020)	β-HOMO-5→ β-LUMO+2 (49) β-HOMO-4→ β-LUMO+2 (32)

[a]Oscillator strength greater than 0.010 and [b]Components with greater than 10% contribution shown.

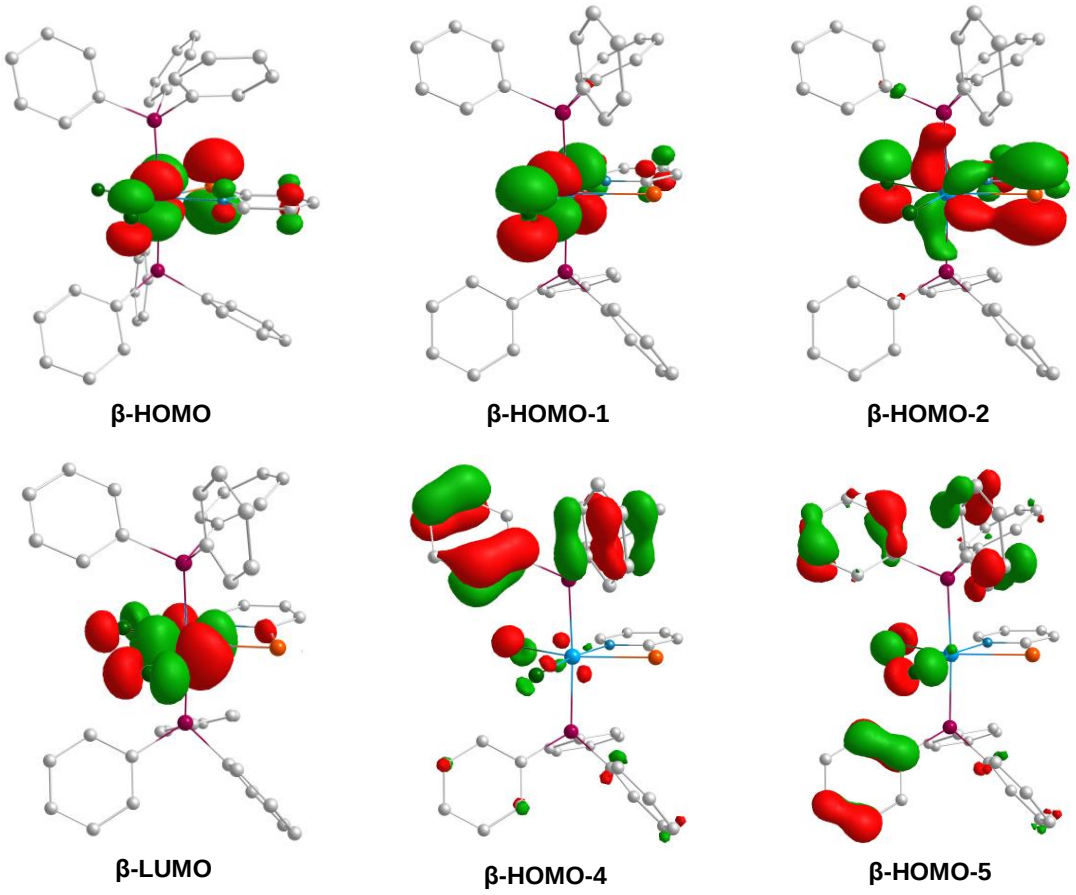


Figure S56. Selected molecular orbitals of **1** (isocontour values: ±0.043 [e.bohr^{-3}]$^{1/2}$).

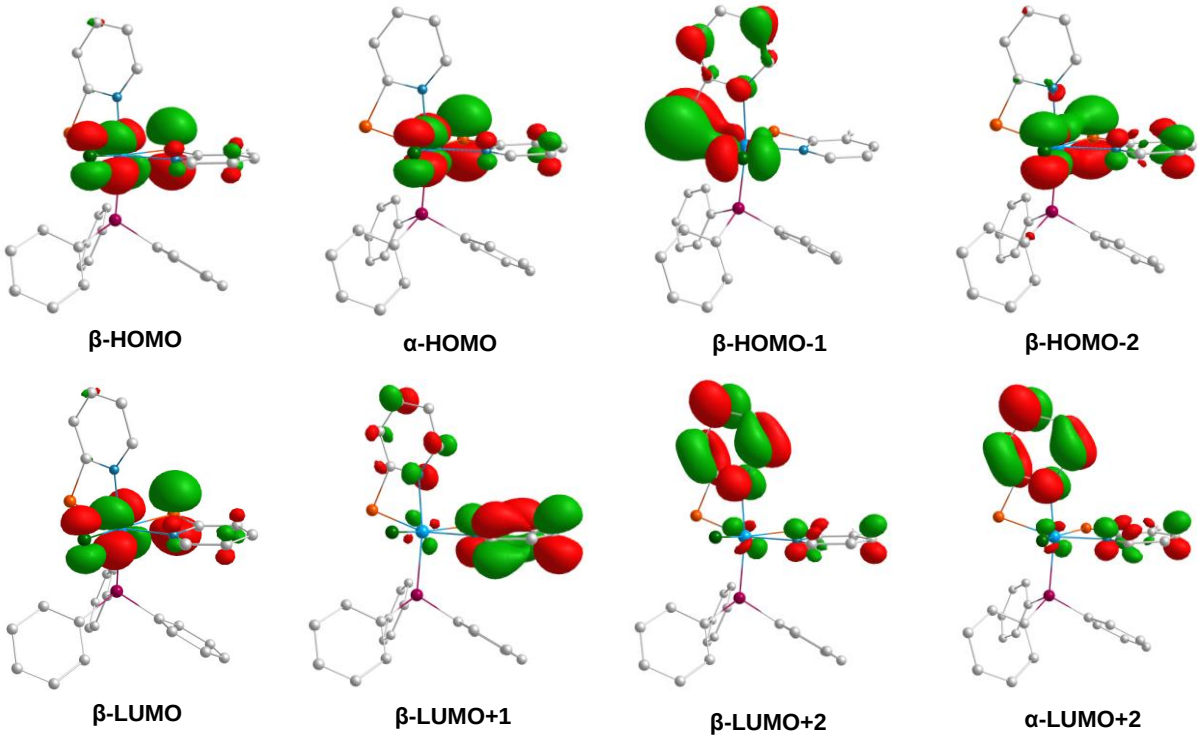
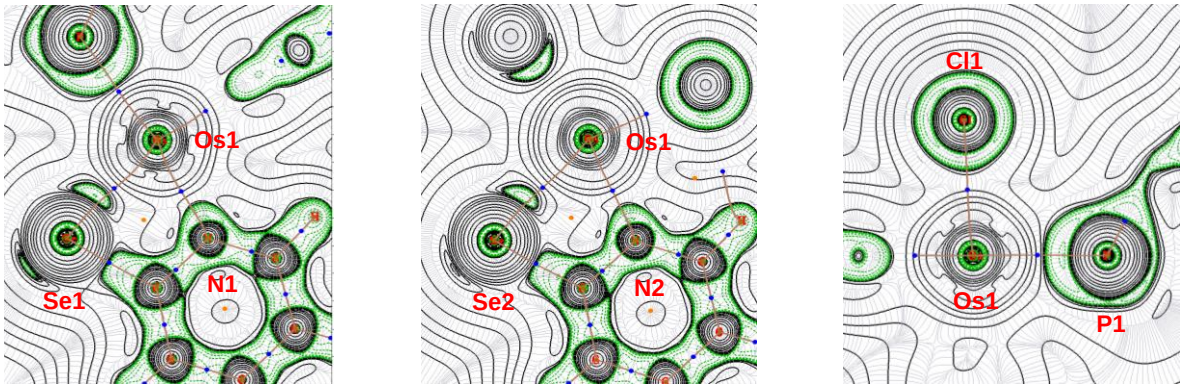
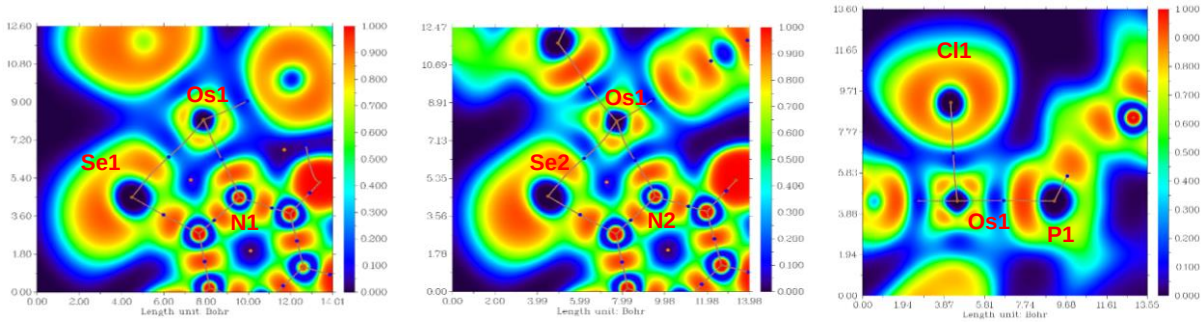


Figure S57. Selected molecular orbitals of **2** (isocontour values: ±0.043 [e.bohr^{-3}]$^{1/2}$).



(a) (b) (c)

Figure S58. Contour-line diagram of the Laplacian of electron density in the Os1-N1-Se1 (a) Os1-N2-Se2 (b) and Cl1-Os1-P1 (c) planes of **2**. The solid brown lines are bond paths, while blue spheres indicate the bond critical points and orange sphere indicates ring critical points. Area of charge concentration [$\nabla^2\rho(r)<0$] are indicated by solid lines and area of charge depletion [$\nabla^2\rho(r)>0$] are shown by dashed lines.



(a) (b) (c)

Figure S59. ELF plot in the Os1-N1-Se1 (a) Os1-N2-Se2 (b) and Cl1-Os1-P1 (c) planes of **2**.

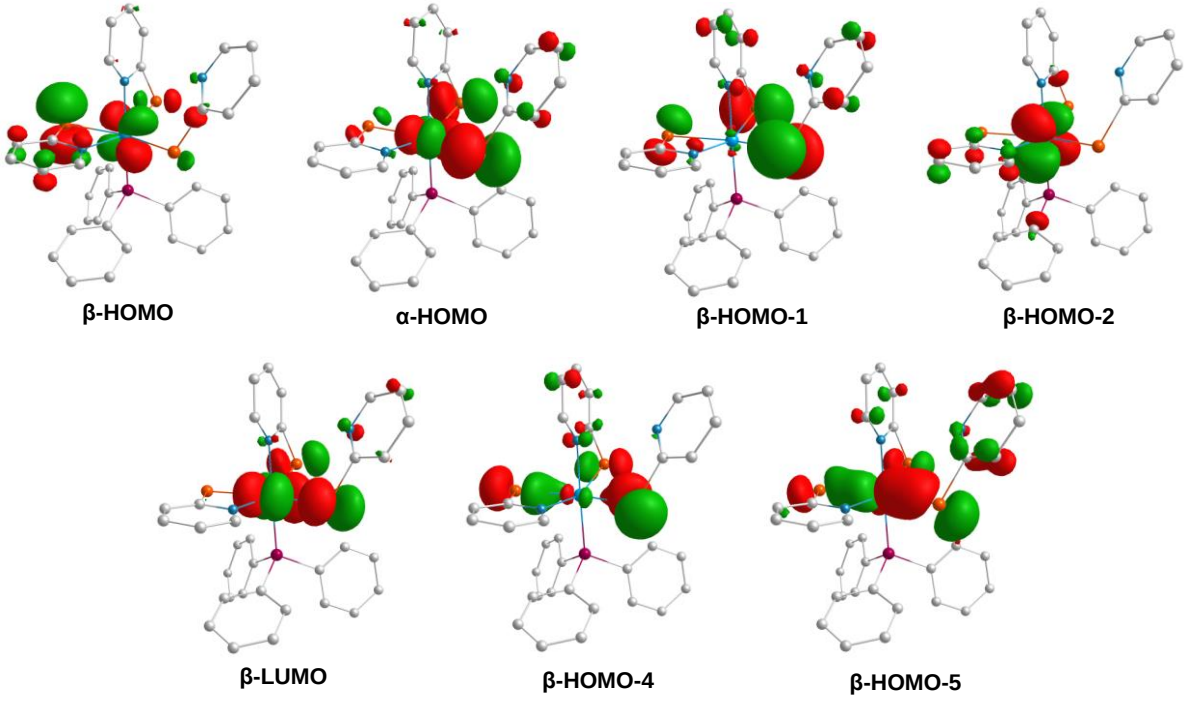
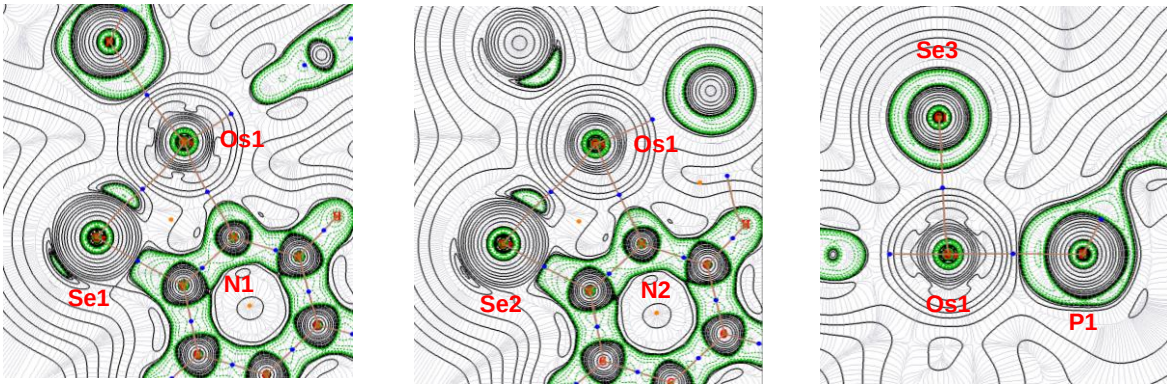
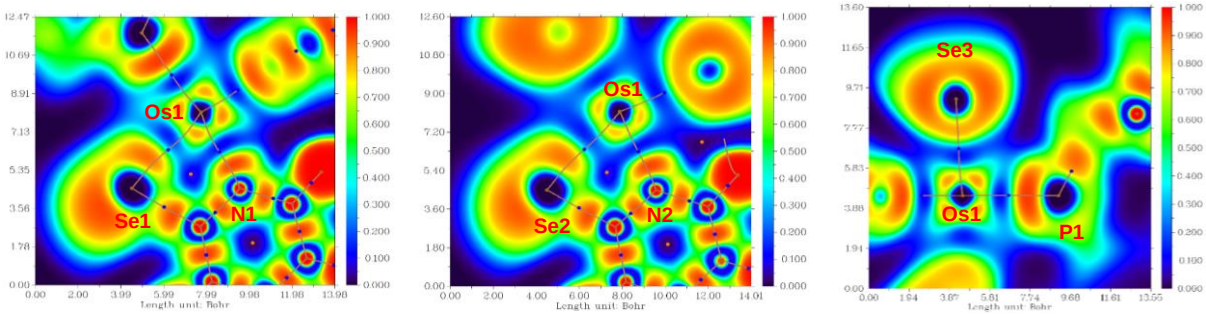


Figure S60. Selected molecular orbitals of **3** (isocontour values: ±0.043 [e.bohr^{-3}]$^{1/2}$).



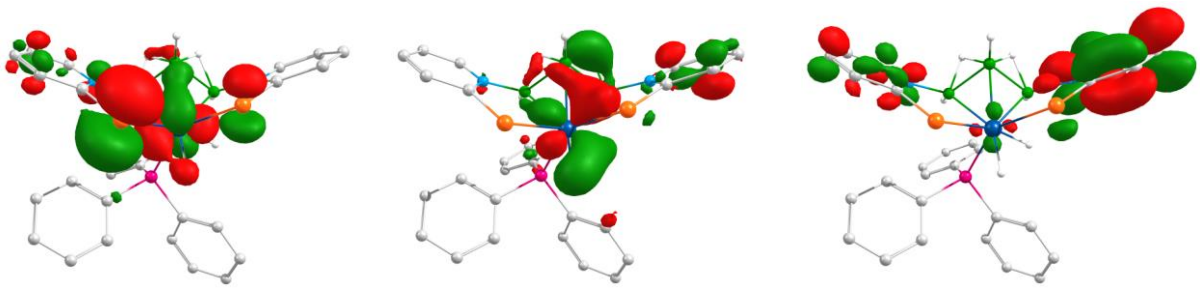
(a) (b) (c)

Figure S61. Contour-line diagram of the Laplacian of electron density in the Os1-N1-Se1 (a) Os1-N2-Se2 (b) and Se1-Os1-P1 (c) planes of **3**.



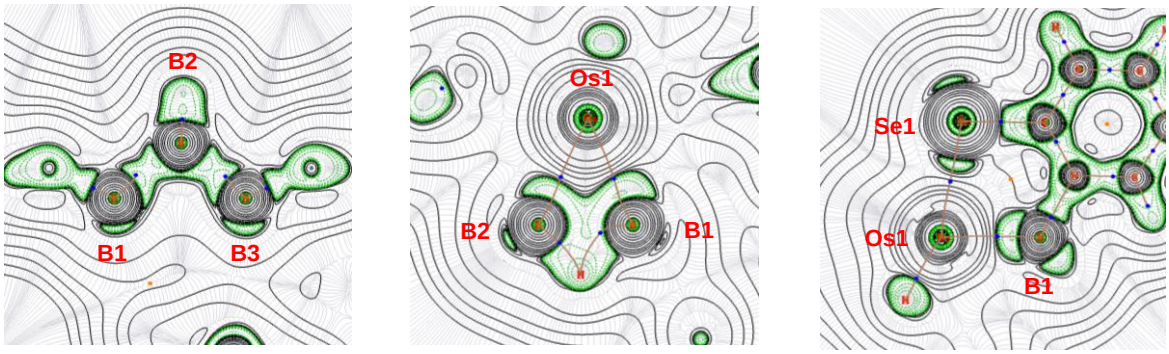
(a) (b) (c)

Figure S62. ELF plot in the Os1-N1-Se1 (a) Os1-N2-Se2 (b) and Se1-Os1-P1 (c) planes of **3**.



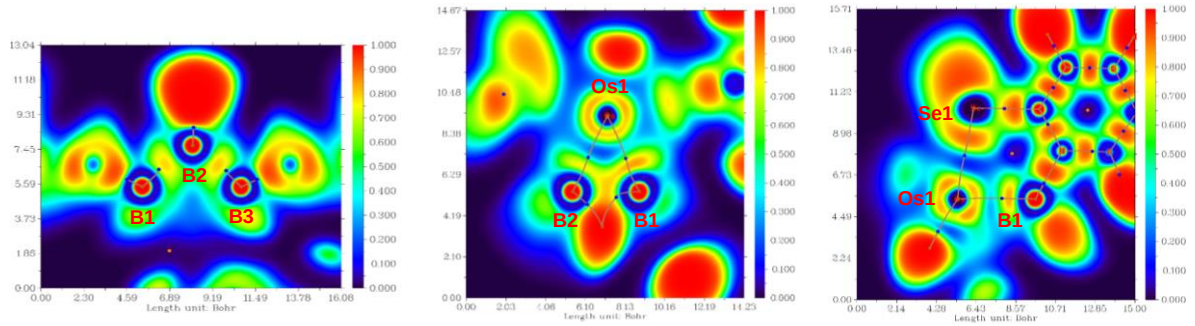
HOMO **HOMO-12** **LUMO**

Figure S63. Selected molecular orbitals of **6** (isocontour values: ±0.043 [e.bohr^{-3}]$^{1/2}$).



(a) (b) (c)

Figure S64. Contour-line diagram of the Laplacian of electron density in the B1-B2-B3 (a), Os1-B1-B2 (b) and Se1-Os1-B1 (c) planes of **6**.



(a) (b) (c)

Figure S65. ELF plot in the B1-B2-B3 (a), Os1-B1-B2 (b) and Se1-Os1-B1 (c) planes of **6**.

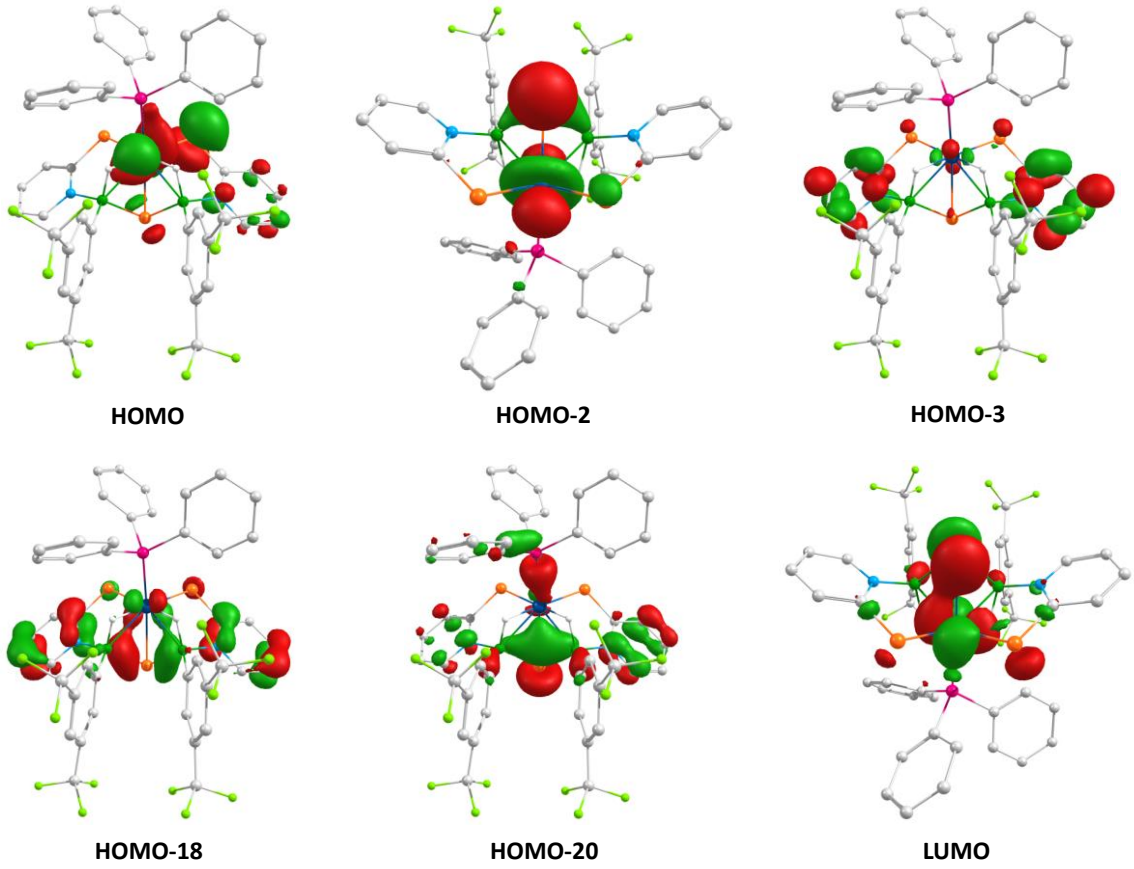


Figure S66. Selected molecular orbitals of **7** (isocontour values: ±0.043 [e.bohr^{-3}]$^{1/2}$).

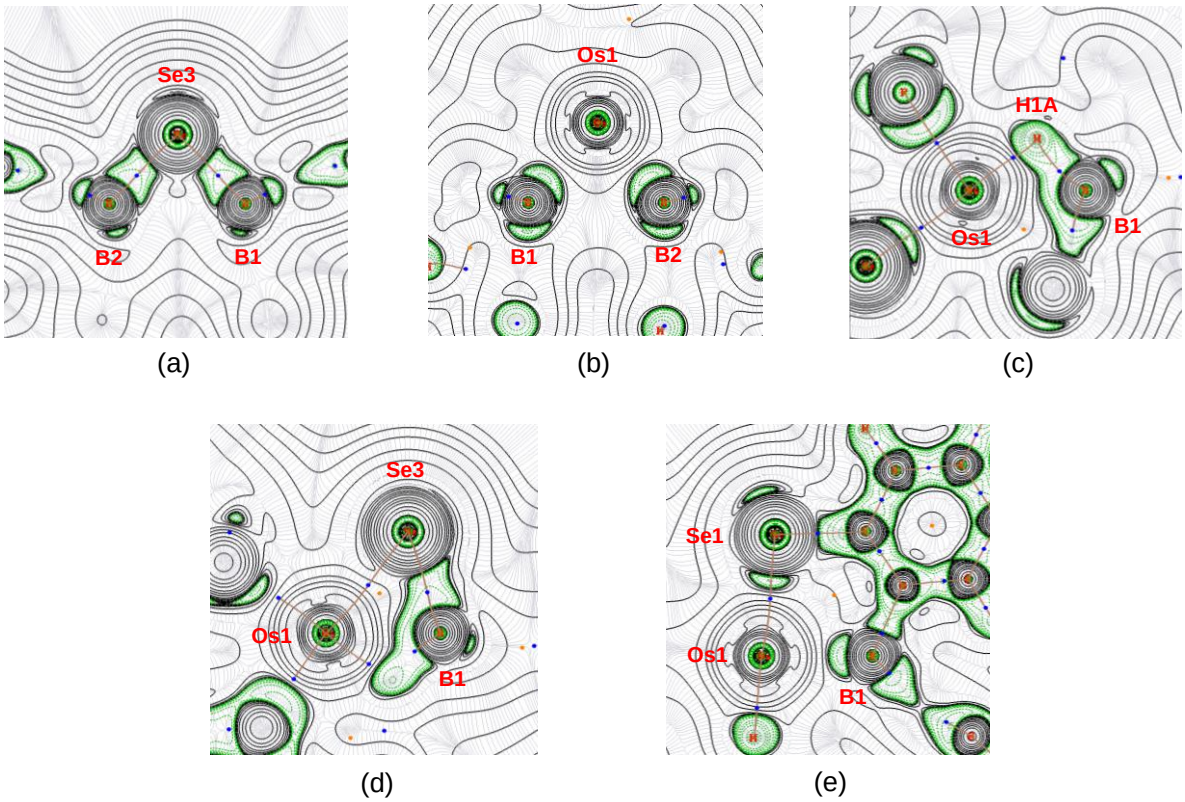


Figure S67. Contour-line diagram of the Laplacian of electron density in the B1-Se3-B2 (a), Os1-B1-B2 (b), Os1-H1A-B1 (c), Os1-Se3-B1 (b) and Os1-B1-Se1 (c) planes of **7**.

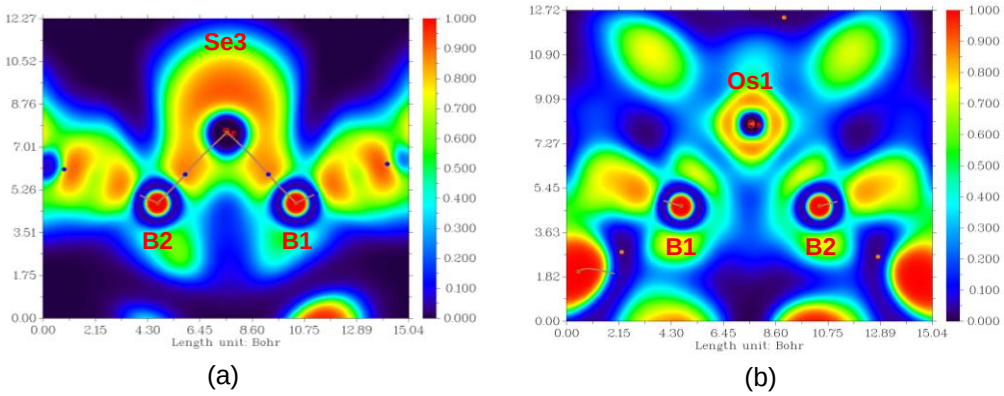


Figure S68. ELF plot in the B1-Se3-B2 (a) and Os1-B1-B2 (b) planes of **7**.

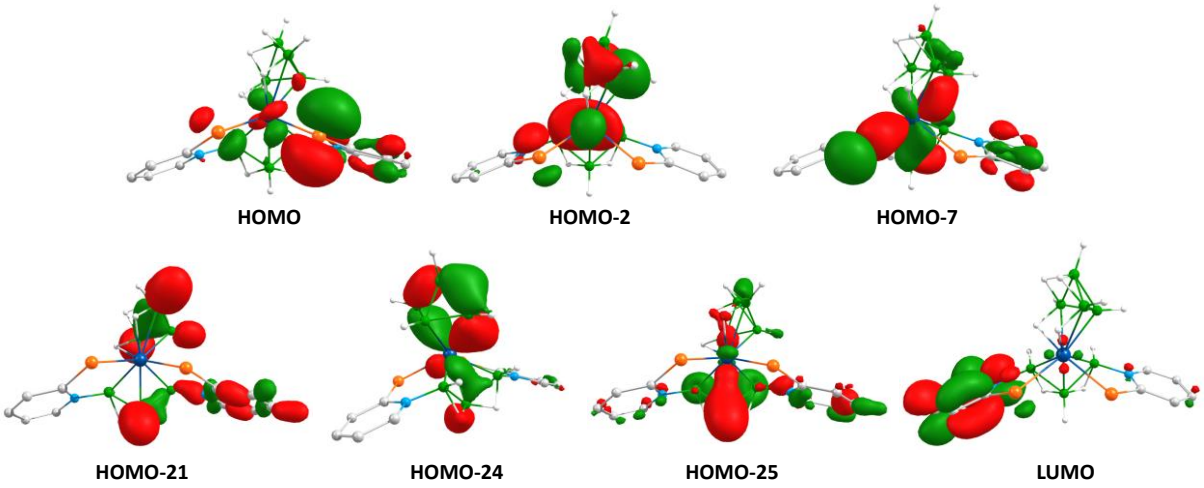


Figure S69. Selected molecular orbitals of **8** (isocontour values: ±0.043 [e.bohr^{-3}]$^{1/2}$).

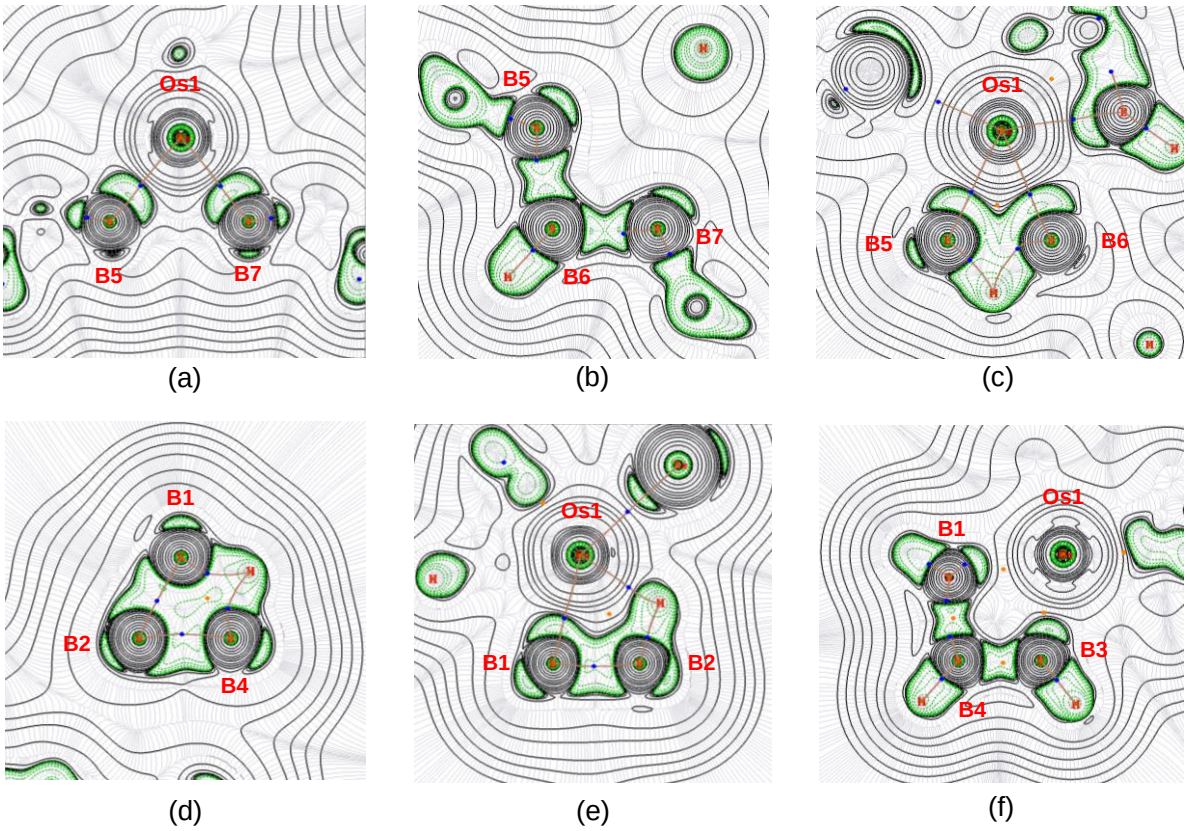


Figure S70. Contour-line diagram of the Laplacian of electron density in the Os1-B5-B7 (a) B5-B6-B7 (b), Os1-B5-B6 (c) B1-B2-B4 (d), Os1-B1-B2 (e) and Os1-B1-B4-B3 (f) planes of **8**.

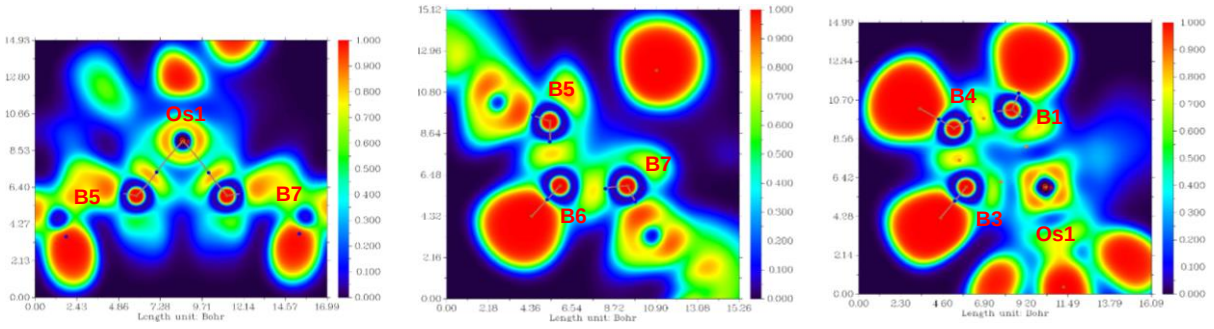


Figure S71. ELF plot in the Os1-B5-B7 (a) B5-B6-B7 (b) and Os1-B1-B4-B3 (c) planes of **8**.

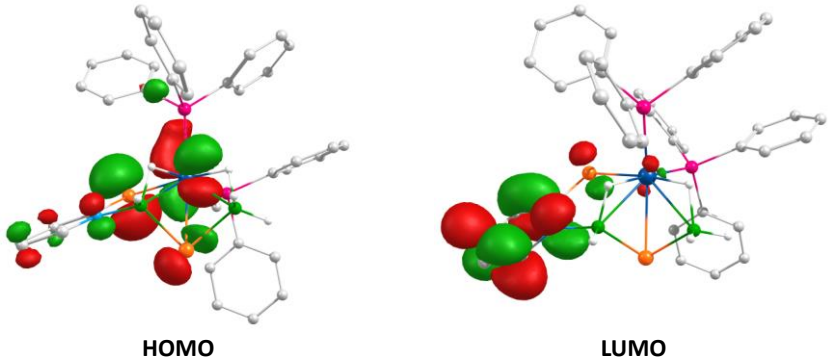


Figure S72. Selected molecular orbitals of **10** (isocontour values: ±0.043 [e.bohr^{-3}]$^{1/2}$).

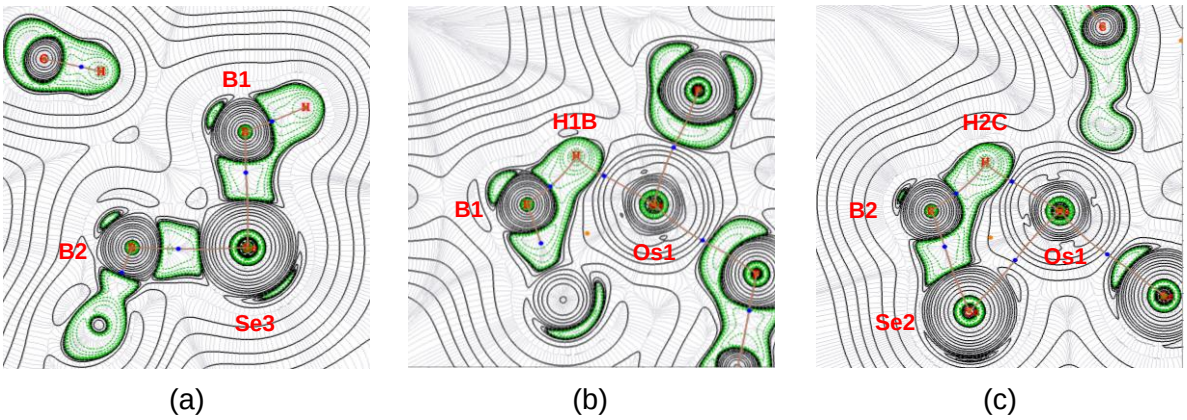


Figure S73. Contour-line diagram of the Laplacian of electron density in the B1-Se2-B2 (a) Os1-H1B-B1 (b) and Os1-H2C-B2-Se2 (c) planes of **10**.

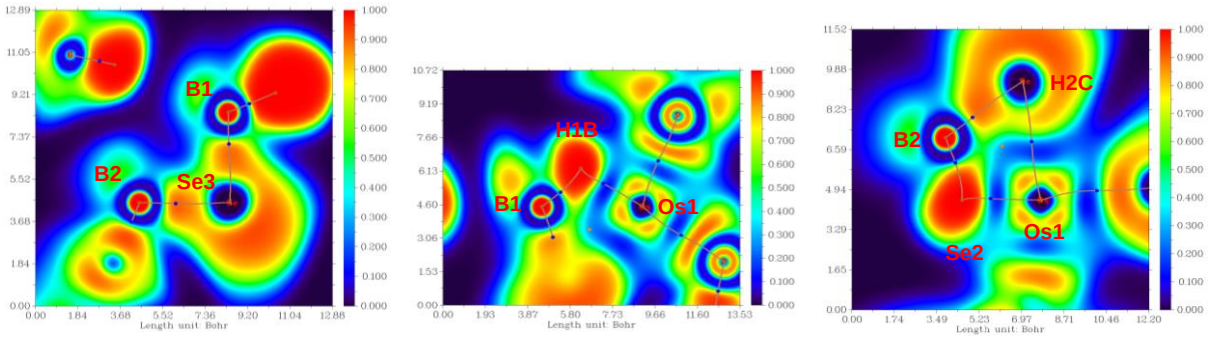


Figure S74. ELF plot in the B1-Se2-B2 (a) Os1-H1B-B1 (b) and Os1-H2C-B2-Se2 (c) planes of **10**.

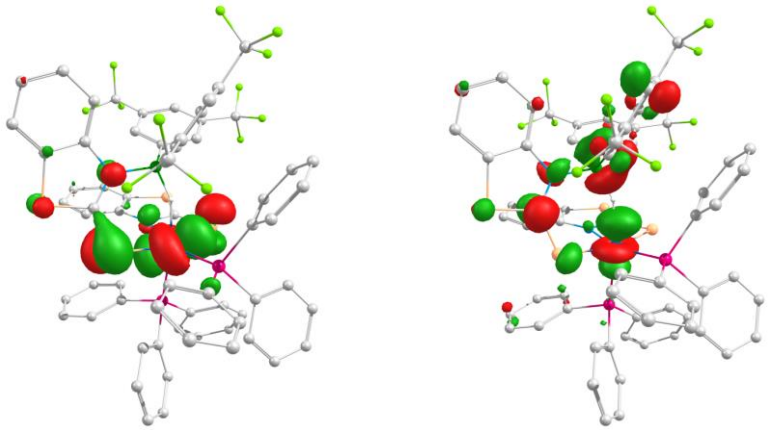


Figure S75. Selected molecular orbitals of **11** (isocontour values: ±0.043 [e.bohr^{-3}]$^{1/2}$).

IV Cartesian Coordinates of all Optimized Structures

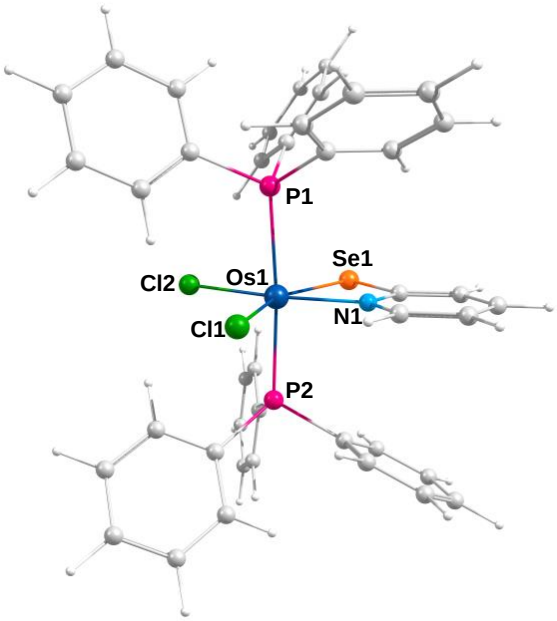


Figure S76. Optimized geometry of **1**.
Total energy = -2671.85535246 a.u.
Cartesian coordinates for the calculated structure **1** (in Å)

C	0.086480000	2.534786000	-1.647536000	H	-5.248327000	-2.237494000	4.643772000
C	-0.051734000	2.534429000	0.686067000	C	-3.294659000	1.330082000	-0.376959000
C	-0.070019000	3.928985000	0.721285000	C	-3.217205000	1.893094000	-1.655998000
N	0.015537000	1.855675000	-0.486607000	C	-3.922595000	2.043680000	0.646476000
P	-2.477917000	-0.333675000	-0.055024000	C	-3.787950000	3.137164000	-1.912624000
C	-3.249057000	-1.476023000	-1.351453000	H	-2.718878000	1.356032000	-2.454844000
C	-4.387747000	-1.123782000	-2.079271000	C	-4.484682000	3.294006000	0.388680000
C	-2.690868000	-2.745856000	-1.524895000	H	-3.976706000	1.627666000	1.644223000
C	-4.960375000	-2.029277000	-2.973231000	C	-4.426233000	3.839856000	-0.891243000
H	-4.837218000	-0.146757000	-1.958407000	H	-3.732099000	3.556992000	-2.910868000
C	-3.268719000	-3.648479000	-2.413468000	H	-4.971577000	3.836701000	1.191369000
H	-1.807414000	-3.030349000	-0.968346000	H	-4.872133000	4.807668000	-1.092110000
C	-4.402680000	-3.293455000	-3.142374000	Os	0.007441000	-0.218558000	-0.059174000
H	-5.842731000	-1.741392000	-3.534285000	C	0.073959000	3.918189000	-1.679073000
H	-2.825273000	-4.629894000	-2.539460000	C	-0.010517000	4.625975000	-0.476471000
H	-4.847297000	-3.997000000	-3.837504000	H	-0.131415000	4.436504000	1.675391000
C	-3.362054000	-0.949646000	1.499423000	H	-0.027059000	5.710105000	-0.476794000
C	-2.664380000	-1.237296000	2.671746000	H	0.131467000	4.428585000	-2.631474000
C	-4.748007000	-1.143405000	1.466615000	Se	-0.085989000	1.290935000	2.132828000
C	-3.343005000	-1.699164000	3.799969000	H	0.153646000	1.930591000	-2.541583000
H	-1.591980000	-1.125084000	2.702852000	Cl	-0.027352000	-0.677172000	-2.443251000
C	-5.425192000	-1.599573000	2.593987000	Cl	0.044954000	-2.445665000	0.844264000
H	-5.306148000	-0.946077000	0.559744000	P	2.498471000	-0.313677000	-0.055335000
C	-4.722761000	-1.877252000	3.766337000	C	3.329562000	-0.151502000	1.635277000
H	-2.785483000	-1.924146000	4.702196000	C	2.819855000	-0.920332000	2.686771000
H	-6.499130000	-1.743635000	2.552938000	C	4.456601000	0.642627000	1.854164000

C	3.428410000	-0.888356000	3.938072000
H	1.952551000	-1.548550000	2.525638000
C	5.059965000	0.678199000	3.112619000
H	4.877158000	1.232338000	1.050316000
C	4.547597000	-0.085659000	4.157075000
H	3.024199000	-1.490786000	4.743978000
H	5.933248000	1.301891000	3.269133000
H	5.017179000	-0.058628000	5.134067000
C	3.296061000	-1.910493000	-0.683167000
C	2.614183000	-2.760502000	-1.555266000
C	4.598238000	-2.235558000	-0.288093000
C	3.232149000	-3.916869000	-2.031324000
H	1.607641000	-2.523761000	-1.867413000
C	5.211514000	-3.391776000	-0.764514000
H	5.137409000	-1.595831000	0.398792000
C	4.529333000	-4.236015000	-1.639130000
H	2.689842000	-4.569540000	-2.706283000
H	6.219992000	-3.632736000	-0.446955000
H	5.004881000	-5.138477000	-2.007180000
C	3.329737000	1.004771000	-1.110662000
C	3.913374000	0.676941000	-2.336863000
C	3.315115000	2.342150000	-0.698929000
C	4.487532000	1.668933000	-3.131275000
H	3.924451000	-0.351254000	-2.674563000
C	3.899640000	3.329441000	-1.488492000
H	2.855399000	2.618576000	0.242560000
C	4.488501000	2.995551000	-2.707195000
H	4.936953000	1.399129000	-4.080454000
H	3.889683000	4.359922000	-1.151290000
H	4.942731000	3.764591000	-3.322043000

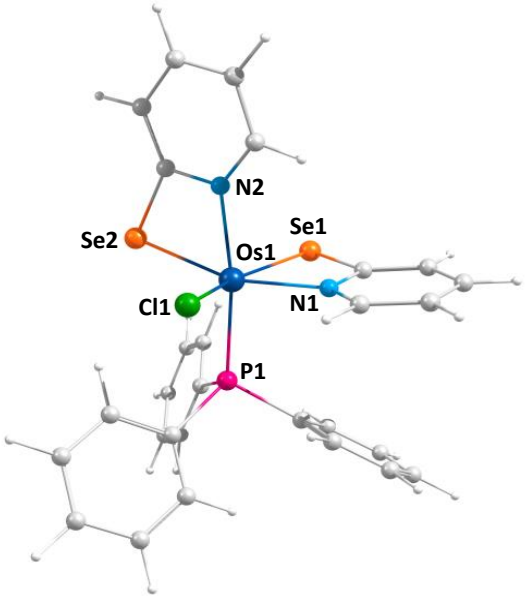


Figure S77. Optimized geometry of **2**.
Total energy = -1321.71260555 a.u.
Cartesian coordinates for the calculated structure **2** (in Å)

C	-3.205760000	1.672000000	0.080904000	H	3.047914000	-4.194166000	-1.832945000
C	-3.976137000	-0.375459000	-0.720562000	H	3.999754000	-3.482184000	2.293810000
C	-5.287093000	0.081510000	-0.713594000	H	4.056817000	-4.964839000	0.302556000
C	-0.890841000	-2.749260000	-1.619216000	C	2.463739000	1.081384000	-1.476029000
C	-1.100770000	-2.552331000	0.702638000	C	1.910040000	2.255320000	-1.993779000
C	-1.242788000	-3.933898000	0.843213000	C	3.684853000	0.622437000	-1.975721000
N	-0.919373000	-1.981422000	-0.513529000	C	2.574035000	2.963085000	-2.991925000
N	-2.959689000	0.404196000	-0.326758000	H	0.952159000	2.608982000	-1.635349000
P	1.561331000	0.222953000	-0.055291000	C	4.344395000	1.329367000	-2.981683000
Se	-1.498575000	2.465767000	0.538533000	H	4.129461000	-0.284900000	-1.587016000
C	2.267197000	1.171941000	1.424480000	C	3.791738000	2.502184000	-3.489708000
C	3.582486000	1.649461000	1.397310000	H	2.130043000	3.868804000	-3.389172000
C	1.487929000	1.401614000	2.559550000	H	5.289965000	0.960519000	-3.363732000
C	4.108774000	2.335129000	2.489163000	H	4.303388000	3.051054000	-4.272512000
H	4.199400000	1.498201000	0.520721000	Os	-0.873921000	0.134506000	-0.231939000
C	2.014799000	2.090286000	3.652534000	C	-1.033627000	-4.124859000	-1.547866000
H	0.467971000	1.048297000	2.593636000	C	-1.205373000	-4.724939000	-0.295283000
C	3.325818000	2.556904000	3.621453000	H	-1.383436000	-4.359169000	1.828573000
H	5.128817000	2.701047000	2.450756000	H	-1.314542000	-5.800593000	-0.213565000
H	1.393774000	2.262776000	4.524368000	H	-1.008904000	-4.713072000	-2.456088000
H	3.734354000	3.094689000	4.469796000	C	-4.490958000	2.198361000	0.114577000
C	2.420720000	-1.448136000	0.074781000	C	-5.545815000	1.385417000	-0.289767000
C	2.440242000	-2.293887000	-1.039850000	H	-4.649779000	3.216817000	0.444382000
C	2.981560000	-1.888847000	1.274939000	H	-6.560861000	1.765810000	-0.276990000
C	3.030728000	-3.552413000	-0.959058000	H	-6.085320000	-0.573419000	-1.037963000
H	2.006293000	-1.966185000	-1.977305000	H	-3.718231000	-1.374347000	-1.047565000
C	3.566214000	-3.152887000	1.356031000	Se	-1.177106000	-1.183256000	2.038235000
H	2.966476000	-1.249368000	2.148273000	H	-0.762919000	-2.218162000	-2.553786000
C	3.596596000	-3.985034000	0.239942000	Cl	-0.874953000	0.463832000	-2.684065000

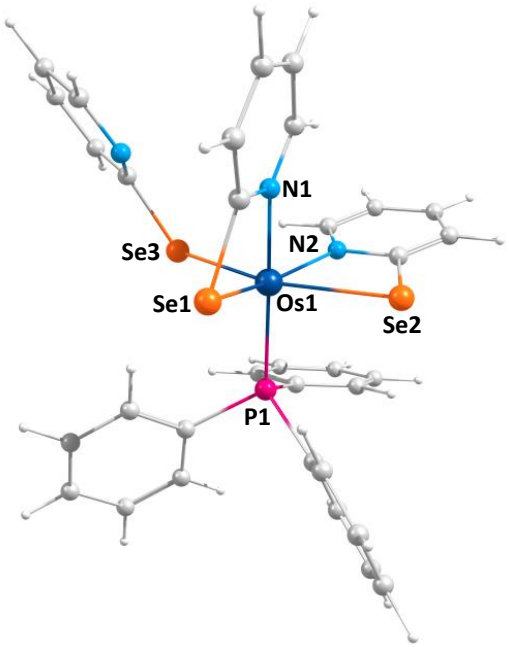


Figure S78. Optimized geometry of **3**.

Total energy = -1563.71855705 a.u.

Cartesian coordinates for the calculated structure **3** (in Å)

C	-3.207054000	1.683105000	1.206243000
C	-5.166496000	1.630842000	0.024381000
C	-5.880212000	2.253909000	1.041549000
C	-2.597724000	-0.729596000	-1.919978000
C	-3.104417000	-2.000052000	-0.034333000
C	-4.288667000	-2.438038000	-0.608013000
C	0.413890000	-2.907144000	0.835874000
C	-0.180595000	-1.658763000	2.721913000
C	0.046154000	-2.740127000	3.556099000
N	-0.006690000	-1.739200000	1.388529000
N	-2.277786000	-1.163844000	-0.677736000
N	-3.859379000	1.349189000	0.097096000
P	1.843208000	0.623964000	-0.046272000
Se	-1.269528000	1.403635000	1.354947000
Se	0.582548000	-2.716460000	-1.055340000
Se	-1.191151000	0.448533000	-2.505895000
C	2.675733000	0.268753000	1.605470000
C	2.553336000	1.164278000	2.671736000
C	3.314126000	-0.958344000	1.814541000
C	3.085228000	0.847881000	3.921495000
H	2.045477000	2.110804000	2.532152000
C	3.846672000	-1.270769000	3.063551000
H	3.398043000	-1.671847000	1.003348000
C	3.738070000	-0.366913000	4.119274000
H	2.988673000	1.554669000	4.738274000
H	4.345066000	-2.222605000	3.209920000
H	4.155899000	-0.609716000	5.089967000
C	3.129690000	0.091172000	-1.331478000
C	2.698114000	-0.221504000	-2.622002000
C	4.498627000	0.093516000	-1.052638000
C	3.622128000	-0.524901000	-3.619544000
H	1.640038000	-0.236255000	-2.850803000
C	5.422050000	-0.219852000	-2.049234000
H	4.855802000	0.330020000	-0.058374000
C	4.986218000	-0.528078000	-3.335907000
H	3.271325000	-0.766830000	-4.616552000
H	6.481386000	-0.220671000	-1.816916000
H	5.704474000	-0.770651000	-4.111140000
C	1.997522000	2.503405000	-0.229938000
C	0.945358000	3.236496000	-0.779620000
C	3.186085000	3.160694000	0.105565000
C	1.080039000	4.609048000	-0.991560000
H	0.017855000	2.742791000	-1.034996000
C	3.315713000	4.531946000	-0.097510000
H	4.015957000	2.610252000	0.531250000
C	2.261677000	5.259904000	-0.649506000
H	0.253841000	5.165362000	-1.419825000
H	4.240818000	5.029569000	0.171863000
H	2.363511000	6.327379000	-0.810869000
Os	-0.387010000	-0.363085000	-0.215601000
C	0.654546000	-4.036824000	1.624461000
C	0.469459000	-3.950373000	2.995073000
H	-0.517910000	-0.700897000	3.095165000
H	-0.104214000	-2.633505000	4.622526000
H	0.650509000	-4.814116000	3.625323000
H	0.979267000	-4.954154000	1.150052000
C	-3.837231000	2.309466000	2.295542000
C	-5.190638000	2.598154000	2.205540000
H	-5.660292000	1.341658000	-0.899502000
H	-6.937160000	2.461773000	0.926182000
H	-5.702202000	3.083434000	3.029928000
H	-3.268383000	2.561328000	3.182120000
C	-3.760267000	-1.137328000	-2.564013000
C	-4.618129000	-2.000922000	-1.892810000
H	-3.976998000	-0.778895000	-3.562055000
H	-5.533971000	-2.335981000	-2.366864000
H	-4.935403000	-3.109721000	-0.058449000
H	-2.797399000	-2.313505000	0.955249000

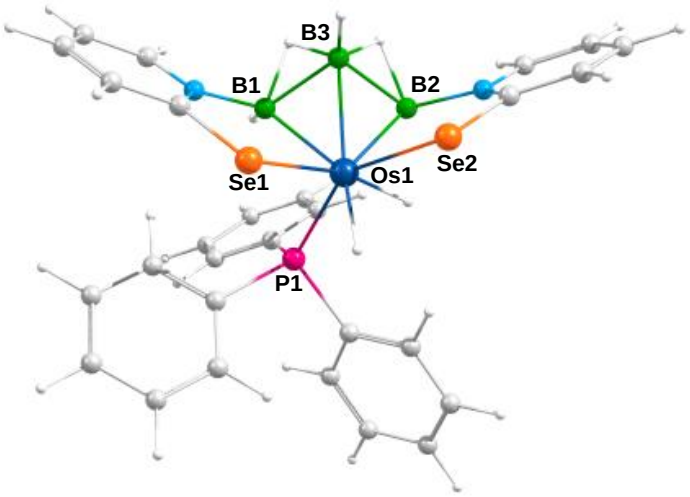


Figure S79. Optimized geometry of **6**.
Total energy = -1720.43700776 a.u.
Cartesian coordinates for the calculated structure **6** (in Å)

B	-0.260968000	-1.282977000	1.554953000
B	2.107100000	0.202869000	1.454588000
N	-1.115101000	-2.580552000	1.279550000
C	-1.788293000	-3.131756000	2.330726000
C	-2.497521000	-4.298133000	2.224451000
C	-2.527312000	-4.949086000	0.972585000
C	-1.865201000	-4.392224000	-0.093393000
C	-1.150662000	-3.178907000	0.053428000
N	3.624151000	0.386175000	1.072209000
C	4.452916000	0.963439000	1.988306000
C	5.800444000	1.099087000	1.784741000
C	6.344885000	0.615787000	0.577421000
C	5.515222000	0.041687000	-0.354732000
C	4.126286000	-0.069804000	-0.113358000
C	-2.674859000	0.522235000	-0.917052000
C	-3.431214000	-0.522344000	-0.369444000
C	-4.517856000	-1.054037000	-1.058363000
C	-4.855379000	-0.565814000	-2.319494000
C	-4.097124000	0.456614000	-2.883299000
C	-3.017208000	0.998386000	-2.187267000
C	-0.938002000	2.829007000	-0.825114000
C	0.148715000	3.116903000	-1.656756000
C	0.276373000	4.372419000	-2.252989000
C	-0.679888000	5.358646000	-2.030503000
C	-1.768167000	5.083371000	-1.203200000
C	-1.893913000	3.832603000	-0.605515000
C	-1.829806000	1.718330000	1.620021000
C	-3.184050000	1.723415000	1.969273000
C	-3.596979000	2.216040000	3.209095000
C	-2.664827000	2.719701000	4.111245000
C	-1.312908000	2.727886000	3.767021000
C	-0.900313000	2.229123000	2.535629000
B	1.529268000	-1.552346000	1.500025000
Os	0.713010000	-0.256841000	-0.157093000
P	-1.172085000	1.152401000	-0.028861000
H	-0.798570000	-0.651208000	2.412299000
H	1.853593000	0.949958000	2.352670000
H	2.246247000	-0.931869000	2.333665000
H	2.080549000	-2.553181000	1.184491000
H	0.697681000	-1.817994000	2.425407000
H	1.362574000	1.222894000	0.057402000
H	-3.176291000	-0.926174000	0.602337000
H	-2.440961000	1.793553000	-2.642634000
H	-4.343441000	0.838260000	-3.868022000
H	-5.697482000	-0.983824000	-2.859731000
H	-5.094647000	-1.856855000	-0.612412000
H	0.152714000	2.227186000	2.285841000
H	-0.577385000	3.120418000	4.460726000
H	-2.986740000	3.104845000	5.072455000
H	-4.651825000	2.209767000	3.461699000
H	-3.929368000	1.352752000	1.278002000
H	0.896003000	2.358061000	-1.840153000
H	1.128328000	4.573299000	-2.893238000
H	-0.579131000	6.333857000	-2.494084000
H	-2.519531000	5.843313000	-1.018658000
H	-2.743584000	3.643865000	0.038553000
H	-1.720174000	-2.577334000	3.257226000
H	-3.013865000	-4.697285000	3.086913000
H	-3.073781000	-5.877586000	0.851313000
H	-1.877930000	-4.860283000	-1.068873000
H	5.900946000	-0.328607000	-1.295525000
H	7.407769000	0.702248000	0.382227000
H	6.416241000	1.568394000	2.539876000
H	3.965505000	1.306890000	2.890410000
H	0.431626000	0.358790000	-1.676305000
Se	-0.291056000	-2.358136000	-1.397595000
Se	2.961554000	-0.802593000	-1.396017000

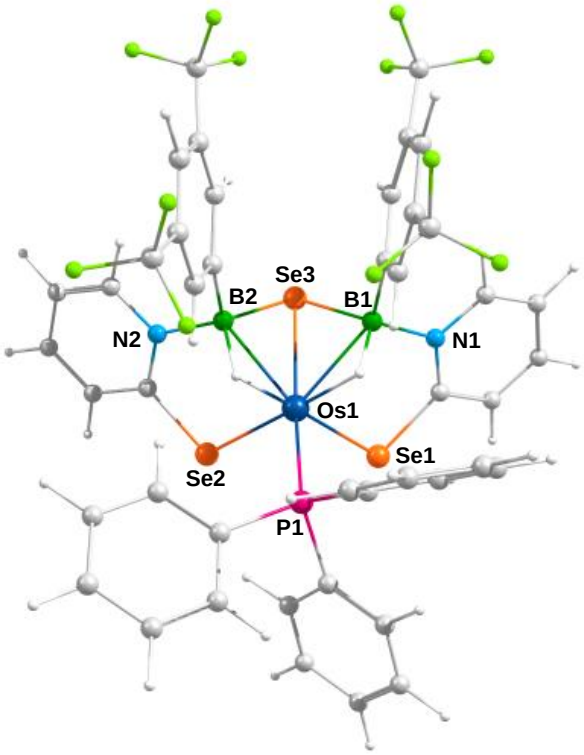


Figure S80. Optimized geometry of **7**.
Total energy = -3513.88669676 a.u.
Cartesian coordinates for the calculated structure **7** (in Å)

C	4.480716000	0.312858000	1.160999000
C	5.007463000	1.139911000	2.158042000
H	4.380603000	1.491246000	2.966549000
C	6.350195000	1.520636000	2.129325000
H	6.738657000	2.165203000	2.910205000
C	7.185804000	1.075762000	1.109709000
H	8.228295000	1.372855000	1.088146000
C	6.672710000	0.240341000	0.117796000
H	7.314954000	-0.119327000	-0.678355000
C	5.333727000	-0.136546000	0.143124000
H	4.958611000	-0.797666000	-0.628794000
C	-0.758201000	-3.070738000	-3.095165000
H	-1.758936000	-2.960089000	-2.712326000
C	-0.469542000	-4.015945000	-4.041041000
H	-1.259230000	-4.655496000	-4.411376000
C	0.855528000	-4.126517000	-4.501395000
H	1.123178000	-4.864559000	-5.248525000
C	1.805415000	-3.282203000	-3.984468000
H	2.836731000	-3.334443000	-4.307980000
C	1.470508000	-2.312912000	-3.012048000
C	2.975917000	-2.054531000	1.592208000
C	2.299887000	-3.086508000	0.938749000
H	1.627412000	-2.852854000	0.125877000
C	2.500434000	-4.417475000	1.305820000
H	1.965193000	-5.204012000	0.785903000
C	3.387090000	-4.733401000	2.330546000
H	3.543866000	-5.767181000	2.617233000
C	4.079791000	-3.712452000	2.981882000
H	4.779546000	-3.949141000	3.775607000
C	3.879798000	-2.385684000	2.613298000
H	4.434540000	-1.606319000	3.121957000
C	1.991918000	0.468144000	2.671513000
C	1.516249000	-0.305394000	3.734650000
H	1.582697000	-1.384992000	3.698505000
C	0.939888000	0.302878000	4.851428000
H	0.561051000	-0.313849000	5.658294000
C	0.840881000	1.688312000	4.924792000
H	0.390706000	2.158099000	5.791484000
C	1.307615000	2.469080000	3.866301000
H	1.226847000	3.549604000	3.910196000
C	1.867340000	1.864869000	2.744989000
H	2.210871000	2.481153000	1.921363000
C	1.472564000	3.595705000	-1.181882000
C	1.827517000	4.941866000	-1.429901000
H	2.854976000	5.146876000	-1.700757000
C	0.907000000	5.952795000	-1.323203000
H	1.191924000	6.980580000	-1.515697000
C	-0.408850000	5.627046000	-0.947719000
H	-1.174906000	6.381528000	-0.830581000
C	-0.721810000	4.313981000	-0.725795000
H	-1.717453000	4.031601000	-0.430958000
C	-1.654861000	-1.561415000	-0.774925000

C	-2.937397000	-1.376599000	-1.312275000
H	-3.051858000	-0.822593000	-2.236812000
C	-4.073978000	-1.885867000	-0.684593000
C	-3.971894000	-2.581938000	0.517765000
H	-4.855036000	-2.966319000	1.010663000
C	-2.711270000	-2.754436000	1.079311000
C	-1.575979000	-2.256268000	0.439985000
H	-0.612527000	-2.406271000	0.911901000
C	-1.754630000	1.723899000	0.100859000
C	-1.853966000	1.401728000	1.458641000
H	-0.962351000	1.132101000	2.013070000
C	-3.086423000	1.406435000	2.122609000
C	-4.251970000	1.761865000	1.456806000
H	-5.202394000	1.772496000	1.973059000
C	-4.173072000	2.086849000	0.102402000
C	-2.952507000	2.046716000	-0.562508000
H	-2.929647000	2.269958000	-1.622792000
C	-5.420680000	-1.752782000	-1.344702000
C	-2.563049000	-3.545284000	2.352283000
C	-3.135234000	1.061646000	3.587755000
C	-5.413811000	2.554162000	-0.611178000
B	-0.300950000	-1.101577000	-1.531243000
B	-0.329367000	1.783038000	-0.658873000
F	-1.513502000	-3.132377000	3.094702000
F	-2.356295000	-4.865653000	2.095124000
F	-3.662757000	-3.474285000	3.132458000
F	-5.687949000	-2.824637000	-2.140597000
F	-6.427675000	-1.681478000	-0.447783000
F	-5.503030000	-0.661271000	-2.133024000
F	-5.323396000	2.426376000	-1.950383000
F	-5.652401000	3.873152000	-0.364893000
F	-6.517951000	1.887641000	-0.211775000
F	-2.621536000	2.057690000	4.356431000
F	-2.420355000	-0.049897000	3.871358000
F	-4.394842000	0.846907000	4.026265000
N	0.176351000	-2.215856000	-2.571522000
N	0.177058000	3.285981000	-0.852237000
Os	1.428842000	0.224564000	-0.769786000
P	2.710179000	-0.273726000	1.131680000
Se	2.832695000	-1.179967000	-2.367489000
Se	2.828718000	2.291657000	-1.294637000
Se	-0.416060000	0.781475000	-2.564583000
H	0.461278000	1.174445000	0.283669000
H	0.537633000	-1.164455000	-0.400865000

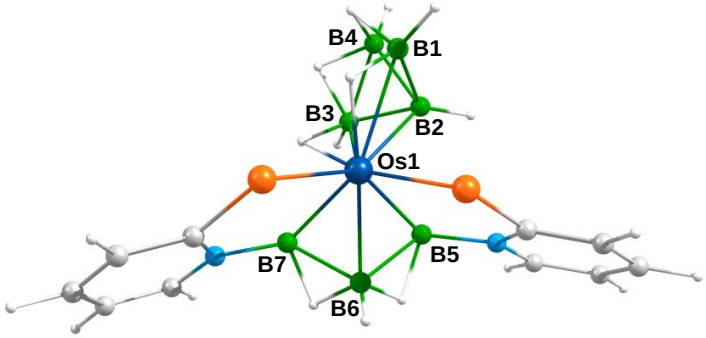


Figure S81. Optimized geometry of **8**.
Total energy = -786.983998268 a.u.
Cartesian coordinates for the calculated structure **8** (in Å)

C	-2.973832000	-0.978605000	-0.232252000
C	-4.225116000	-1.516674000	-0.597240000
H	-4.385887000	-1.790324000	-1.631740000
C	-5.221237000	-1.658938000	0.340902000
H	-6.187046000	-2.058336000	0.053192000
C	-4.981950000	-1.265890000	1.670963000
H	-5.744817000	-1.343221000	2.433542000
C	-3.743132000	-0.773594000	1.990802000
H	-3.481934000	-0.457376000	2.991049000
C	3.204902000	-0.657476000	-0.216862000
C	4.506636000	-1.086669000	-0.557808000
H	4.745651000	-1.217030000	-1.604932000
C	5.445640000	-1.311636000	0.420818000
H	6.446145000	-1.630880000	0.152254000
C	5.102549000	-1.109748000	1.772000000
H	5.816737000	-1.261005000	2.569804000
C	3.825608000	-0.708667000	2.063725000
H	3.487844000	-0.533622000	3.076129000
B	-0.873254000	2.609829000	-1.155361000
B	-1.494305000	2.257296000	0.410682000
B	-0.099139000	2.554382000	1.358447000
B	-0.679173000	3.742093000	0.206203000
B	-1.348443000	-0.138395000	1.549267000
B	0.091997000	-1.253958000	1.153698000
B	1.436655000	-0.116142000	1.555839000
N	-2.748502000	-0.646411000	1.069241000
N	2.885296000	-0.501704000	1.097970000
Os	0.005899000	0.653618000	0.057323000
Se	-1.613264000	-0.684123000	-1.506573000
Se	1.917709000	-0.273515000	-1.537722000
H	-0.095893000	3.680678000	-1.010240000
H	-1.469012000	2.772268000	-2.167340000
H	-2.625001000	2.189760000	0.756351000
H	0.062167000	2.677633000	2.524983000
H	-1.139902000	4.819917000	0.370931000
H	-1.438605000	0.352226000	2.630536000

H	0.157957000	-2.261544000	0.536275000
H	1.485106000	0.458618000	2.597880000
H	0.148321000	1.853083000	-1.374021000
H	0.994055000	1.826991000	0.838152000
H	0.493511000	3.650821000	0.815492000
H	-0.825645000	-1.365704000	2.023970000
H	0.987149000	-1.309565000	2.072571000

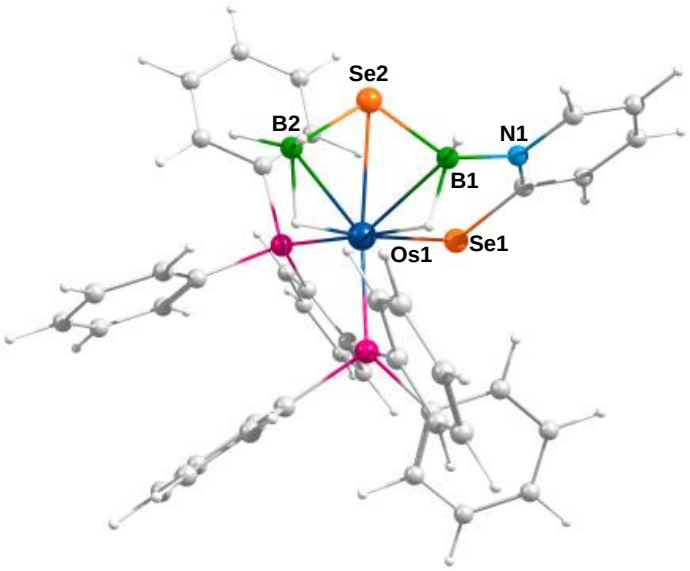


Figure S82. Optimized geometry of **10**.
Total energy = -2483.14378986 a.u.
Cartesian coordinates for the calculated structure **10** (in Å)

C	-2.322049000	-0.240695000	1.932503000
Os	0.275753000	-0.438662000	-0.594550000
Se	0.900422000	-1.762179000	1.544068000
Se	0.132883000	-2.624045000	-2.033804000
P	1.129126000	1.689596000	-0.072071000
P	-1.992363000	-0.287655000	0.096717000
N	2.718838000	-2.750690000	-0.439482000
C	2.604387000	1.703543000	-2.504242000
C	-3.085135000	1.080323000	-0.543202000
C	-2.982045000	-1.790202000	-0.386882000
C	-3.478221000	-0.805090000	2.491598000
C	-1.410746000	0.388087000	2.787820000
C	-2.808004000	-0.085093000	4.699849000
C	-1.038995000	3.399218000	0.595813000
C	-3.718612000	-0.726774000	3.861734000
C	2.306536000	-2.834706000	0.862298000
C	3.636486000	2.093068000	-3.357634000
C	-4.041920000	-1.731858000	-1.295552000
C	-1.651435000	0.469885000	4.158767000
C	-1.833289000	4.537588000	0.492814000
C	3.601700000	1.203567000	3.190355000
C	0.318947000	4.174919000	-1.229765000
C	-0.486551000	5.308003000	-1.343955000
C	2.933912000	1.056183000	1.978738000
C	2.967465000	-3.736671000	1.726681000
C	-1.559430000	5.498507000	-0.478402000
C	4.646724000	2.935798000	-2.900706000
C	4.615503000	3.392616000	-1.584183000
C	3.581136000	3.009805000	-0.732689000
C	1.617664000	3.012143000	2.459680000
C	3.272577000	2.250636000	4.050767000
C	-2.633474000	-3.030763000	0.165863000
C	3.760430000	-3.519095000	-0.875573000
C	-4.199753000	1.540863000	0.169911000
C	2.279433000	3.153082000	3.681289000
C	4.422540000	-4.400754000	-0.063591000
C	-4.777858000	3.043041000	-1.632691000
C	4.009126000	-4.512602000	1.278001000
C	-5.037182000	2.515457000	-0.369437000
C	-4.747660000	-2.887681000	-1.637105000
C	-4.407804000	-4.110662000	-1.068143000
C	-3.347215000	-4.177754000	-0.163442000
C	2.563148000	2.155571000	-1.179666000
C	1.930851000	1.958156000	1.593658000
C	0.060398000	3.206030000	-0.252044000
C	-3.670317000	2.594855000	-2.348323000
C	-2.828583000	1.625705000	-1.806297000
B	2.001775000	-1.874376000	-1.512465000
B	-0.284318000	-0.811338000	-2.980341000
H	2.021743000	3.978867000	4.335567000
H	3.789840000	2.363453000	4.997128000
H	4.377293000	0.496985000	3.464333000
H	3.203364000	0.239611000	1.322057000
H	0.868276000	3.741793000	2.184084000
H	3.572738000	3.381387000	0.283939000
H	5.396092000	4.049624000	-1.216329000
H	5.452017000	3.233822000	-3.563117000
H	3.647973000	1.730152000	-4.379411000

H	1.833352000	1.043299000	-2.876294000
H	1.154142000	4.056915000	-1.906630000
H	-0.266266000	6.043102000	-2.110363000
H	-2.181141000	6.382843000	-0.562911000
H	-2.674869000	4.663984000	1.164113000
H	-1.283998000	2.663163000	1.350198000
H	-1.972065000	1.288549000	-2.372335000
H	-3.453848000	3.001267000	-3.329904000
H	-5.431166000	3.799542000	-2.053363000
H	-5.893639000	2.859948000	0.200163000
H	-4.421079000	1.147524000	1.153288000
H	-1.797510000	-3.101183000	0.852341000
H	-3.066868000	-5.128178000	0.277194000
H	-4.958831000	-5.007128000	-1.330321000
H	-5.564080000	-2.824312000	-2.348105000
H	-4.326643000	-0.788816000	-1.743284000
H	2.629237000	-3.793744000	2.753065000
H	4.507819000	-5.200157000	1.951533000
H	5.240283000	-4.987120000	-0.459976000
H	4.019773000	-3.382426000	-1.915861000
H	-1.430098000	-0.756373000	-3.304824000
H	0.541590000	-0.540512000	-3.801730000
H	-0.114215000	0.276861000	-2.107356000
H	1.952060000	-0.624018000	-1.078772000
H	2.710798000	-1.713092000	-2.458023000
H	-0.491055000	0.796124000	2.390178000
H	-0.924863000	0.956068000	4.799988000
H	-2.994104000	-0.028831000	5.766664000
H	-4.616851000	-1.173600000	4.273741000
H	-4.191510000	-1.318684000	1.859184000

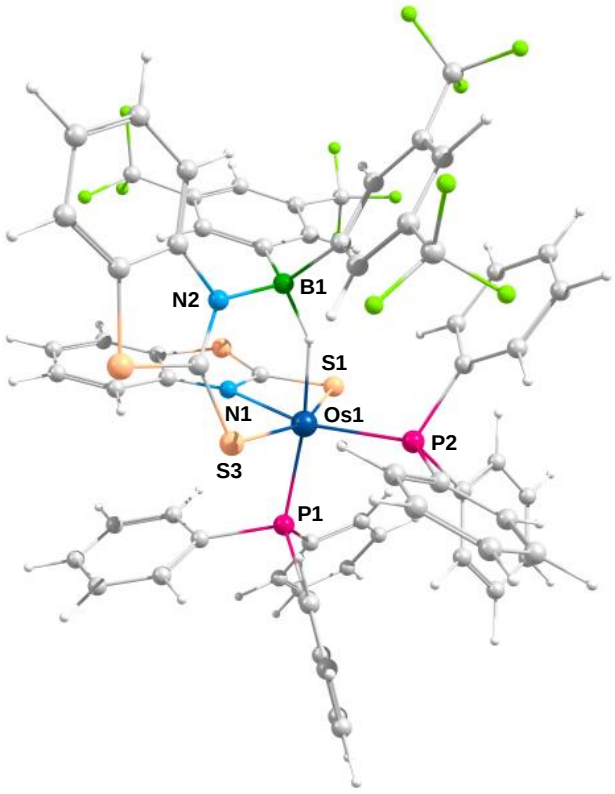


Figure S83. Optimized geometry of **11**.
Total energy = -6242.52235302 a.u.
Cartesian coordinates for the calculated structure **11** (in Å)

Os	1.181770000	0.122059000	0.018383000
S	0.891610000	-0.062501000	-2.366997000
S	1.067548000	1.136410000	2.309322000
S	-1.079806000	0.483902000	-4.477220000
S	0.379123000	4.167279000	1.815946000
N	-1.825963000	0.370095000	-1.987857000
N	0.727629000	2.295364000	0.037032000
C	0.723222000	2.512296000	1.341842000
C	0.343246000	3.562470000	-2.085885000
C	-3.494208000	3.414983000	2.068789000
C	2.554497000	-2.544594000	3.303669000
C	-0.583865000	-4.072047000	1.546458000
C	0.200822000	4.577682000	0.110135000
C	2.771400000	-2.532307000	1.919820000
C	-3.008249000	0.562792000	-2.744318000
C	-3.407741000	3.556824000	0.687035000
C	1.610426000	-3.580352000	-0.492189000
C	-1.594558000	-4.543576000	2.384099000
C	0.023737000	4.797593000	-2.635939000
C	3.601353000	3.383201000	-0.947394000
C	-0.733322000	0.265678000	-2.762830000
C	4.333640000	0.597167000	2.436413000

C	4.545498000	1.149925000	1.170288000
C	-2.425758000	1.331045000	0.451800000
C	-0.071345000	-2.777818000	1.699037000
C	-2.749950000	-1.306225000	-0.254484000
C	-3.020308000	2.240571000	2.644863000
C	-3.802158000	-1.467499000	0.654071000
C	-4.318497000	0.645077000	-2.260888000
C	3.796349000	4.472055000	-1.791718000
C	3.722208000	2.071398000	-1.426745000
C	-0.620855000	-1.964394000	2.700449000
C	3.599802000	-2.803983000	4.190600000
C	4.577698000	-0.644231000	-1.101116000
C	5.613747000	2.047413000	1.008351000
C	-2.530119000	-2.368816000	-1.146963000
C	4.040143000	-1.523267000	-2.047851000
C	4.067883000	-2.791934000	1.452849000
C	-1.622080000	-2.439602000	3.544683000
C	-0.112749000	5.816718000	-0.441797000
C	-2.869851000	2.540436000	-0.102950000
C	1.784934000	-5.733875000	-2.298928000
C	-0.204619000	5.917127000	-1.826529000
C	-2.109898000	-3.735560000	3.393088000
C	1.074415000	-3.484950000	-1.780338000
C	-2.788155000	0.661388000	-4.126603000
C	6.429307000	2.390283000	2.084317000
C	5.149973000	0.939310000	3.513150000
C	-3.828272000	0.857098000	-5.030764000
C	4.881207000	-3.062054000	3.712398000
C	4.849176000	-2.438382000	-2.720395000
C	6.196882000	1.840196000	3.343468000
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C	-5.360706000	0.838039000	-3.160985000
C	2.224415000	-4.786014000	-0.118767000
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C	-4.596786000	-2.616390000	0.666518000
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C	-3.323686000	-3.512116000	-1.145411000
C	2.314199000	-5.848847000	-1.014640000
C	4.098027000	4.271276000	-3.137382000
C	5.109777000	-3.055531000	2.338045000
C	5.953836000	-0.710551000	-0.841240000
C	6.214374000	-2.494394000	-2.453516000
C	-4.371561000	-3.646880000	-0.236434000
C	4.201308000	2.973458000	-3.630002000
C	-2.498026000	1.221082000	1.850088000
C	6.762281000	-1.627252000	-1.509815000
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C	-3.045856000	-4.637653000	-2.103397000
C	-5.755524000	-2.701668000	1.622532000
C	-3.032899000	2.103278000	4.143111000
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P	1.392401000	-2.162453000	0.705520000
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C	0.426254000	3.439486000	-0.695462000
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F	-5.484691000	-2.116497000	2.810591000
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F	-1.959354000	2.704405000	4.717270000
F	-5.317432000	4.831006000	0.094957000
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H	-0.292252000	6.677745000	0.190591000
H	0.551909000	2.710130000	-2.715782000

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