ELECTRONIC SUPPLEMENTARY INFORMATION

Easy Abstraction of a Hydride Anion from an Alkyl C–H Bond of a Coordinated Bis(N-Heterocyclic Carbene)

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Experimental Details

General Procedures. Solvents were dried over sodium diphenyl ketyl (hydrocarbons, THF, hexane) or CaH_2 (dichloromethane) and distilled under nitrogen before use. The reactions were carried out under nitrogen at room temperature, using Schlenk-vacuum line techniques, and were routinely monitored by solution IR spectroscopy (carbonyl stretching region). The salt $[MeHIm(CH_2)_3ImHMe]Br_2$ was prepared as described previously.[1] All remaining reagents were purchased from commercial sources. All reaction products were vacuum-dried for several hours prior to being weighted and analyzed. IR spectra were recorded in solution on a Perkin-Elmer Paragon 1000 FT spectrophotometer. NMR spectra were run on a Bruker DPX-300 instrument. Microanalyses were obtained from the University of Oviedo Analytical Service. FAB mass spectra were obtained from the University of A Coruña Mass Spectrometric Service; data given refer to the most abundant molecular ion isotopomer.

Synthesis of $[Ru(\kappa^2 C_{NHC},C_{NHC}\text{-}MeIm(CH_2)_3ImMe)(CO)_3]$ (1). A toluene solution of $K[N(SiMe_3)_2]$ (3.0 mL, 0.5 M, 1.500 mmol) was added to a suspension of $[MeHIm(CH_2)_3ImHMe]Br_2$ (258 mg, 0.706 mmol) in THF (30 mL). After stirring for 20 min, finely powdered $[Ru_3(CO)_{12}]$ (150 mg, 0.235 mmol) was added. The color changed from orange to yellow. The mixture was stirred for 1 h. The solid was filtered off and the filtered solution was evaporated to dryness under reduced pressure. The residue was washed with hexane (5 mL) to give compound

1 as a yellow solid (233 mg, 85%). Anal. Calcd for $C_{14}H_{16}N_4O_3Ru$ (389.37): C, 43.19; H, 4.14; N, 14.39. Found: C, 43.35; H, 4.23; N, 14.03. (+)-FAB MS: *m/z* 390 $[M]^+$. IR (THF, cm^{-1}): ν_{CO} 1986 (s), 1974 (s), 1852 (vs). ^1H NMR (C_6D_6, 298 K, 300.09 MHz): δ 6.23 (d, *J* = 1.9 Hz, 2 H), 6.01 (d, *J* = 1.9 Hz, 2 H), 3.75 (s, 6 H), 3.50–2.88 (br, 4 H), 1.00 (quint, *J* = 6.0 Hz, 2 H). $^{13}C\{^1H\}$ and DEPT-135 NMR (C_6D_6, 75.48 MHz, 300 K): δ 218.3 (s, 3 *C*O), 188.5 (s, 2 $C_{carbene}$), 122.5 (s, 2 *C*H), 119.9 (s, 2 *C*H), 46.0 (s, CH_2), 40.3 (s, 2 CH_3), 32.8 (s, 2 CH_2) (Figure SI-1).

Synthesis of [Ru(κ^3*C*$_{NHC}$,*C*,*C*$_{NHC}$-MeImCH$_2$CHCH$_2$ImMe)(CO)$_3$][Ru$_3$(*μ*-H)(*μ*-CO)(CO)$_{10}$] (2[Ru$_3$(*μ*-H)(*μ*-CO)(CO)$_{10}$]). ***Method 1:*** A toluene solution of K[N(SiMe$_3$)$_2$] (1.5 mL, 0.5 M, 0.750 mmol) was added to a suspension of [MeHIm(CH$_2$)$_3$ImHMe)]Br$_2$ (129 mg, 0.353 mmol) in THF (30 mL). After stirring for 20 min, finely powdered [Ru$_3$(CO)$_{12}$] (300 mg, 0.470 mmol) was added. The color changed from orange to red. The mixture was stirred for 2 h. The solid was filtered off and the filtered solution was evaporated to dryness under reduced pressure. The residue was washed with hexane (5 mL) to give **2**[Ru$_3$(*μ*-H)(*μ*-CO)(CO)$_{10}$] as a red solid (402 mg, 85%). ***Method 2:*** Finely powdered [Ru$_3$(CO)$_{12}$] (450 mg, 0.704 mmol) was added to a THF solution (30 mL) of compound **1** (275 mg, 0.706 mmol) and the mixture was stirred for 45 min. The color changed from yellow to red. The solvent was removed under reduced pressure and the solid residue was recrystallized from dichloromethane/hexane to give **2**[Ru$_3$(*μ*-H)(*μ*-CO)(CO)$_{10}$] as a red solid (635 mg, 90%). Anal. Calcd for $C_{25}H_{16}N_4O_{14}Ru_4$ (1000.70): C, 30.01; H, 1.61; N, 5.60. Found: C, 30.12; H, 1.67; N, 5.49. (+)-FAB MS: *m/z* 389 $[\mathbf{2}]^+$. IR (THF, cm^{-1}): ν_{CO} 2097 (m), 2072 (w), 2035 (m), 2014 (vs), 1986 (vs), 1950 (s), 1730 (w). ^1H NMR (CD_2Cl_2, 296 K, 300.09 MHz): δ 7.18 (d, *J* = 1.9 Hz, 2 H), 6.99 (d, *J* = 1.9 Hz, 2 H), 4.43 (dd, *J* = 13.4, 7.7 Hz, 2 H), 4.18 (dd, *J* = 13.4, 1.7 Hz, 2 H), 3.84 (s, 6 H), 3.59 (tt, *J* = 7.7, 1.7 Hz, 1 H), –12.65 (s, 1 H). $^{13}C\{^1H\}$ and DEPT-135 NMR (CD_2Cl_2, 75.47 MHz, 300 K): δ 202.5 (s, 1 *C*O), 192.1 (s, 2 *C*O), 172.8 (s, 2 $C_{carbene}$), 125.01 (s, 2 *C*H), 120.2 (s, 2 *C*H), 62.4 (s, 2 CH_2), 39.2 (s, 2 CH_3), 31.8 (s, *C*H) (Figure SI-1).

Reaction of Compound 1 with [CPh$_3$][PF$_6$]. A mixture of [CPh$_3$][PF$_6$] (39 mg, 0.100 mmol) and compound **1** (39 mg, 0.100 mmol) in dichloromethane (15 mL) was stirred for 2 h. The solvent was evaporated to dryness and the solid residue was analyzed by IR and NMR. The corresponding spectra indicated the presence of $\mathbf{2}^+$ among other reaction products. All attempts to isolate **2**[PF$_6$] in pure form were unsuccessful.

X-Ray Diffraction Analyses. Crystals of **1**·(C$_6$H$_6$)$_{0.5}$ and **2**[Ru$_3$(*μ*-H)(*μ*-CO)(CO)$_{10}$] were analyzed by X-ray diffraction methods. A selection of crystal measurement and refinement data is

given in Table SI-1. Diffraction data were collected on an Oxford Diffraction Xcalibur Onyx Nova single crystal diffractometer, using Cu-Kα radiation. Empirical absorption corrections were applied using the SCALE3 ABSPACK algorithm as implemented in the program CrysAlisPro RED,[2] for **1**·$(C_6H_6)_{0.5}$ and XABS2[3] for **2**[$Ru_3(\mu\text{-H})(\mu\text{-CO})(CO)_{10}$]. The structures were solved using the program SIR-97.[4] Isotropic and full matrix anisotropic least square refinements were carried out using SHELXL.[5] Two independent molecules of **1** were found in the asymmetric unit of **1**·$(C_6H_6)_{0.5}$ (Figure SI-2). The cation $\mathbf{2}^+$ of **2**[$Ru_3(\mu\text{-H})(\mu\text{-CO})(CO)_{10}$] was found disordered over two positions (Figure SI-3) with a 52:48 occupancy ratio (restraints on the geometrical and thermal parameters were required to model the disorder). The anion $[Ru_3(\mu\text{-H})(\mu\text{-CO})(CO)_{10}]^-$ was not disordered (Figure SI-4). All non-H atoms were refined anisotropically. The position of the hydride ligand of **2**[$Ru_3(\mu\text{-H})(\mu\text{-CO})(CO)_{10}$] was calculated with XHYDEX.[6] The remaining hydrogen atoms were set in calculated positions and refined riding on their parent atoms. The molecular plots were made with the X-SEED program package.[7] The WINGX program system[8] was used throughout the structure determinations.

Computational Details. Density functional theory (DFT) calculations were carried out using the Becke's three-parameter hybrid exchange-correlation functional[9] and the B3LYP non-local gradient correction.[10] The LanL2DZ basis set, with relativistic effective core potentials, was used for the Ru atoms.[11] The basis set used for the remaining atoms was the 6-31G, with addition of (d,p)-polarization.[12] No simplified model compounds were used for the calculations. All stationary points of the mechanistic study were confirmed as energy minima (reactants, products, and intermediates; all positive eigenvalues) or transition states (one imaginary eigenvalue) by analytical calculation of frequencies. IRC calculations were used to verify that the transition states found were correct saddle points connecting the proposed minima. All energies given in this article are electronic energies calculated in gas phase. All calculations were carried out without symmetry constraints utilizing the Gaussian03 package.[13] Atomic coordinates of all optimized structures are given in Tables SI-2 to SI-8.

References

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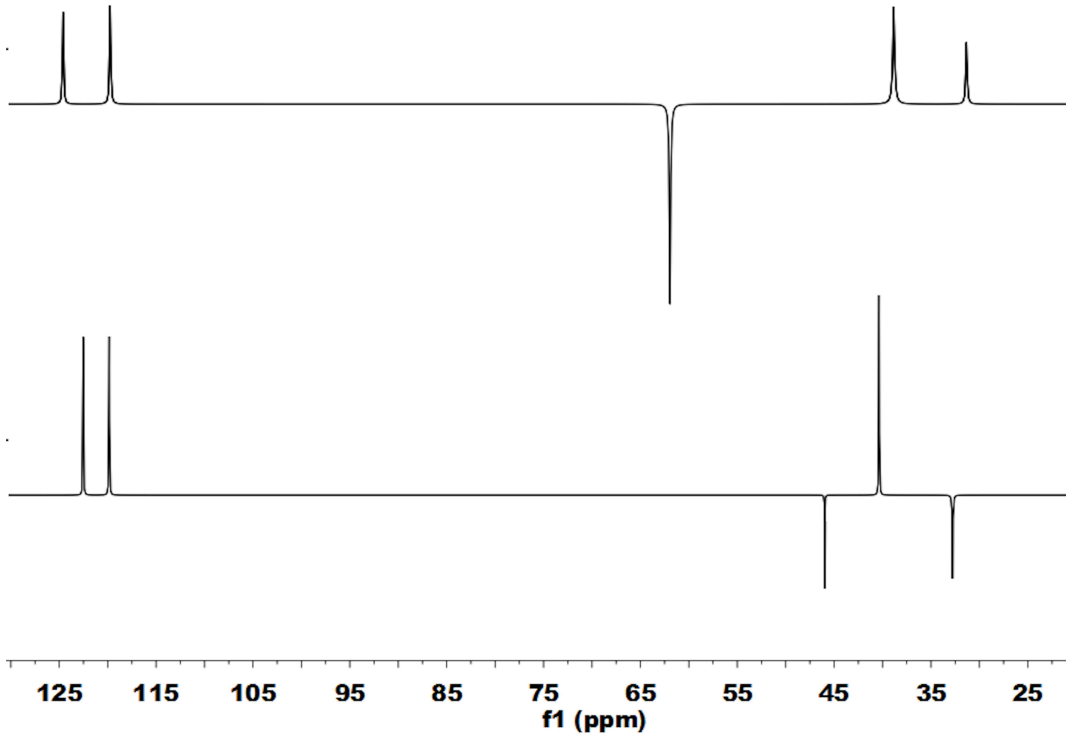


Figure SI-1. DEPT-135 ^{13}C NMR spectra (75.48 MHz, 300 K) of **1** in C_6D_6 (bottom) and **2**[Ru(μ-H)(μ-CO)$(CO)_{11}$] in CD_2Cl_2 (top).

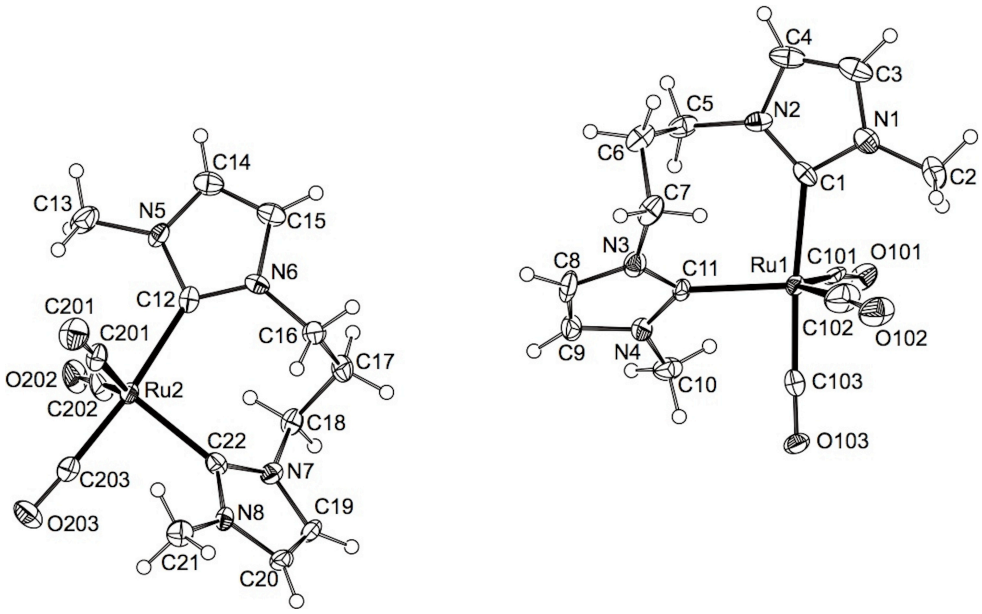


Figure SI-2. The two independent molecules of **1** found in the asymmetric unit of **1**·$(C_6H_6)_{0.5}$. Thermal ellipsoids are shown at 40% probablity.

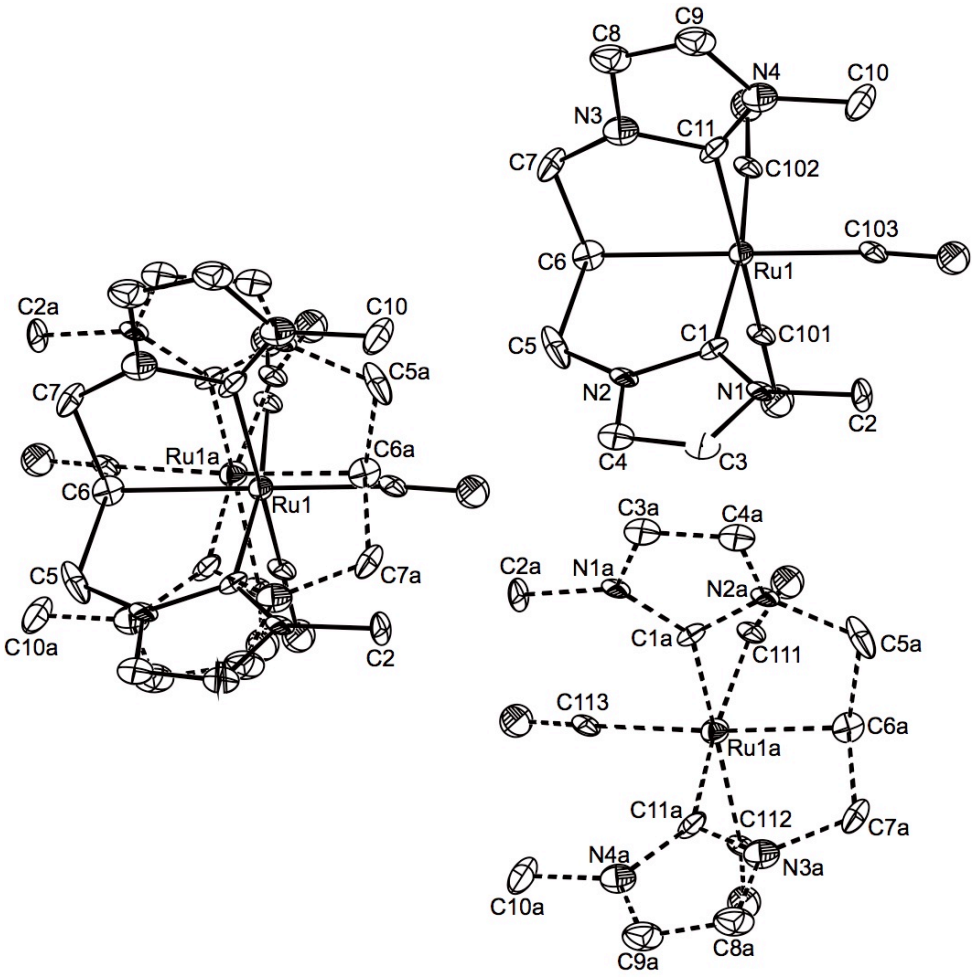


Figure SI-3. Structure of the cation of **2**[$Ru_3(\mu\text{-}H)(\mu\text{-}CO)(CO)_{10}$], showing its disorder (left) and separate representations of the two parts involved in its disorder (right). Hydrogen atoms have been omitted for clarity. Thermal ellipsoids are shown at 20% probability.

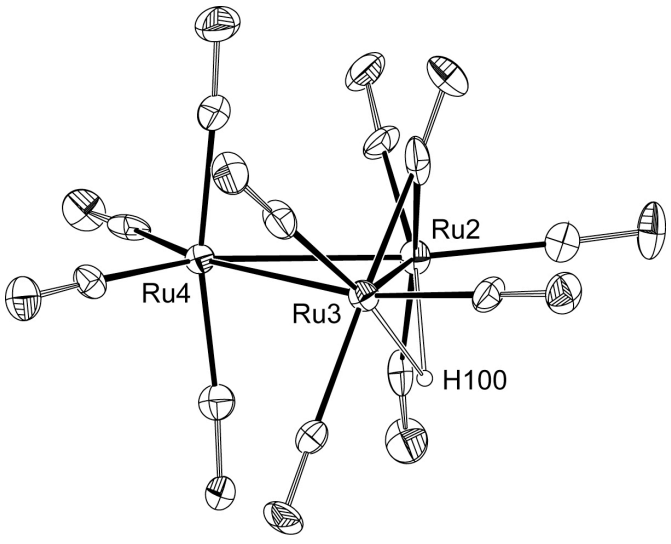


Figure SI-4. Structure of the anion of **2**[$Ru_3(\mu\text{-}H)(\mu\text{-}CO)(CO)_{10}$]. Thermal ellipsoids are shown at 20% probability.

Table SI-1. Crystal, Measurement, and Refinement Data for Compounds 1 and 2[$Ru_3(\mu$-H)(μ-CO)$(CO)_{10}$].

	1·$(C_6H_6)_{0.5}$	**2**[$Ru_3(\mu$-H)(μ-CO)$(CO)_{10}$]
formula	$C_{14}H_{16}N_4O_3Ru$·0.5 C_6H_6	$[C_{14}H_{15}N_4O_3Ru_1][C_{11}H_1O_{11}Ru_3]$
fw	428.43	1000.70
cryst syst	monoclinic	monoclinic
space group	*P*21/n	*P*21/n
a, Å	20.5311(8)	13.3007(5)
b, Å	8.6613(3)	15.6862(5)
c, Å	20.6671(7)	15.2570(5)
α, β, γ, deg	90, 104.761(4), 90	90, 92.473(3), 90
V, $Å^3$	3553.9(2)	3180.2(2)
Z	8	4
F(000)	1736	1928
D_{calcd}, g cm^{-3}	1.601	2.090
μ(CuKα), mm^{-1}	7.349	15.748
cryst size, mm	0.19 x 0.11 x 0.05	0.06 x 0.05 x 0.03
T, K	123.0(1)	123(2)
θ range, deg	3.52 to 67.50	4.04 to 62.51
min./max. *h*, *k*, *l*	–24/23, –10/7, –22/24	–14/15, 0/17, 0/17
no. collected reflns	12764	4889
no. unique reflns	6367	4889
no. reflns with $I > 2\sigma(I)$	5576	3341
no. params/restraints	443/0	457/25
GOF on F^2	1.193	1.199
R_1 (on *F*, $I > 2\sigma(I)$)	0.075	0.078
wR_2 (on F^2, all data)	0.198	0.235
min./max. $\Delta\rho$, e $Å^{-3}$	–2.177/2.222	–1.297/1.388

Table SI-2. Atomic coordinates for the DFT-optimized structure of 1

38			
C	-3.758981	0.670151	-0.330091
C	-2.921305	1.690124	-0.627211
C	-3.539852	-1.730502	0.304652
C	-0.811311	-1.714596	1.985391
H	-0.684678	3.234644	0.962829
C	-0.458270	2.007152	-0.810077
C	-1.630214	-0.140427	-0.170064
C	-0.638732	-2.487991	-1.363768
C	0.113561	2.699471	0.434306
C	1.203512	-2.970699	0.649832
C	0.805457	1.736085	1.411326
C	1.674226	-0.181694	0.081403
C	3.206912	-1.535298	-1.375212
C	3.162628	1.490904	0.576358
C	3.804664	0.601594	-0.218799
H	-4.835857	0.627179	-0.298300
H	-3.119863	2.711461	-0.910745
H	-4.596922	-1.720793	0.030078
H	-3.440998	-1.907843	1.377186
H	-0.761252	2.760329	-1.543391
H	-3.025384	-2.525468	-0.230713
H	0.290672	1.367880	-1.278809
H	0.836100	3.453603	0.097107
H	1.230018	2.291567	2.253492
H	3.932076	-1.221249	-2.130263
H	0.105282	0.995412	1.803460
H	2.288735	-1.870158	-1.858671
H	3.624168	-2.362833	-0.795041
H	3.503794	2.408069	1.030748
H	4.819225	0.586791	-0.585097
N	-2.965767	-0.434174	-0.060379
N	-1.631641	1.182760	-0.532257
N	1.875429	0.998999	0.754143
N	2.889198	-0.399009	-0.519212
O	-1.362253	-1.839644	3.009455
O	-1.090414	-3.076253	-2.266490
O	1.947116	-3.821643	0.913694
Ru	-0.039069	-1.573904	0.229142

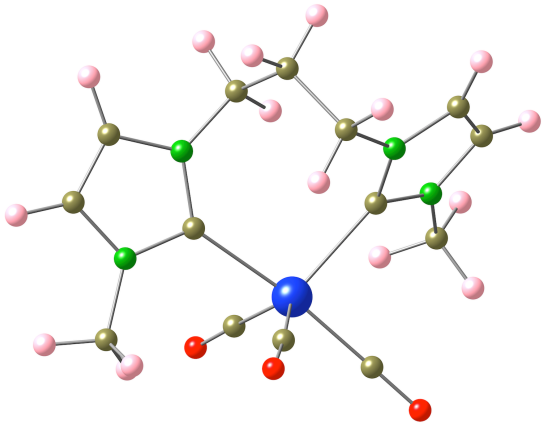
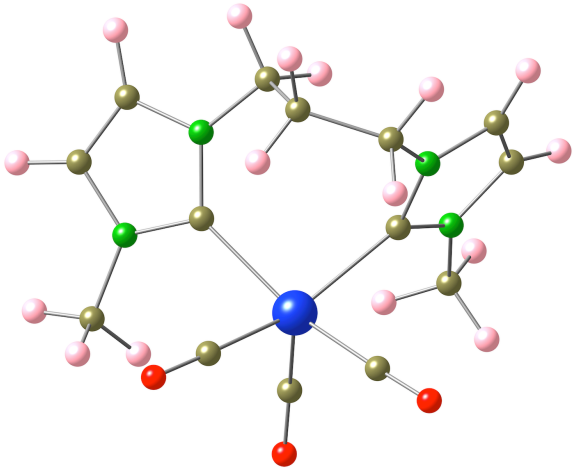
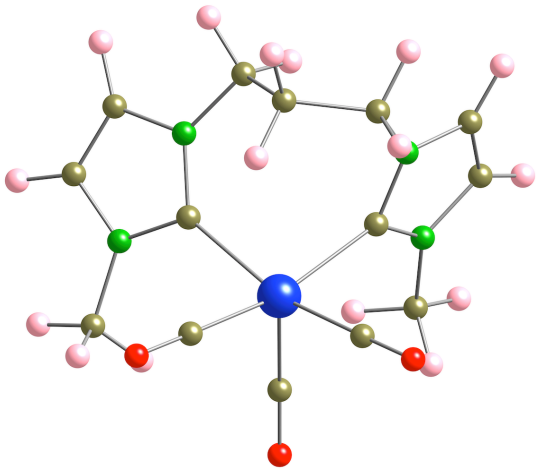


Table SI-3. Atomic coordinates for the DFT-optimized structure of ts1



38			
C	3.244416	1.566811	0.794130
C	2.271015	2.415438	0.391706
C	3.554591	-0.906935	0.882388
C	1.745536	-1.833323	-1.715036
H	0.334228	0.971322	-2.295794
C	-0.008172	2.238267	-0.595022
C	1.493924	0.289806	0.062449
C	0.726878	-2.656317	1.018615
C	-0.443183	1.657109	-1.955621
C	-0.820600	-2.611929	-1.521816
C	-1.791648	0.894539	-1.961857
C	-1.525947	-0.486031	0.114260
C	-2.030730	-1.502425	2.357982
C	-3.394802	0.819338	-0.029462
C	-3.446644	0.168384	1.158179
H	4.225879	1.757425	1.197989
H	2.233134	3.493101	0.382542
H	4.447671	-0.597281	1.428846
H	3.852153	-1.411164	-0.039212
H	0.213743	3.303095	-0.697024
H	2.976729	-1.601683	1.490961
H	-0.802475	2.152204	0.150991
H	-0.514952	2.468419	-2.688438
H	-2.598046	1.539817	-2.321898
H	-2.483394	-1.080491	3.258249
H	-1.710752	0.036617	-2.633535
H	-0.953933	-1.590135	2.486656
H	-2.444644	-2.498311	2.175879
H	-4.075935	1.523817	-0.479782
H	-4.184178	0.190901	1.944711
N	2.761527	0.283275	0.589264
N	1.215532	1.629721	-0.055355
N	-2.225019	0.405469	-0.652660
N	-2.303961	-0.615625	1.232318
O	2.640727	-2.028822	-2.438183
O	1.042155	-3.623957	1.591426
O	-1.547712	-3.282628	-2.135825
Ru	0.326881	-1.477814	-0.493007

Table SI-4. Atomic coordinates for the DFT-optimized structure of 1'.



38			
C	2.760568	2.569680	0.977904
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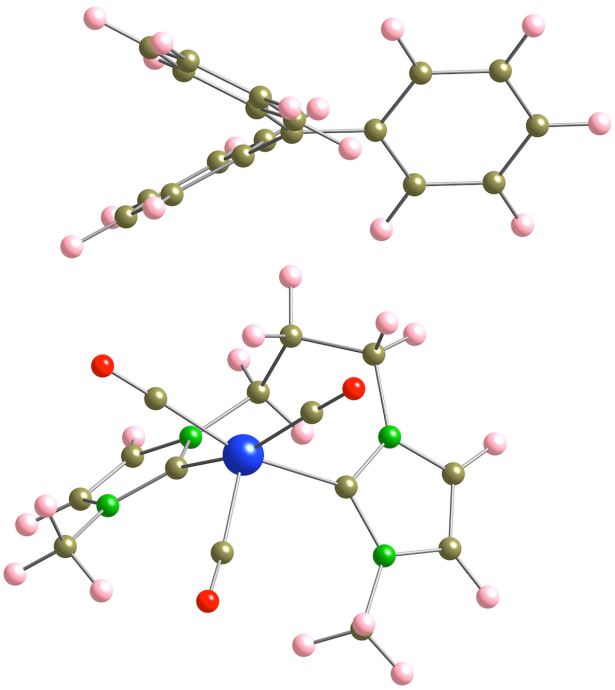
H	-0.103998	1.300729	-2.108326
C	-0.460939	2.801609	-0.609782
C	1.219755	1.059630	0.208026
C	0.585804	-2.195471	1.108757
C	-0.901647	1.996849	-1.844676
C	-0.669146	-1.628399	-1.785943
C	-2.207447	1.197302	-1.664787
C	-1.574017	-0.127962	0.399535
C	-1.725869	-1.063592	2.733399
C	-3.559743	1.000830	0.428285
C	-3.424849	0.391721	1.629675
H	3.683583	2.893963	1.431824
H	1.579860	4.341677	0.371636
H	3.959527	0.538259	2.066353
H	3.927596	-0.220213	0.453685
H	-0.303031	3.848052	-0.881943
H	2.700564	-0.662869	1.657376
H	-1.232151	2.790539	0.167758
H	-1.049600	2.675396	-2.692407
H	-3.072327	1.833013	-1.876236
H	-2.358277	-0.845614	3.596262
H	-2.224766	0.366386	-2.373005
H	-0.695435	-0.789502	2.956585
H	-1.764848	-2.132294	2.515978
H	-4.349273	1.620904	0.034958
H	-4.072773	0.377304	2.491412
N	2.434700	1.230288	0.826164
N	0.808246	2.352190	-0.015503
N	-2.428185	0.677149	-0.309793
N	-2.214186	-0.284484	1.599980
O	2.934964	-1.563426	-1.772555
O	0.805011	-3.156256	1.735550
O	-1.294179	-2.132138	-2.636987
Ru	0.305079	-0.898428	-0.311163

Table SI-5. Atomic coordinates for the DFT-optimized structure of 1' + $[CPh_3]^+$.



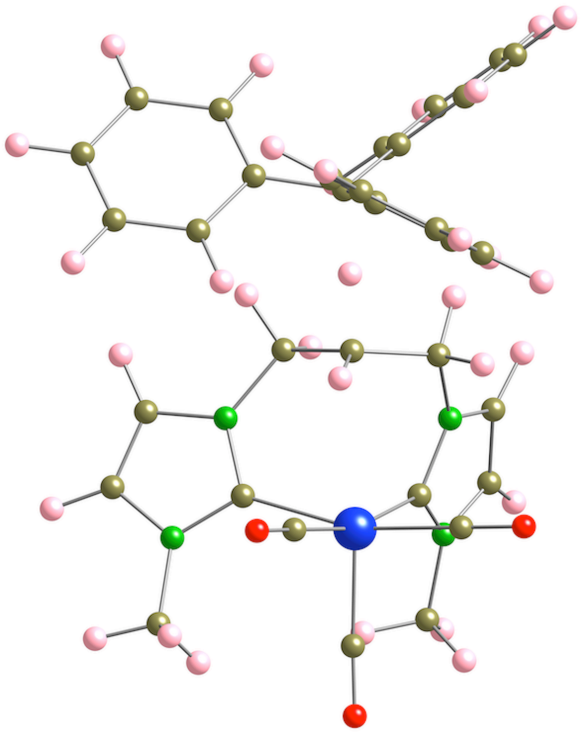
72			
C	3.377868	3.366927	-1.918001
C	2.070400	3.272770	-1.576946
C	5.494425	2.182828	-1.314749
C	3.459254	1.383216	2.375361
H	0.972285	-0.173216	0.712647
C	0.705928	1.832210	-0.042680
C	3.194332	1.664472	-0.424469
C	5.479148	-0.058430	0.849775
C	0.424981	0.323713	-0.097728
C	3.315078	-1.401635	2.243785
C	0.779281	-0.337684	-1.440730
C	2.954848	-1.374535	-0.577898
C	4.834295	-3.040393	-0.353515
C	1.788719	-2.482646	-2.187872
C	2.894367	-3.214492	-1.916677
H	3.884636	4.037536	-2.593317
H	1.214696	3.848675	-1.891153
H	5.880661	2.873243	-2.065295
H	5.980863	2.387660	-0.360242
H	0.698459	2.168572	0.998584
H	5.717323	1.161042	-1.622898
H	-0.076645	2.382287	-0.572293
H	-0.641767	0.185439	0.114102
H	1.088887	0.408458	-2.180154
H	5.124774	-3.937954	-0.900390
H	-0.098366	-0.847628	-1.843997
H	5.628230	-2.299103	-0.439464
H	4.692550	-3.296329	0.697979
H	0.982897	-2.652098	-2.884118
H	3.238965	-4.151665	-2.323618
N	4.048529	2.380532	-1.209823
N	1.974834	2.230000	-0.664615
N	1.832844	-1.367089	-1.361567
N	3.592774	-2.529701	-0.934652
O	3.417949	2.200112	3.190282
O	6.614428	-0.128503	1.066291
O	3.183725	-2.281922	2.979882
Ru	3.547728	0.044753	0.968108
C	-3.638670	-3.024119	-2.580742
C	-4.491608	-1.916734	-2.542682
C	-2.667742	-3.183034	-1.585328
C	-4.373409	-0.972370	-1.529437
C	-3.400099	-1.112030	-0.507074
C	-3.286168	-0.130028	0.566795
C	-2.909303	-0.555738	1.907375
C	-3.603938	1.270925	0.306393
C	-2.552310	-2.249827	-0.561492
C	-3.255340	-1.843379	2.395468
C	-2.184630	0.300891	2.778216
C	-2.890210	-2.247798	3.673329
C	-1.809834	-0.115521	4.048716
C	-2.158978	-1.392275	4.504054
C	-3.330941	1.869019	-0.951492
C	-4.211456	2.082337	1.298131
C	-3.635324	3.202733	-1.194652
C	-4.525446	3.412707	1.043741
C	-4.235666	3.982732	-0.199349
H	-3.734504	-3.760100	-3.373010
H	-5.261021	-1.797444	-3.299305
H	-2.002449	-4.041314	-1.606824
H	-5.062859	-0.136317	-1.494321

H	-1.789466	-2.379305	0.199013
H	-3.848946	-2.504365	1.773986
H	-1.892956	1.286107	2.430594
H	-3.184953	-3.229923	4.029879
H	-1.239750	0.551895	4.687693
H	-1.869553	-1.714023	5.499531
H	-2.857385	1.276212	-1.727256
H	-4.470688	1.644186	2.255396
H	-3.407439	3.639042	-2.162730
H	-5.009930	4.005413	1.813543
H	-4.481185	5.022054	-0.394070

Table SI-6. Atomic coordinates for the DFT-optimized structure of ts2.

72

C	-3.039888	-3.371456	-1.845873
C	-1.734462	-3.277751	-1.488002
C	-5.165082	-2.169109	-1.259738
C	-2.804888	-1.433758	2.443302
H	-0.535716	0.267100	1.060976
C	-0.429082	-1.760940	0.080990
C	-2.871917	-1.661583	-0.372078
C	-5.002024	-0.056556	1.185302
C	-0.288778	-0.243343	0.131651
C	-2.762710	1.405310	2.295563
C	-0.525333	0.483510	-1.194827
C	-2.795098	1.340772	-0.521018
C	-4.872154	2.755125	-0.446337
C	-1.677006	2.513947	-2.107143
C	-2.877176	3.116949	-1.925275
H	-3.538774	-4.043781	-2.525559
H	-0.873397	-3.850550	-1.793576
H	-5.537101	-2.728844	-2.118312
H	-5.671610	-2.521288	-0.358437
H	-0.392325	-2.177929	1.089801
H	-5.379600	-1.110825	-1.408046
H	0.404446	-2.189454	-0.485167
H	1.032630	-0.121230	0.264725
H	-0.679126	-0.251917	-1.992906
H	-5.188245	3.675062	-0.938399
H	0.362970	1.065083	-1.457563
H	-5.598808	1.970963	-0.664760
H	-4.835976	2.926007	0.630743
H	-0.852423	2.760551	-2.756529
H	-3.304666	3.992397	-2.387443
N	-3.721154	-2.374809	-1.157191
N	-1.652895	-2.224410	-0.588757
N	-1.642977	1.433926	-1.235545
N	-3.548526	2.390403	-0.950645
O	-2.650661	-2.239297	3.249166
O	-6.135738	-0.054833	1.394477
O	-2.596522	2.293869	3.006354
Ru	-3.069656	-0.078769	1.042850
C	2.851209	3.983861	-1.425260
C	3.635323	2.924564	-1.884354
C	1.959484	3.768460	-0.369624
C	3.525394	1.659086	-1.307544
C	2.628323	1.417969	-0.247695
C	2.482861	0.073172	0.396177
C	2.581245	0.038908	1.893827
C	3.037416	-1.106454	-0.345646
C	1.853169	2.507966	0.211711
C	3.185378	1.076905	2.629111
C	2.069351	-1.058572	2.617688
C	3.259956	1.025275	4.021140
C	2.136960	-1.107697	4.007184
C	2.729440	-0.060121	4.718568
C	2.681340	-1.340878	-1.692829
C	3.931637	-2.013936	0.254673
C	3.183374	-2.431213	-2.398311
C	4.437937	-3.104094	-0.453844
C	4.064724	-3.323490	-1.779871
H	2.941549	4.968218	-1.874335
H	4.347107	3.083545	-2.689040
H	1.352705	4.586882	0.007129
H	4.158686	0.855852	-1.666290

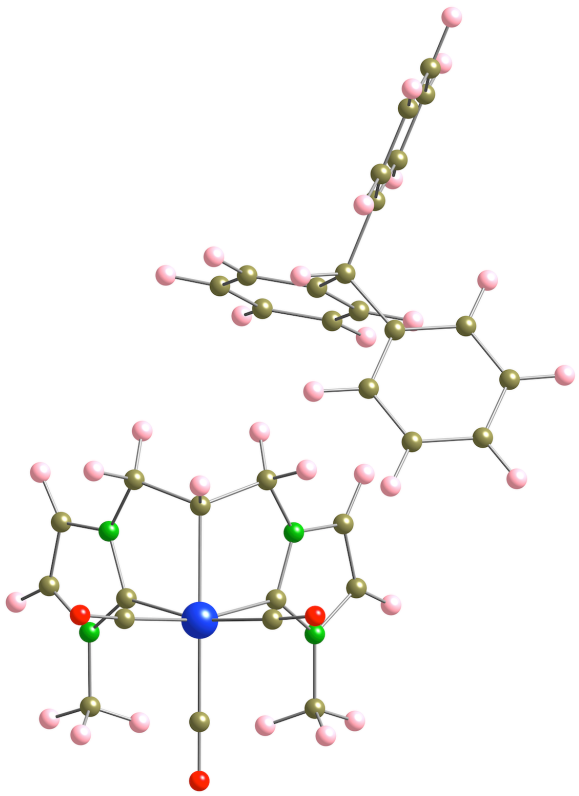


H	1.166414	2.360056	1.040226
H	3.617540	1.922611	2.106837
H	1.629282	-1.893169	2.080693
H	3.741568	1.835820	4.559932
H	1.731478	-1.965083	4.536430
H	2.784181	-0.096315	5.802165
H	2.010938	-0.649288	-2.195394
H	4.247504	-1.853245	1.278863
H	2.892846	-2.582555	-3.434123
H	5.134827	-3.779176	0.033909
H	4.460276	-4.172376	-2.328888

Table SI-7. Atomic coordinates for the DFT-optimized structure of 2^+ + $HCPh_3$.

72

C	1.755492	-0.303939	3.505609
C	0.695089	0.120446	2.769716
C	4.047855	-1.214114	3.013393
C	2.110252	-2.439729	-0.754568
H	0.583504	-0.131876	-1.826231
C	0.240661	0.207942	0.252466
C	2.326884	-0.489013	1.331510
C	4.795267	-1.515698	-0.190395
C	1.120882	0.336406	-0.999332
C	3.417075	-0.651524	-2.586002
C	1.408963	1.788185	-1.408770
C	3.639955	1.323559	-0.518661
C	6.020908	1.533132	0.260700
C	3.170668	3.524829	-0.731221
C	4.452979	3.416782	-0.296588
H	1.892980	-0.370995	4.573114
H	-0.274445	0.490634	3.061637
H	4.051581	-1.336411	4.096552
H	4.219999	-2.187514	2.551662
H	-0.431953	-0.651320	0.162253
H	4.854363	-0.531961	2.735837
H	-0.383878	1.090535	0.425115
H	-3.954036	0.184430	-1.698991
H	0.631849	2.481886	-1.066319
H	6.733101	2.356867	0.311417
H	1.470579	1.872412	-2.499676
H	5.942380	1.076797	1.250117
H	6.390555	0.796668	-0.453397
H	2.573403	4.393178	-0.959018
H	5.185102	4.173412	-0.063075
N	2.750514	-0.678395	2.608340
N	1.072297	0.000881	1.444797
N	2.694303	2.231660	-0.854750
N	4.728653	2.059093	-0.174160
O	1.516781	-3.417242	-0.809452
O	5.807409	-1.995095	0.062788
O	3.591801	-0.575285	-3.714775
Ru	3.050270	-0.709924	-0.648390
C	-8.578200	0.577841	-0.686561
C	-7.828735	1.234102	0.287876
C	-7.927427	-0.212737	-1.637800
C	-6.436602	1.104190	0.315587
C	-5.775459	0.311839	-0.629137
C	-4.253597	0.110869	-0.646055
C	-3.837694	-1.290080	-0.186451
C	-3.482108	1.212305	0.085637
C	-6.540941	-0.342755	-1.606955
C	-4.488870	-1.957616	0.859114
C	-2.759118	-1.930221	-0.814806
C	-4.058534	-3.218332	1.279199
C	-2.329781	-3.193380	-0.402043
C	-2.976700	-3.839865	0.653665
C	-3.012935	2.321328	-0.632964
C	-3.246994	1.173419	1.468502
C	-2.334359	3.363262	0.004885
C	-2.579271	2.218459	2.113039
C	-2.115623	3.317066	1.384023
H	-9.658712	0.682853	-0.709745
H	-8.323337	1.855725	1.028713
H	-8.500281	-0.725108	-2.405266
H	-5.869102	1.635564	1.072310



H	-6.042803	-0.963795	-2.347759
H	-5.352052	-1.498401	1.331673
H	-2.266149	-1.441012	-1.653120
H	-4.580617	-3.721033	2.088280
H	-1.504245	-3.679629	-0.914801
H	-2.652744	-4.826166	0.972201
H	-3.197216	2.377554	-1.703124
H	-3.593603	0.321810	2.045687
H	-1.998103	4.220259	-0.572691
H	-2.439159	2.183618	3.190691
H	-1.607039	4.134238	1.887364

Table SI-8. Atomic coordinates for the DFT-optimized structure of 2^{+}.

37			
C	3.010379	-2.245329	0.317945
C	2.764893	-2.123999	-1.012174
C	2.187831	-1.144146	2.419190
C	1.418197	2.213718	-0.377694
H	-0.002707	1.121461	-2.760412
C	1.298990	-0.522188	-2.373420
C	1.462881	-0.621997	0.066999
C	-0.032602	1.312627	1.958990
C	0.003193	0.254448	-2.097256
C	-1.457603	2.179470	-0.394145
C	-1.278274	-0.544291	-2.379705
C	-1.438931	-0.654316	0.059454
C	-2.170567	-1.163119	2.409369
C	-2.725798	-2.167538	-1.020504
C	-2.961182	-2.300102	0.310668
H	3.674688	-2.903179	0.855611
H	3.182495	-2.647096	-1.857702
H	3.079965	-1.610539	2.837852
H	2.198792	-0.085117	2.674102
H	2.070573	0.153554	-2.758667
H	1.305590	-1.619366	2.856043
H	1.165883	-1.321933	-3.112460
H	-1.127582	-1.341835	-3.117749
H	-2.751659	-1.962035	2.870068
H	-2.059484	0.117808	-2.769209
H	-1.154383	-1.198495	2.803152
H	-2.626458	-0.203727	2.663967
H	-3.135624	-2.696974	-1.865861
H	-3.612199	-2.971167	0.848229
N	2.208034	-1.313285	0.967660
N	1.811530	-1.128793	-1.139179
N	-1.786652	-1.158743	-1.147408
N	-2.163927	-1.364499	0.961306
O	2.258506	2.938590	-0.655919
O	-0.054310	1.566561	3.078092
O	-2.309665	2.885977	-0.683880
Ru	-0.007625	0.908123	0.025085