\(\eta^2\)-Iminoacyl and \(\eta^2\)-Acyl Monocyclopentadienyl Tantalum Complexes Bearing Oxo and Oxo-Borane Ligands

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Dedicated to J. Antonio Abad, an excellent scientist and friend

**Keywords:** Tantalum / Oxo ligands / Insertion reactions / Isocyanide / Carbon monoxide

Alkyl-chloro ligand exchange by the reaction of [TaCp*R2{O-[B(C6F5)3]}] (R = CH2Ph, Me) with Ph3CCl gave the monoalkyl compounds [TaCp*RCl{O-[B(C6F5)3]}] (R = CH2Ph, Me). Insertion of CNAr (Ar = 2,6-Me2C6H3) and CO into a Ta–C bond of the mono- and dialkyl complexes gave the iminoacyl compounds [TaCp*X{\eta^2-C(R)=NAr}O-[B(C6F5)3]}] (X = R = CH2Ph, Me; X = Cl, R = CH2Ph) and the acyl compounds [TaCp*X{\eta^2-C(R)=O}O-[B(C6F5)3]}] (X = R = CH2Ph, Me; X = Cl, R = CH2Ph, Me; X = Cl, R = CH2Ph). The related chloro compound [TaCp*Cl{\eta^2-C(Me)=NAr}O-[B(C6F5)3]}] was isolated from the reaction of the oxo derivative [TaCp*Cl{\eta^2-C(Me)=NAr}O] with the Lewis acid B(C6F5)3.

Addition of CNAr or pyridine to [TaCp*{(CH2Ph)\eta^2-C(Ph)=NAr}O-[B(C6F5)3]}] afforded the borane-free complex [TaCp*{(CH2Ph)\eta^2-C(Ph)=NAr}O] and the acid-base adduct L•B(C6F5)3 (L = py, CNAr). The molecular structures of [TaCp*Cl{\eta^2-C(Me)=NAr}O-[B(C6F5)3]}] and [TaCp*{(CH2Ph)\eta^2-C(Ph)=O}O-[B(C6F5)3]}] were obtained from X-ray diffraction studies.

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**Introduction**

The formation of C–C bonds through insertion of isocyanides (CNR) and carbon monoxide (CO) into M–C bonds is well-documented, and this reaction initially affords imido or acyl units affording diazo- or dioxobutene complexes,[6–7] the imino-\(\eta^2\)-iminoacyl derivatives [TaCp*(NR)Bu}{\eta^2-C(Me)=NAr}{\eta^2-C(Ph)=NAr}].[20] With regard to the CO insertion reactions, double migration of the alkyl group [process (a)] was observed for [TaCp*Cl2Me2][10] whereas the coupling of the acyl groups [process (b)] occurred for [TaCp*Me2(NR)][R = 2,6-Me2C6H3] to give the dinuclear compound [TaCp*(NR)Me2{\mu-\eta^2-OC(Me)=C(Me)O}] and ligand exchange [process (c)] for complexes [TaCp*ClMe(NR)][R = 2,6-Me2C6H3][10] leading to the oxo compounds [TaCp*Cl(O){\eta^2-C(Me)=NAr}].

Furthermore, the \(\eta^2\)-(methyl)acyl complexes remained elusive and mixed complexes with imido- \(\eta^2\)-iminoacyl complexes being stable.

It was observed that for monocyclopentadienyl imido tantalum derivatives addition of a second CNR molecule into the iminoacyl compounds [TaCp*(NR)X{\eta^2-C(Me)=NAr}][X = Cl, Me, OR, NH/BU] gave the results determined by the R group of the imido ligand. No reaction was found for R = 2,6-Me2C6H3[10] whereas a second insertion occurred for R = RBU.[20] Conversely, the imido complexes [TaCp*MeX(NR)](R = 2,6-Me2C6H3, RBU; X = Cl, Me) reacted with CO to give the dinuclear compound [TaCp*(NR)Me2{\mu-\eta^2-OC(Me)=C(Me)O}] for X = Me[10] and one of the few mononuclear derivatives [TaCp*Cl(O){\eta^2-C(Me)=NAr}] containing a terminal tantalum-oxo double bond for X = Cl.[10,11]

The versatility and potential applications of all these results determined by the R and X substituents of the imido...
complexes \([\text{TaCp}^*\text{MeX(NR)}]\) led us to extend our studies to similar insertion reactions of CNR and CO into the Ta–alkyl bonds of the related oxo-borane compounds \([\text{TaCp}^*\text{X}_2\{\text{O}-\text{B}(\text{C}_6\text{F}_5)_3\}]\) that have recently been isolated.\(^{[21]}\)

**Results and Discussion**

The monoalkyl oxo-borane compounds \([\text{TaCp}^*\text{RCl}\{\text{O-}\text{B}(\text{C}_6\text{F}_5)_3\}]\) \((R = \text{CH}_3\text{Ph}\ 1\text{c}, \ \text{Me}\ 1\text{d})\) were synthesised by an alkyl-chloro metathesis reaction of \([\text{TaCp}^*\text{R}_2\{\text{O-}\text{B}(\text{C}_6\text{F}_5)_3]\}\) \((R = \text{CH}_3\text{Ph}\ 1\text{a}, \ \text{Me}\ 1\text{b})\) with one equiv. of Ph$_3$CCl as chlorinating agent (Scheme 1). The reaction for \(1\text{c}\) proceeded smoothly at room temperature to give a pale yellow solid in good yield, whereas complex \(1\text{d}\) could not be isolated in the solid state. The formation of \(1\text{d}\) was demonstrated on a small scale by \(^1\text{H}\) NMR spectroscopy with a \(\text{C}_2\text{D}_6\) solution of \(1\text{b}\) that was treated with Ph$_3$CCl and heated to 40 °C. All attempts made to obtain the monomethyl complex through alkylation of \([\text{TaCp}^*\text{Cl}_2\{\text{O-}\text{B}(\text{C}_6\text{F}_5)_3]\}\) and redistribution reactions between \([\text{TaCp}^*\text{Cl}_2\{\text{O-}\text{B}(\text{C}_6\text{F}_5)_3]\}\) and \([\text{TaCp}^*\text{Me}_2\{\text{O-}\text{B-}(\text{C}_6\text{F}_5)_3]\}\) failed. The \(^{1}\text{B}\) and \(^{19}\text{F}\) NMR spectra of complexes \(1\text{c}\) and \(1\text{d}\) are consistent with the presence of a tetrahedral coordination environment around the tantalum atom. The \(^{1}\text{H}\) NMR spectra for the methyl-iminoacyl group, \(^{[21-31]}\) and the \(^{13}\text{C}\) NMR spectra with the monosubstitution of only one of the alkyl ligands. These pale yellow complexes are air and thermally stable in solution. The \(^{13}\text{C}\) NMR resonance at ca. \(\delta = 240\ \text{ppm}\) is the most apparent spectroscopic feature and confirms the presence of the \(\eta^2\)-iminoacyl ligand in complexes 2.

We previously reported\(^{[10]}\) on the isolation of the related oxo complex \([\text{TaCp}^*\text{Cl}\{\eta^2\text{-}\text{C(Me)=NAr}\}{\text{O}}]\) from the reaction of the monomethyl compound \([\text{TaCp}^*\text{ClMe(NAr)}]\) with CO through a process that involved insertion of CO into the Ta–Me bond and further rearrangement of the \(\eta^2\)-acyl intermediate with intramolecular oxo-imido exchange. Since the starting oxo-borane chloro-methyl complex could not be isolated, thus preventing access to the oxo-borane compound \([\text{TaCp}^*\text{Cl}\{\eta^2\text{-}\text{C(Me)=NAr}\}{\text{O}-\text{B}(\text{C}_6\text{F}_5)_3]\}]\) \(2\text{d}\) by insertion of CNAr into the Ta–C bond of the corresponding alkyl-chloro compound, we tried to obtain this compound by an alternative route. With this aim, we investigated the reaction of the oxo iminoacyl compound \([\text{TaCp}^*\text{Cl}\{\text{O-}\text{B}(\text{C}_6\text{F}_5)_3\}]\) with B(C$_6$F$_5$)$_3$, which afforded \(2\text{d}\) in high yield. The formation of \(2\text{d}\) was confirmed by \(^{13}\text{C}\) and \(^{19}\text{F}\) NMR spectroscopy. The \(^{13}\text{C}\) NMR spectrum shows the resonance corresponding to the Cp$_*$ atom of the \(\eta^2\)-iminoacyl ligand \((\delta = 240.1\ \text{ppm})\) to be slightly low-field shifted with respect to that of the starting compound \([\text{TaCp}^*\text{Cl}\{\eta^2\text{-}\text{C(Me)=NAr}\}{\text{O}}]\) \((\delta = 236.8\ \text{ppm})\). An analogous behaviour was observed in the \(^1\text{H}\) NMR spectrum for the methyl-iminoacyl group, which was shifted from \(\delta = 2.65\ \text{ppm}\) in \([\text{TaCp}^*\text{Cl}\{\eta^2\text{-}\text{C(Me)=NAr}\}{\text{O}}]\) to \(\delta = 2.94\ \text{ppm}\) in \(2\text{d}\).

The molecular structure of compound \([\text{TaCp}^*\text{Cl}\{\eta^2\text{-}\text{C(Me)=NAr}\}{\text{O}-\text{B}(\text{C}_6\text{F}_5)_3]\}]\) \((\text{Ar} = 2,6\text{-Me}_2\text{C}_6\text{H}_3)\) \(2\text{d}\) was obtained by X-ray diffraction studies. Figure 1 depicts an ORTEP drawing of \(2\text{d}\) with selected bond lengths and angles. Compound \(2\text{d}\) exhibits the typical geometry known for group 5 half-sandwich iminoacyl compounds with a tetrahedral coordination environment around the tantaluatom. Considering the centroid of the Cp$_*$ ring and the midpoint of the C(10)–N bond as coordination sites, the other two positions are occupied by the chloro and oxo ligands. The N atom of the \(\eta^2\)-iminoacyl group is located in a trans position with respect to the oxo ligand, as in analogous half-sandwich imido complexes and in \([\text{TaCp}^*\text{Cl}\{\eta^2\text{-}\text{C(Me)=NAr}\}{\text{O}}]\). Furthermore, the oxygen atom is attached to the boron atom of the B(C$_6$F$_5$)$_3$ group. All the values of the bond lengths and angles of compound \(2\text{d}\) are very close to the corresponding bond lengths and angles found for the parent compound \([\text{TaCp}^*\text{Cl}\{\eta^2\text{-}\text{C(Me)=NAr}\}{\text{O}}]\)\(^{[10]}\) except for the Ta–O bond, which is longer in \([\text{TaCp}^*\text{Cl}\{\eta^2\text{-}\text{C(Me)=NAr}\}{\text{O}}]\) \((1.731(7)\ \text{Å})\) than in \(2\text{d}\) \((1.809(5)\ \text{Å})\) as a consequence of the coordination of the oxygen atom to the B(C$_6$F$_5$)$_3$ ligand.\(^{[21,23-28,30,31]}\) The linear Ta–O–B angle \((174.4(6)°)\) and the B–O bond length \((1.52(1)\ \text{Å})\) are typical of oxo-borane compounds.\(^{[21-31]}\) The Ta–O bond in \(2\text{d}\) is longer than that in the oxo-borane compound \([\text{TaCp}^*\text{Cl}_2\{\text{O-}\text{B}(\text{C}_6\text{F}_5)_3]\}]\) \((1.784(2)\ \text{Å})\). This bond length is similar to the lower end of the range of Ta–O bond lengths for compounds with Ta–O–Ta bridges \((1.82–1.20\ \text{Å})\)\(^{[32-35]}\) and with terminal Ta–OH bonds \((1.85–1.97\ \text{Å})\)\(^{[36-38]}\). However, the single bond Ta–O distances are ca. 2.18 Å,
and thus a bond order of two should be considered for the Ta–O bond in 2d.

The reaction of the dialkyl [TaCp*R₂{O·B(C₆F₅)₃}] (R = CH₂Ph 1a, Me 1b) and the monobenzyl [TaCp*(CH₂Ph)-Cl·O·B(C₆F₅)₃]] (1c) complexes with CO in toluene gave the η²-acyl compounds [TaCp*X{η²-C(R)=O}·O·B(C₆F₅)₃] (X = R = CH₂Ph 3a, Me 3b; X = Cl, R = CH₂Ph 3c) after ca. 24 h at room temperature in moderate yields (Scheme 1). These pale yellow compounds were air and thermally stable below 120 °C for several hours. The ¹³C NMR spectra showed a resonance at ca. 305 ppm corresponding to the Cᵥσ² atom of the η²-acyl fragment. The slowness of the insertion reactions of CO is in contrast with the rapid transformations observed for complexes 1 with isocyanide and the behaviour observed for the analogous tert-buty1 imido complexes. Rather, all complexes 2–3 released the acid-base adduct L·B(C₆F₅)₃ (L = py, CNAr) in the presence of donor ligands such as isocyanide or pyridine. Only in the case of compound 2a were we able to isolate the borane-free 18-electron compound [TaCp*(CH₂Ph){η²-C(CH₂Ph)=NAr}·O] (4a), with a terminal oxo-tantalum bond. The remaining oxo-borane complexes decomposed under similar conditions. The ¹³C NMR spectrum of the new oxo iminoacyl compound 4a showed the η²-iminoacyl Cᵥσ² resonance at δ = 240.3 ppm. A comparison of this ¹³C resonance and that assigned to the CH₂-iminoacyl group in the ¹H NMR spectrum with the corresponding resonances in the parent compound 2a, showed a behaviour that is opposite to that observed for 2d and [TaCp*Cl{η²-C(Me)=NAr}·O].

**Conclusions**

The dialkyl oxo-borane compounds [TaCp*R₂{O·B(C₆F₅)₃}] can be transformed into the monoalkyl derivatives [TaCp*RX·O·B(C₆F₅)₃] by alkyl-chloro exchange with Ph₃CCl. All of these complexes reacted with one molecule of isocyanide or carbon monoxide to give the η²-iminoacyl or η²-acyl complexes, respectively. No further insertion processes have been observed. This behaviour is analogous to that observed for the imido compounds [TaCp*MeX(NR)] (R = 2,6-Me₂C₆H₃; X = Cl, Me), although in the oxo-borane compounds the insertion of CO gave stable η²-acyl derivatives because of the presence of a Ta–O multiple bond, which prevents further rearrangement.

Figure 1. ORTEP diagram of the X-ray structure of compound 2d. Thermal ellipsoids are drawn at the 50% level, and hydrogen atoms and C₆F₅ groups have been omitted for clarity. Selected bond lengths [Å] and angles [°]: Ta–O 1.809(5), Ta–N 2.127(7), Ta–C(10) 2.120(9), Ta–Cl 2.392(2), B–O 1.525(11), N–C(10) 1.26(1), N–Ta–C(10) 34.5(3), C(10)–N–Ta 72.4(5), N–C(10)–Ta 73.1(5), B–O–Ta 174.4(6).

Figure 2. ORTEP diagram of the X-ray structure of compound 3a. Thermal ellipsoids are drawn at the 50% level, and hydrogen atoms and C₆F₅ groups have been omitted for clarity. Selected bond lengths [Å] and angles [°]: Ta–O(1) 1.816(2), Ta–O(2) 2.110(3), Ta–C(1) 2.299(4), Ta–C(2) 2.011(3), B–O(1) 1.512(4), O(2)–C(2) 1.209(4), O(2)–Ta–C(2) 34.02(12), C(2)–O(2)–Ta 68.5(2), O(2)–C(2)–Ta 77.5(2), B–O(1)–Ta 173.02(19).
Experimental Section

All manipulations were performed under argon, and solvents were distilled from appropriate drying agents. NMR spectra were recorded at 300.13 (1H NMR), 183.31 (13F NMR), 75.47 (13C NMR) and 128.38 Hz (19B NMR) at room temperature with a Varian Unity 300 (1H, 13C, 19F) or Bruker Advance 400 (19B NMR) instrument. Chemical shifts (δ, ppm) are given in ppm, relative to internal TMS (1H and 13C NMR), and external CFCI3 (19F NMR) and BF3·OEt2 (13B NMR). Elemental analyses were performed with a Perkin–Elmer 2400 instrument. Compounds [TaCp*Me2]3, [TaCp*(CH2Ph)2{O–B(C6F5)3}]19, [TaCp*Me2–O{B(C6F5)3}]17, [TaCp*Cl{η2–C(Me)=NAr}{O}]40 and B(C6F5)322 were prepared by literature methods, and H2O–B(C6F5)223 was prepared from H2O and B(C6F5)3 in toluene at room temperature and used in situ without further purification.

[TaCp*(CH2Ph)2{O–B(C6F5)3}] (1e): A suspension of Ph3CCl (0.14 g, 0.50 mmol) and [TaCp*(CH2Ph)2{O–B(C6F5)3}] (7a) (0.50 g, 0.49 mmol) in toluene (5 mL) was stirred at room temperature with a colour change from yellow to brown. Later, all volatile components were removed under vacuum until the volume was ca. 1 mL, leaving a dark oil that was washed with hexane (2 × 10 mL) to yield 1e as a brownish solid (0.36 g, 76%). C7H15BFC11F13O7 (970.75): calc. C 43.31, H 2.28; found C 42.99, H 2.25. 1H NMR: δ = 2.14 (s, 15 H, C6Me5), 2.52 (d, JHH = 14.0 Hz, 1 H, C6H5), 2.80 (d, JHH = 14.0 Hz, 1 H, CH2Ph), 6.74 (m, 2 H, C6H5), 7.02 (m, 3 H, C6H3) ppm. 13B NMR: δ = 0.10 (O–B(C6F5)3) ppm. 13C NMR {1H}: δ = 11.5 (C6Me5), 82.3 (C6H5), 125.7 (C6H5), 127.1 (C6H5), 128.2 (C6H5), 128.3 (C6H5), 131.4 (C6H5), 135.0 (C6F3), 138.3 (C6F3), 145.9 (C6F3), 149.1 (C6F3) ppm. 19F NMR: δ = −132.9 (a–C6F3), −157.7 (p–C6F3), −163.3 (m–C6F3) ppm.

[TaCp*Cl{η2–C(Me)=NAr}{O–B(C6F5)3}] (3a): A solution of TaCl5 (0.50 g, 0.49 mmol) and B(C6F5)3 (0.28 g, 0.51 mmol) in toluene (5 mL) was stirred at room temperature for 1 h. Later, the solution was filtered, layered with hexane (5 mL) and cooled to −10 °C, obtaining 2d as yellow crystals (0.40 g, 74%). C6H5BF2ClF15O2Ta (1146.56): calc. C 32.8, H 3.25; found C 32.4, H 3.17. 1H NMR: δ = 1.88 (s, 15 H, C6Me5), 2.54 (d, JHH = 12.1 Hz, 1 H, Ta–CH2Ph), 2.70 (d, JHH = 12.1 Hz, 1 H, Ta–CH2Ph), 4.59 (d, JHH = 19.4 Hz, 1 H, C–CH2Ph), 4.88 (d, JHH = 19.4 Hz, 1 H, C–CH2Ph), 6.70–6.90 (m, 6 H, C6H5), 7.20–7.45 (m, 4 H, C6F5) ppm. 13C NMR {1H}: δ = 11.8 (C6Me5), 17.6 (C6Me5), 18.8 (C6Me5), 42.2 (C–CH2Ph), 54.8 (Ta–CH2Ph), 120.1 (C6Me5), 123.5–149.2 (C6Me5, C6H5 and C6F3), 237.7 (Ta–C) ppm. 19F NMR: δ = −130.7 (a–C6F3), −158.3 (p–C6F3), −163.8 (m–C6F3) ppm. IR (KBr): ν = 1601 (C≡N) cm−1.

[TaCp*Cl{η2–C(Me)=NAr}{O–B(C6F5)3}] (3b): The procedure used for 2a, but starting from [TaCp*Me2–O{B(C6F5)3}] (1b) (0.50 g, 0.57 mmol) and CNAr (0.079 g, 0.960 mmol), gave 2b (0.52 g, 91%). C6H5BF2ClF15O2Ta (1005.41): calc. C 46.59, H 3.09, N 1.28. 1H NMR: δ = 0.79 (s, 3 H, Ta–Me), 1.48 (s, 3 H, Me2C6H5), 1.69 (s, 3 H, Me2C6H5), 1.95 (s, 15 H, C6Me5), 2.72 (s, 3 H, C–Me), 7.06 (m, 2 H, Me2C6H5), 7.13 (m, 1 H, Me2C6H5) ppm. 13C NMR {1H}: δ = 11.0 (C6Me5), 17.2 (Me2C6H5), 18.6 (Me2C6H5), 20.0 (Ta–Me), 31.6 (C–Me), 118.7 (C6Me5), 127.6–148.8 (Me2C6H5 and C6F3), 238.3 (Ta–C=O) ppm. 19F NMR: δ = −131.7 (a–C6F3), −158.4 (p–C6F3), −163.8 (m–C6F3) ppm. IR (KBr): ν = 1629 (C≡N) cm−1. \n
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(10 mL) was added to precipitate a white solid that was washed with KAPPA-CCD single crystal diffractometer. The crystal structure was solved by direct methods and refined using full-matrix least-squares on $F^2$ (SHELXL-97). All non-hydrogen atoms were anisotropically refined. Hydrogen atoms were geometrically placed and left riding on their parent atoms. Two molecules of toluene crystal.

Crystal Structure Determination for 2d and 3a: Selected crystals were covered with perfluropolyether oil and mounted on a Nonius KAPPA-CCD single crystal diffractometer. The crystal structure was solved by direct methods and refined using full-matrix least-squares on $F^2$ (SHELXL-97). All non-hydrogen atoms were anisotropically refined. Hydrogen atoms were geometrically placed and left riding on their parent atoms. Two molecules of toluene crystal.

Table 1. Crystallographic data for compounds 2d and 3a.

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References


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