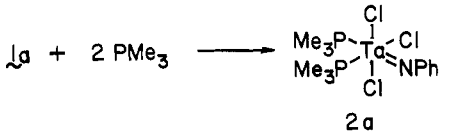
tatively.[7] It seems reasonable that a Ta═N bond should replace a Ta═C bond, since the often nearly linear metal–imido bond is closer to a triple bond than a double bond.[1] The cis,mer configuration for **1** is proposed on the basis of the presence of two types of THF ligands in the ^1H NMR spectra.

The reaction between **1a** and PMe_3 gives light yellow **2a** (eq 2). The cis,mer configuration of **2a** is based on inequivalence

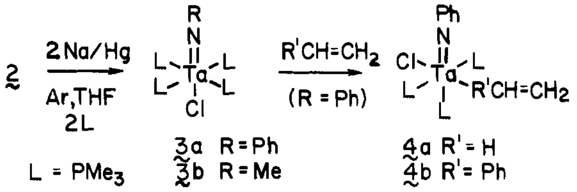

(2)

2a

of the PMe_3 ligands (−11.65 and −40.0 ppm in ^{31}P NMR spectrum at −30 °C) and faster exchange of one of them (we propose the one trans to the imido ligand) with added PMe_3 at temperatures above ~−30 °C. Interestingly, the orange PEt_3 derivative (**2b**) has a trans,mer geometry on the basis of its ^{31}P NMR spectrum (+15.5 ppm). A cis,fac geometry seems less likely but cannot be excluded by this data alone.

No ^{15}N NMR and few IR studies of labeled imido complexes have been reported.[1] The ^{15}N NMR spectrum[8a] of **1a**-^{15}N shows a peak at 369 ppm (referenced to liquid NH_3[8b]) while that for **2b**-^{15}N shows a singlet at 353 ppm. Coupling of ^{15}N to the two cis ^{31}P nuclei in **2b** evidently is small. In the IR spectrum of **1a** there is a medium-strength peak at ~1365 cm^{-1} and in the spectrum of **2b** at ~1345 cm^{-1}. The IR spectra of the ^{15}N-labeled complexes are identical with the unlabeled ones except these peaks are shifted to ~1335 and ~1315 cm^{-1}, respectively. The peak in the 1350-cm^{-1} region in each case is probably some combination of the Ta═N stretching mode coupled to the N—C stretching mode.[1,9]

The reduction of **2** with 2 equiv of sodium amalgam gives green **3a** in high yield (eq 3). This Ta(III) imido complex, like Ta-$(CHCMe_3)(PMe_3)_4Cl$,[10] its carbon analogue, did not analyze well due to the lability of the PMe_3 ligands. Therefore it was identified by NMR methods.[11] The proposed structure is based on the coupling of the imidomethyl group in **3b** equally to four phosphorus nuclei (J_{HP} = 3.5 Hz). One PMe_3 ligand can be displaced readily from **3a** by ethylene or styrene to give **4**.[12] The structure of **4** is believed to be mer with Cl cis to the imido ligand since one and only one phosphine ligand (we propose the one trans to the imido ligand) is lost in solution and exchanges readily on the NMR time scale with added PMe_3. The olefin must lie in the TaL_2Cl plane since the two ethylene carbon atoms are identical in the ^{13}C NMR spectrum. The Ta═N—R stretching frequencies are proposed to be 1340 cm^{-1} in **3a** and 1355 cm^{-1} in **4a**, although these assignments need to be confirmed by ^{15}N labeling studies.


(3)

3a R = Ph
3b R = Me
4a R′ = H
4b R′ = Ph

For some time we have been trying to prepare an imidoalkylidene complex in order to test whether the imido ligand, a good π-electron donor, would cause the alkylidene ligand to react with olefins to give metathesis products, just as alkoxide ligands apparently do.[13] Such a species can be prepared by using an organic azide,[1,14] as shown in eq 4. Complex **5**[15] is believed to be


(4)

5

structurally similar to bisneopentylidene complexes.[16] It sublimes at 120 °C and 1 μm without decomposing. It reacts slowly with ethylene (40 psi, 40 °C, 3 h) to give a quantitative yield of the two expected products of *rearrangement* of an intermediate metallacyclobutane complex, 4,4-dimethyl-1-pentene (80%), and *trans*-4,4-dimethyl-2-pentene (20%), but no metathesis products. It reacts with 1 equiv of acetone to give a 90% yield of 2,4,4-trimethyl-2-pentene[5] and a white, insoluble complex which shows a Ta═O stretch at ~835 cm^{-1} in the IR spectrum.

Complexes **1–4** react with ketones and aldehydes to give imines[17] in high yield (e.g., eq 5). The initial metal-containing

$$\underset{\mathbf{1a}}{Ta(NPh)(THF)_2Cl_3} + PhCHO \rightarrow PhCH{=}NPh \qquad (5)$$

products probably are oxo analogues of the imido complexes, but none has been identified. (We believe that **5** reacts under mild conditions with acetone to give only 4,4-dimethylpentenes because the initial oxo complex is insoluble.) These results suggest that for "metathesis-like" reactions the preference for bonding to tantalum is O > NR > CHR.

Acknowledgment. We thank the National Science Foundation for support (Grant CHE 79-05307).

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[7] Anal. R = Ph, Calcd for $TaC_{14}H_{21}O_2Cl_3N$: C, 32.17; H, 4.05. Found: C, 31.91; H, 3.99. R = CH_3, Calcd for $TaC_9H_{19}O_2Cl_3N$: C, 23.47; H, 4.16. Found: C, 23.09; H, 4.18. R = CMe_3, Calcd for $TaC_{12}H_{25}O_2Cl_3N$: C, 28.67; H, 5.01. Found: C, 28.81; H, 5.26.

[8] (a) Recorded at 9.04 MHz in benzene containing ~5% $Cr(acac)_3$ on a JEOL FX90Q spectrometer. (b) Levy, G. C.; Lichter, R. L. "Nitrogen-15 Nuclear Magnetic Resonance Spectroscopy"; Wiley: New York, 1979.

[9] We thank the referees for pointing this out.

[10] Fellmann, J. D.; Turner, H. W.; Schrock, R. R. *J. Am. Chem. Soc.* **1980**, *102*, 6608–6609.

[11] For **3b**, ^1H NMR (C_6D_6, 250 MHz) δ 1.45 (t, 36, J_{HP} = 2.4, PMe_3), 2.96 (qt, 3, J_{HP} = 3.5 NCH_3). $^{31}P\{^1H\}$ ($C_6D_5CD_3$, 36 MHz): δ −7.8 (brs, s); $^{13}C\{^1H\}$ NMR ($C_6D_5CD_3$, 22 MHz, −20 °C) δ 22.2 (PMe_3), 48.0 (NCH_3).

[12] For **4a**, ^1H NMR ($C_6D_5CD_3$, 250 MHz) δ 1.19 (d, 9, PMe_3), 1.31 (t, 18, PMe_3), 1.58 (m, 2, C_2H_4), 1.77 (m, 2, C_2H_4), 6.49–6.94 (m, 5, phenyl); $^{31}P\{^1H\}$ NMR ($C_6D_5CD_3$, 36 MHz) δ −22.9 (t, J_{PP} = 14.6 Hz), −11.7 (d, J_{PP} = 14.6 Hz).

[13] Schrock, R. R.; Rocklage, S.; Wengrovius, J. H.; Rupprecht, G. A.; Fellmann, J. D. *J. Mol. Catal.* 1980, *8*, 73–83.

[14] (a) McGlinchey, M. J.; Stone, F. G. A. *J. Chem. Soc., Chem. Commun.* **1970**, 1265. (b) Cenini, S.; LaMonica, G. *Inorg. Chim. Acta* **1976**, *18*, 279.

[15] ^1H NMR (C_6D_6, 250 MHz) δ 0.21 (s, 9, $NSi\mathit{Me}_3$), 1.16 (t, 18, J_{PH} = 3.3 Hz, PMe_3), 1.25 (s, 9, $CHC\mathit{Me}_3$), 7.98 (s, 1, $C\mathit{H}CMe_3$). ^{13}C NMR (C_6D_6, 22 MHz): δ 4.6 (q, J_{CH} = 126 Hz, $NSiMe_3$), 34.9 (q, J_{CH} = 125 Hz, $CHC\mathit{Me}_3$), 15.7 (t, J_{CH} = 126 Hz, PMe_3), 43.8 (s, $CH\mathit{C}Me_3$), 275.8 (d, J_{CH} = 99 Hz, $\mathit{C}HCMe_3$).

[16] (a) Fellmann, J. D.; Rupprecht, G. A.; Wood, C. D.; Schrock, R. R. *J. Am. Chem. Soc.* **1978**, *100*, 5964. (b) Churchill, M. R.; Youngs, W. J. *Inorg. Chem.* **1979**, *18*, 1930.

[17] Nugent noted that $Ta(NR)(NR_2)_3$ reacts with ketones to give imines: Nugent, W. A.; Harlow, R. L. *J. Chem. Soc., Chem. Commun.* **1978**, 578.

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Tantalum Complexes Containing Diimido Bridging Dinitrogen Ligands

Sir:

There is no published example of a characterized dinitrogen complex containing a group 5 metal.[1] Here we show how several tantalum complexes containing a bridging dinitrogen ligand can

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[1] (a) Chatt, J.; Dilworth, J. R.; Richards, R. L. *Chem. Rev.* **1978**, *78*, 589–625. (b) Chatt mentions[1a] that $[Nb(dmpe)_2Cl]_2N_2$ is obtained in low yield upon reducing $Nb(dmpe)_2Cl_4$ with Mg in THF under N_2 (Chatt, J.; Burt, R. J. Unpublished results).

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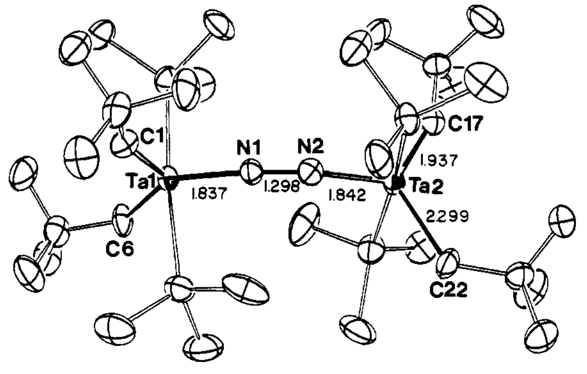

Figure 1. ORTEP-II diagram of the molecule, with hydrogen atoms omitted for clarity. The equatorial ligands about Ta1 are C1, C6, and N1, and about Ta2 are C17, C22, and N2.

be prepared and show by a crystal structure of one of them that the bridging dinitrogen is "diimidolike".

The two-electron, sodium amalgam reduction of Ta$(CHCMe_3)(PMe_3)_2Cl_3$ in the presence of PMe_3 under argon gives $Ta(CHCMe_3)(PMe_3)_4Cl$.[2] Under dinitrogen the product is an extremely air-sensitive, THF- and benzene-soluble, yellow powder (~50% yield) whose ^1H, ^{13}C, and ^{31}P NMR spectra suggest that it contains a neopentylidene ligand and two trans, equivalent PMe_3 ligands.[3] It must contain a chloride ligand since it can be alkylated by $LiCH_2CMe_3$ or LiMe. Elemental analyses show that approximately one nitrogen atom per Ta is present.[4] We formulate these complexes as bridging dinitrogen complexes as shown in eq 1. **1a** also forms slowly in pentane solutions of $Ta(CHCMe_3)(PMe_3)_4Cl$ under dinitrogen. The reverse reaction does not occur in neat PMe_3 at 70 °C.

$$\underset{\mathrm{L = PMe_3}}{\mathrm{Ta(CHCMe_3)L_2Cl_3}} \xrightarrow[\mathrm{L,\,N_2}]{\mathrm{2Na/Hg}} \underset{\mathbf{1a}}{\mathrm{[Ta(CHCMe_3)L_2Cl]_2N_2}} \xrightarrow{\mathrm{LiR}} \underset{\substack{\mathbf{1b},\ \mathrm{R = CH_2CMe_3} \\ \mathbf{1c},\ \mathrm{R = Me}}}{\mathrm{[Ta(CHCMe_3)L_2R]_2N_2}} \quad (1)$$

Stirring a pentane solution of $Ta(CHCMe_3)(PMe_3)_4Cl$ under $^{15}N_2$ for 24 h gives an ~50% yield of **1a**-$^{15}N_2$. The IR spectrum of **1a**-$^{15}N_2$ differs from that of **1a**-$^{14}N_2$ in only one respect. A medium-strength, sharp peak at 847 cm^{-1} in the IR spectrum of **1a**-$^{14}N_2$ shifts to 820 cm^{-1}. Since this type of molecule is not centrosymmetric, we do not know if this is an N–N stretching mode, a Ta–N stretching mode, or a combination mode.

The ^{15}N NMR spectrum of **1a**-$^{15}N_2$[5] consists of a singlet at 414 ppm (referenced to liquid NH_3[6]). This chemical shift should be compared with chemical shifts of the order of 350–370 ppm for imido complexes such as $Ta(NPh)(THF)_2Cl_3$,[7] and, equally importantly, with the rather different chemical shift of 679 ppm for the bridging dinitrogen in $[Zr(\eta^5\text{-}C_5Me_5)_2(^{15}N_2)]_2{}^{15}N_2$.[6,8]

The overall molecular geometry of **1b** is shown in Figure 1.[9] It is a binuclear molecule with no crystallographically imposed symmetry. Each tantalum atom is in a trigonal-bipyramidal coordination environment in which the PMe_3 ligands occupy the axial sites (Ta–P = 2.582 (3)–2.601 (3) Å) and a neopentyl, a neopentylidene, and a μ-N_2 ligand occupy the equatorial sites. Each half of the molecule therefore resembles $Ta(CHCMe_3)_2(mesityl)(PMe_3)_2$,[11] and the two halves are close to orthogonal (dihedral angle between equatorial planes = 82.6°). The tantalum–neopentyl bond lengths are Ta(1)–C(6) = 2.285 (10) and Ta(2)–C(22) = 2.299 (10) Å, with Ta–C(α)–C(β) angles of 129.4 (7)° and 127.6 (7)°. The tantalum–neopentylidene linkages are substantially shorter, with Ta(1)=C(1) = 1.932 (9) and Ta(2)=C(17) = 1.937 (9) Å; the Ta=C(α)—C(β) angles are 158.5 (7)° and 160.3 (7)°, respectively.

The bridging dinitrogen ligand is the most unusual feature of this molecule. The tantalum–nitrogen bond lengths are shorter than the tantalum–neopentylidene linkages (Ta(1)–N(1) = 1.837 (8) and Ta(2)–N(2) = 1.842 (8) Å) and the N(1)–N(2) bond is very long (1.298 (12) Å). The Ta–(μ-N_2)–Ta unit is close to linear, with ∠ Ta(1)–N(1)–N(2) = 171.4 (7) and ∠ Ta(2)–N(2)–N(1) = 172.4 (7)°. There are other examples of short M–μ-N_2 bond lengths where M is a third row metal such as Re,[1a] but there is no simple bridging dinitrogen complex in which the N–N bond is so long.[1a,12] In particular, these results contrast sharply with studies of $[(\eta^5\text{-}C_5Me_5)_2Zr(N_2)]_2(\mu\text{-}N_2)$[13a] and $[(\eta^5\text{-}C_5Me_5)_2Ti]_2(\mu\text{-}N_2)$,[13b] where the metal–nitrogen distances are substantially longer and the μ-N–N distances shorter (N–N = 1.182 (5) and 1.155 (14)–1.165 (14) Å, respectively). We propose that the μ-N_2 ligand is close to being a unique "diimido type", Ta=N—N=Ta.[14] A mononuclear imido analogue is known.[7]

Several other findings corroborate this point of view and suggest further that this type of dinitrogen complex will be a common one in Nb and Ta chemistry. First, THF and PEt_3 complexes which are analogous to $Ta(NPh)L_2Cl_3$ complexes[7] can be prepared quantitatively from *cis,mer*-$Ta(CHCMe_3)(THF)_2Cl_3$ (eq 2 and 3). Secondly, a dinitrogen ethylene complex analogous to $Ta(NPh)(PMe_3)_3(C_2H_4)Cl$[7] can be prepared as shown in eq 4.

$$\mathrm{2Ta(CHCMe_3)(THF)_2Cl_3} \xrightarrow[\mathrm{-2PhCH{=}CHCMe_3}]{\mathrm{PhCH{=}N{-}N{=}CHPh}} \underset{\mathbf{2a}^{15}}{\mathrm{[(THF)_2Cl_3Ta{=}N\text{-}]_2}} \quad (2)$$

$$\mathbf{2a} + \mathrm{4PEt_3} \rightarrow \underset{\mathbf{2b}^{16}}{\mathrm{[(PEt_3)_2Cl_3Ta{=}N\text{-}]_2}} \quad (3)$$

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(2) Fellmann, J. D.; Turner, H. W.; Schrock, R. R. *J. Am. Chem. Soc.* **1980**, *102*, 6608–6609.

(3) ^1H NMR (C_6D_6, 270 MHz) δ 8.79 (br s, 1, C*H*CMe$_3$), 1.32 (t, 18, $^2J_{HP}$ = 3.5 Hz, PMe_3), 1.29 (s, 9, CHC*Me*$_3$); ^{13}C NMR (C_6D_6, 15.0 MHz) δ 274.5 (br d, $^1J_{CH}$ = 91 Hz, *C*HCMe$_3$), 46.6 (s, CH*C*Me$_3$), 35.0 (q, $^1J_{CH}$ = 125 Hz, CHC*Me*$_3$), 15.2 (qt, $^1J_{CP}$ = 12.0 Hz, $^1J_{CH}$ = 129 Hz, PMe_3); ^{31}P NMR ($C_6D_5CD_3$, 109.3 MHz, 30 and −30 °C) δ −11.2 (s).

(4) Anal. Calcd for $[Ta(CHCMe_3)(CH_2CMe_3)(PMe_3)_2]_2N_2$: N, 2.87. Found, 2.52.

(5) Recorded at 9.04 MHz in the presence of ~5% $Cr(acac)_3$ on a JEOL FX-90Q spectrometer.

(6) Levy, G. C.; Lichter, R. L. "Nitrogen-15 Nuclear Magnetic Resonance Spectroscopy," Wiley: New York, 1979.

(7) See: Rocklage, S. M.; Schrock, R. R. *J. Am. Chem. Soc.* **1980**, *102*, preceding paper in this issue.

(8) Manriquez, J. M.; McAlister, D. R.; Rosenberg, E.; Shiller, A. M.; Williamson, K. L.; Chan, S. I.; Bercaw, J. E. *J. Am. Chem. Soc.* **1978**, *100*, 3078–3083.

(9) Triclinic crystals grown from pentane. Space group $P\bar{1}$, with a = 9.712 (2) Å, b = 14.845 (2) Å, c = 18.448 (4) Å, α = 67.278 (14)°, β = 84.812 (15)°, and γ = 71.743 (12)°. The cell volume is 2328.5 (7) Å^3 and ρ(calcd) = 1.39 g cm^{-3} for Z = 2 and mol wt = 976.9. Diffraction data ($4.0 < 2\theta < 40°$) were collected at 23 (1) °C by using a Syntex P2$_1$ diffractometer equipped with graphite-monochromatized Mo Kα radiation.[10] Data were corrected for absorption and the structure was solved by a combination of Patterson and difference-Fourier techniques. Full-matrix least-squares refinement using anisotropic thermal parameters for all 40 nonhydrogen atoms, with all 78 hydrogen atoms included in calculated positions, converged with R_F = 4.5% and R_{wF} = 3.1% for all 4308 independent data (none rejected) and R_F = 3.1% and R_{wF} = 3.0% for those 3626 data with $|F_o| > 3\sigma(F_o)$.

(10) Churchill, M. R.; Lashewycz, R. A.; Rotella, F. J. *Inorg. Chem.* **1977**, *16*, 265–271.

(11) Churchill, M. R.; Youngs, W. J. *Inorg. Chem.* **1979**, *18*, 1930–1935.

(12) Slightly longer N–N bonds (~1.35 Å) have been reported in complexes such as $[(PhLi)_6Ni_2N_2(ether)_2]_2$ and related species: Krüger, C.; Tsay, Y.-H. *Angew. Chem., Int. Ed. Engl.* **1973**, *12*, 998,999.

(13) (a) Sanner, R. D.; Manriquez, J. M.; Marsh, R. E.; Bercaw, J. E. *J. Am. Chem. Soc.* **1976**, *98*, 8351–8357. (b) Sanner, R. D.; Duggan, D. M.; McKenzie, T. C.; Marsh, R. E.; Bercaw, J. E. *J. Am. Chem. Soc.* **1976**, *78*, 8358–8365.

(14) A referee suggested that a resonance structure such as $Ta^-{=}N^+{=}N^+{=}Ta^-$ might be equally appropriate.

(15) Insoluble orange powder quantitatively. The $\nu_{Ta_2N_2}$ region is obscured by THF modes.

(16) Green crystals quantitatively; $\nu_{Ta_2N_2}$ = 855 cm^{-1} (cf. **1a**). ^{31}P NMR: δ −14.2 (trans,mer). An analogous PEt_2Ph derivative analyzed for one N per Ta.

$$trans,mer\text{-}Ta(C_2H_4)(PMe_3)_2Cl_3{}^{17} \xrightarrow[N_2,\ Na/Hg,\ 2\ h]{PMe_3,\ ether/THF} \underset{3^{18}}{{}^{1}/_{2}[Cl(PMe_3)_3(C_2H_4)Ta{=}N\text{-}]_2} \quad (4)$$

Extensive ^1H, ^{13}C, and ^{31}P NMR studies suggest that the two ends of this molecule are "locked" ~90° to one another when the phosphine on each Ta which is trans to the μ-N_2 ligand stops exchanging rapidly on the NMR time scale (cf. **1b**). Thirdly, **2b** and **3** react with acetone to give dimethylketazine quantitatively (eq 5). This reaction is analogous to the reaction of Ta-

$$\mathbf{2b}\ \text{or}\ \mathbf{3} \xrightarrow[8\ h,\ 25\ °C]{excess\ Me_2C{=}O} Me_2C{=}N{-}N{=}CMe_2 \quad (5)$$

$(NPh)(PEt_3)_2Cl_3$ with the carbonyl function to give an imine.[7] To our knowledge, this reaction of bridging dinitrogen is unique.

Acknowledgment. This work was supported by NSF Grants CHE79-05307 (to R.R.S.) and CHE79-24560 (to M.R.C.).

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(17) Fellmann, J. D.; Rupprecht, G. A.; Schrock, R. R. *J. Am. Chem. Soc.* **1979**, *101*, 5099-5101.

(18) Ether-soluble yellow-orange powder, 60% yield; $\nu_{Ta_2N_2}$ = 825 cm^{-1} (cf. **1a**, **2b**).

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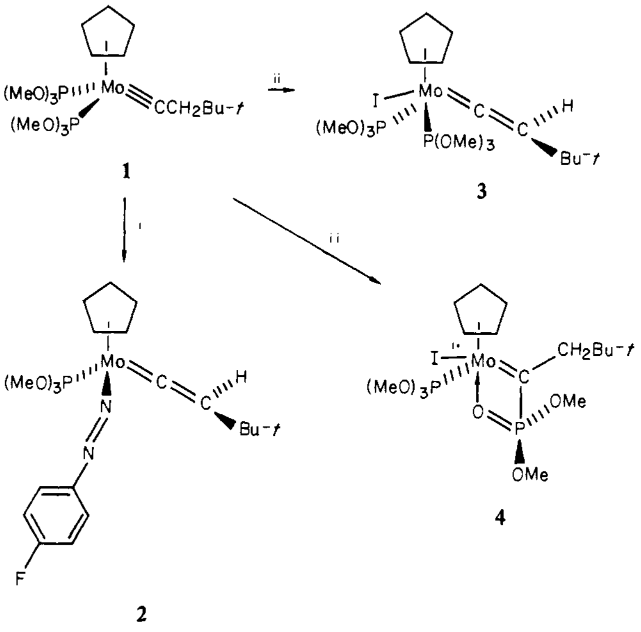
Transformation of a Molybdenum-Alkylidyne Complex into Molecules Containing Vinylalkylidene and Alkylidene Ligands. Crystal and Molecular Structure of $[\overline{IMo{=}C(CH_2Bu\text{-}t)P(O})(OMe)_2(P(OMe)_3)(\eta^5\text{-}C_5H_5)]$

Sir:

Alkylidene transition-metal complexes in which the carbon atom bonded directly to the metal carries either a partial negative[1] or positive charge[2] have been described. This distinction has been emphasized by the use of the descriptive terms—Schrock or Fischer-type complexes. Group 6 alkylidene chemistry has, so far, been largely dominated by Fischer-type complexes despite the obvious importance of expanding the range of alkylidene complexes in this area of the periodic table. The availability of the alkylidyne complex **1**[3] $[Mo{\equiv}CCH_2Bu\text{-}t(P(OMe)_3)_2(\eta^5\text{-}C_5H_5)]$, whose synthesis we have previously described, suggested a possible new synthetic approach to species containing carbon to molybdenum double bonds.

Reaction of **1** with 4-fluorophenyldiazonium tetrafluoroborate in methylene chloride at room temperature led to the rapid formation of a deep red solution. Chromatography of the reaction mixture on an alumina packed column afforded (31% yield) from hexane (−78 °C) dark red crystals, which were characterized by elemental analysis and mass, IR, and NMR spectroscopy[4] as the vinylalkylidene complex **2**. (See Scheme I.) Thus, an apparent *two*-electron oxidative reaction has occurred between an aryldiazonium salt[5] and an alkylidyne complex with loss of a molecule of trimethyl phosphite. It is likely that the displaced phosphite facilitates loss of HBF_4 from an intermediate cationic species to give the neutral vinylalkylidene complex **2**, containing a chiral molybdenum center.

Scheme I^a



^{a} (i) $4\text{-}FC_6H_4N_2BF_4/CH_2Cl_2$, (ii) CF_3I/Et_2O.

A vinylalkylidene complex was also obtained as one of the products on treatment of complex **1** with trifluoroiodomethane in diethyl ether at room temperature. Column chromatography on alumina at low temperature (−30 °C) led, on elution with hexane, to the separation of the red crystalline (−20 °C, hexane) product **3**, which was identified[6] as a molybdenum vinylalkylidene with the illustrated structure. Recent calculations[7,8] suggest that in **3** the plane of the CHBu-*t* fragment should bisect the molecular symmetry plane as shown, although the barrier to rotation is calculated to be relatively low. This prediction was borne out by the observation of *one* ^{31}P resonance at −90 °C for the two phosphite ligands present in **3**.

The formation of **3** probably involves an initial oxidative reaction followed by elimination of a proton, this being supported by the

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(1) Schrock, R. R. *Acc. Chem. Res.* **1979**, *12*, 98-104.

(2) Fischer, E. O.; Schubert, U. *J. Organomet. Chem.* **1975**, *100*, 59-81. Cardin, D. J.; Cetinkaya, B.; Lappert, M. F. *Chem. Rev.* **1972**, *72*, 545-574. Cotton, F. A.; Lukehart, C. M. *Prog. Inorg. Chem.* **1972**, *16*, 487-613.

(3) Bottrill, M.; Green, M. *J. Am. Chem. Soc.* **1977**, *99*, 5795-5796.

(4) The spectroscopic data for complex **2** is as follows: γ(Nujol) 1608 m (Mo=C=C), 1594 m ($MoN_2C_6H_4F$) cm^{-1}; ^1H NMR ($C_6D_5CD_3$, room temperature) τ 2.49 (m, 2 H, FC_6H_4), 3.08 (t, 2 H, FC_6H_4, J_{HH} = 8 Hz), 4.15 (d, 1 H, C=C*H*Bu-*t*, J_{PH} = 6 Hz), 4.61 (s, 5 H, C_5H_5), 6.64 (d, 9 H, POMe, J_{PH} = 12 Hz), 8.78 (s, 9 H, Bu-*t*); ^{13}C NMR ($C_6D_5CD_3$, room temperature, ^1H decoupled) δ 31.2 (s, C*Me*$_3$), 34.7 (s, *C*Me$_3$), 52.0 (s, POMe), 96.7 (d, C_5H_5, J_{CP} = 22 Hz), 115.5 (d, FC_6H_4, J_{CF} = 25 Hz), 120.5 (d, FC_6H_4, J_{CF} = 7 Hz), 141.3 (s, C=*C*HBu-*t*), 146.4 (s, FC_6H_4), 159.4 (d, *F*C$_6$H$_4$, J_{CF} = 242 Hz), 348.6 (d, Mo=*C*=CHBu-*t*, J_{CP} = 30 Hz); ^{31}P NMR ($C_6D_5CD_3$, room temperature, ^1H decoupled) δ 184.7 (s).

(5) Oxidative-addition reactions between low-valent metal complexes and aryldiazonium salts have been previously observed; see: Sutton, D. *Chem. Soc. Rev.* **1975**, *4*, 443-470.

(6) The spectroscopic data for complex **3** is as follows: γ(Nujol) 1605 m (Mo=C=C) cm^{-1}; ^1H NMR (C_6D_6, room temperature) τ 4.69 (t, 1 H, C=C*H*Bu-*t*, J_{PH} = 13 Hz), 5.08 (t, 5 H, C_5H_5, J_{PH} = 2 Hz), 6.32 (t, 18 H, POMe, J_{PH} = 5 Hz), 8.94 (s, 9 H, Bu-*t*); ^{13}C NMR (C_6D_6, room temperature, ^1H decoupled) δ 31.3 (t, C*Me*$_3$, J_{CP} = 3 Hz), 35.0 (t, *C*Me$_3$, J_{CP} = 6 Hz), 54.5 (t, POMe, J_{CP} = 3 Hz), 91.0 (t, C_5H_5, J_{CP} = 20 Hz), 132.7 (t, C=*C*HBu-*t*, J_{CP} = 12 Hz), 326.4 (t, Mo=*C*=CHBu-*t*, J_{CP} = 51 Hz); ^{31}P NMR (C_6D_6, room temperature, H decoupled) δ 157.9 (s); ^{31}P NMR ($C_6D_5CD_3$, −90 °C, ^1H decoupled) δ 159.3 (s).

(7) Schilling, B. E. R.; Hoffmann, R.; Lichtenberger, D. L. *J. Am. Chem. Soc.* **1979**, *101*, 585-590.

(8) Schilling, B. E. R.; Hoffmann, R.; Faller, J. W. *J. Am. Chem. Soc.* **1979**, *101*, 592-598.