full-matrix least-squares refinement was based on 1204 observed reflections ($I > 3.00\sigma(I)$) and 154 variable parameters and converged (largest parameter shift was 0.00 times its esd) with unweighted and weighted agreement factors of $R = \sum ||F_o| - |F_c||/\sum|F_o| = 0.054$ and $R_w = [(\sum w(|F_o| - |F_c|)^2/\sum wF_o^2)]^{1/2} = 0.063$.
The standard deviation of an observation of unit weight was 1.97. The maximum and minimum peaks on the final difference Fourier map corresponded to 0.30 and −0.27 e⁻/Å³, respectively.
Neutral atom scattering factors were taken from Cromer and Waber.[26] Anomalous dispersion effects were included in F_c;[27] the values of $\Delta f'$ and $\Delta f''$ were those of Cromer.[28] All calculations were performed with use of the TEXSAN[29] crystallographic software

(26) Cromer, D. T.; Waber, J. T. *International Tables for X-ray Crystallography*; The Kynoch Press: Birmingham, England, 1974; Vol. IV, Table 2.2A.
(27) Ibers, J. A.; Hamilton, W. C. *Acta Crystallogr.* **1964**, *17*, 781.
(28) Cromer, D. T. *International Tables for X-ray Crystallography*; The Kynoch Press: Birmingham, England, 1974; Vol. IV, Table 2.2A.

package of Molecular Structure Corp.
Table VII is a listing of the positional parameters and the B(eq) values for the atoms.

Registry No. 1, 129152-58-5; 1,1,3,3-tetramethyl-1,3-disilacyclobutane, 1627-98-1.

Supplementary Material Available: Tables of U values, intramolecular distances involving the hydrogen atoms, and torsion or conformation angles (5 pages); listings of observed and calculated structure factor amplitudes (9 pages). Ordering information is given on any current masthead page.

(29) TEXSAN—TEXRAY Structure Analysis Package, Molecular Structure Corp., 1985.

Bis((2,6-diisopropylphenyl)imido) Complexes of Molybdenum(VI)

N. Bryson, M.-T. Youinou, and J. A. Osborn*

Laboratoire de Chimie des Métaux de Transition et de Catalyse (URA 424 du CNRS), 4 Rue Blaise Pascal, 67000 Strasbourg, France

Summary: The molybdenum(VI) complex (Ar′N)₂MoCl₂·THF (Ar′ = 2,6-diisopropylphenyl) was prepared by reaction of MoO₂Cl₂ with Ar′NCO at 70 °C in THF. The preparations of the derivatives Mo(NAr′)₂Np₂, Mo(NAr′)₂Mes₂, Mo(NAr′)₂MesBr, Mo(NAr′)₂(NEt₂)Cl, and Mo(NAr′)₂(NEt₂)₂ from Mo(NAr′)₂Cl₂·THF are described. In addition, the complex Mo(NAr′)₂(NHAr′)₂ has been synthesized and characterized crystallographically. Crystal data: MoC₄₈H₇₀N₄, M_r = 799.06, monoclinic, $P2_1/n$, a = 9.874 (3) Å, b = 24.004 (8) Å, c = 19.220 (7) Å, β = 93.34 (2) Å, V = 4547 Å³, Z = 4, ρ_{calc} = 1.167 g cm⁻³, λ = 1.5418 Å, μ = 26.35 cm⁻¹, T = −100 °C, R = 4.22% for 4426 unique observed $[I \geq 3\sigma(I)]$ reflections, R_w = 7.65%.

The organoimido ligand =NR²⁻ exerts a stabilizing influence on high oxidation states similar to that of the oxo ligand O²⁻ but additionally can provide, by modification of the R groups, variations in electronic and steric effects. For example, complexes involving imido groups have found use as designed electrophilic catalysts for olefin metathesis[1,2] and for C–H activation.[3] Also, interesting protonation–deprotonation reactions of tungsten and molybdenum imido complexes have recently been reported.[4,5] We have been interested in generating coordinatively unsaturated

molybdenum(VI) complexes of the (2,6-diisopropylphenyl)imido ligand (Ar′) for further investigations in the area of hydrocarbon activation. Although molybdenum complexes of this ligand exist, they have been prepared by rather tedious routes.[1] In this paper, we describe the convenient preparation of the (Ar′N)₂MoCl₂·THF complex, which serves as an important starting material for this chemistry. The synthesis of some alkyl (aryl) and amido derivatives are presented along with an X-ray structure of one such derivative.

Results and Discussion

The complex Mo(NAr′)₂Cl₂·THF (1) was prepared under an inert atmosphere by reacting freshly sublimed MoO₂Cl₂ with 2 equiv of Ar′NCO in THF at 70 °C according to a method previously reported.[2] The reaction was complete after 3 days. Following evaporation of the solvent, 1 was recovered as a dark red powder in excellent yield. Mo(NAr′)₂Cl₂·THF, which is very soluble in nonpolar organic solvents, was recrystallized from pentane to give analytically pure material. The product is very sensitive to air and moisture both in solution and in the solid state.

Alkylation of 1 using neopentyllithium in diethyl ether yielded the orange air-sensitive Mo(NAr′)₂Np₂ (2), which was recrystallized from pentane. If 1 equiv of LiNp was used, only the starting material 1 and the dineopentyl complex 2 (in a 1:1 ratio) were observed by ¹H NMR spectroscopy. Comparison of the ¹H NMR spectra of Mo(NAr′)₂Np₂ (2) and Mo(NBuᵗ)₂Np₂[2] prepared previously shows that the stronger electron-withdrawing arylimido ligand leads to a significant downfield shift of the neopentyl α-hydrogens from 1.89 to 2.35 ppm.

The compound Mo(NAr′)₂Cl₂·THF (1) was allowed to react with 2 equiv of mesitylmagnesium bromide (MesMgBr) in THF. After 12 h, the monosubstituted product Mo(NAr′)₂MesX (4) (X = Br) comprised ca. 75% of the reaction products as determined by ¹H NMR spectroscopy. The analytical results and mass spectrum

(1) (a) Schrock, R. R.; Murdzek, J. S.; Bazan, G.C.; Robbins, J.; DiMare, M.; O'Regan, M. *J. Am. Chem. Soc.* **1990**, *112*, 3875. (b) Murdzek, J. S.; Schrock, R. R. *Organometallics* **1987**, *6*, 1373–1374. (c) Schaverien, C. J.; Dewan, J. C.; Schrock, R. R. *J. Am. Chem. Soc.* **1986**, *108*, 2771.
(2) Schoettel, G.; Kress, J.; Osborn, J. A. *J. Chem. Soc., Chem. Commun.* **1989**, 1062.
(3) (a) Walsh, P. J.; Hollander, F. J.; Bergman, R. G. *J. Am. Chem. Soc.* **1988**, *110*, 8729. (b) Cummins, C. C.; Baxter, S. M.; Wolczanski, P. T. *J. Am. Chem. Soc.* **1988**, *110*, 8731.
(4) Danopoulos, A. A.; Wilkinson, G.; Hursthouse, M. B.; Hussain, B. *Polyhedron* **1989**, *8*, 2947.
(5) Danopoulos, A. A.; Wilkinson, G.; Hussain, B.; Hursthouse, M. B. *J. Chem. Soc., Chem. Commun.* **1989**, 896.

analysis of 4 revealed that an exchange between chloride and bromide had taken place. Lengthening the reaction time to several days gave the disubstituted complex Mo-(NAr')₂Mes₂ (3). The monomesityl derivative 4 was subsequently prepared in a separate experiment in which the reagents were mixed in a 1:1 ratio. Both 3 and 4 were isolated as red crystalline solids after recrystallization from pentane and are stable in the absence of air and moisture. Furthermore, the ¹H NMR spectrum of Mo(NAr')₂MesBr (4) shows that the complex does not undergo ligand redistribution (disproportionation) at room temperature. It thus appears that the stability of the monomesityl complex toward redistribution (as compared with the neopentyl analogue) must result from increased steric bulk about the metal. This may well imply that such reactions proceed via bridging dimeric intermediates.

Treatment of the dihalide Mo(NAr')₂Cl₂·THF (1) with 2 equiv of LiNEt₂ in diethyl ether gave a deep red solution, which was filtered to remove LiCl. Evaporation of the solvent, dissolution in pentane, and refrigeration precipitated a deep red crystalline solid (<5% yield based on Mo) whose IR and ¹H NMR spectra, as well as analysis, are consistent with the formulation Mo(NAr')₂(NEt₂)Cl (5). The complex is stable in solution and in the solid state in the absence of air and moisture. The complex Mo-(NAr')₂(NEt₂)₂ (6) was subsequently isolated as a red oil in high yield after complete precipitation of the monohalide Mo(NAr')₂(NEt₂)Cl (5) from the reaction mixture. Recrystallization of Mo(NAr')₂(NEt₂)₂ (6) from pentane gave a red-orange crystalline product; however, because of its extreme solubility only very low yields of crystalline solid could be obtained.

The bis(imido)–bis(amido) complex Mo(NAr')₂(NHAr')₂ (7) was prepared directly from the reaction of the dihalide 1 with 2 equiv of LiNHAr' (eq 1) (prepared at -20 °C by

$$Mo(NAr')_2Cl_2 + 2LiNHAr' \rightarrow Mo(NAr')_2(NHAr')_2 \quad (1)$$

reaction of Ar'NH₂ and LiBu) in diethyl ether in ca. 65% yield (¹H NMR). The formation of Mo(NAr')₂(NHAr')₂ (7) is accompanied by the formation of the amine Ar'NH₂ and a second molybdenum species, which appear in a 1:1 ratio. However, attempts to isolate this molybdenum species have as yet proven unsuccessful. Complex 7 is stable in solution, showing no decomposition in refluxing benzene over a period of several days but is not stable in the solid state and decomposes slowly at room temperature even in an inert atmosphere.

¹H NMR spectra of 7 showed a 1:1 ratio of the Mo=NAr' and Mo–NHAr' groups with doublets at 1.02 and 1.25 ppm and septets at 3.38 and 3.85 ppm (²J_{H-H} = 6.9 Hz). Two sets of multiplets for the phenyl hydrogens appear at 6.92 and 7.08 ppm. The amido hydrogens, which showed an absorption band at 3323 cm⁻¹ in the infrared spectrum (Nujol mull), exhibit a singlet at 8.03 ppm. These results show that the protons in Mo(NAr')₂(NHAr')₂ are fixed at the amido nitrogens and that proton transfer from an amido to an imido does not occur at a significant rate to be observable by NMR spectroscopy. The corresponding W(NBuᵗ)₂(NHBuᵗ)₂, which has been prepared by Nugent,[6] shows a 1:1 ratio of the tert-butylimido and tert-butylamido groups, suggesting a similar behavior.

The structure of 7 was confirmed by X-ray diffraction. Experimental details are given in Table I, and positional and thermal parameters, in Table II. An ORTEP drawing is shown in Figure 1, and selected bond distances and bond angles are in Table III. The geometry about the metal

Table I. Crystallographic Data for Mo(NAr')₂(NHAr')₂

|	
chem formula	MoC₄₈H₇₀N₄
fw	799.06
cryst dimens, mm	0.360 × 0.340 × 0.160
cryst system	monoclinic
space group	$P2_1/n$
a, Å	9.874 (3)
b, Å	24.004 (8)
c, Å	19.220 (7)
β, deg	93.34 (2)
V, Å³	4547
ρ_{calc}, g cm⁻³	1.167
Z	4
$F(000)$	1712
λ, Å	1.5418
scan method	$\theta/2\theta$ flying step-scan
μ, cm⁻¹	26.35
temp, °C	-100
scan speed, deg min⁻¹	0.024
scan width, deg	0.9 + 0.14 tan θ
2θ range, deg	6–102
octants	$\pm h, +k, +l$
no. of reflcns measd, total	5216
no. of reflcns measd, unique	4426
criterion for observns	$I \geq 3\sigma(I)$
no. of variables	478
transm factors (max/min)	1.243/0.880
R/R_w, %	4.22/7.65
GOF	1.791
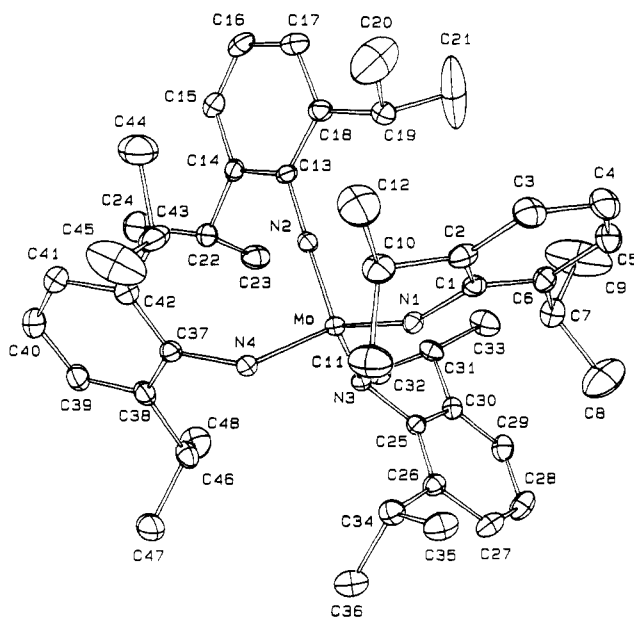
Figure 1. ORTEP drawing of Mo(NAr')₂(NHAr')₂ (7), showing 50% probability ellipsoids.

center is tetrahedral (N–Mo–N$_{av}$ = 109.5°), having one almost linear (Mo–N2–C13 = 172.3 (3)°) and one partially bend imido group (Mo–N1–C1 = 155.7 (3)°) and two bent amido groups (134.3 (2) and 123.4 (2)°). There is a considerable lengthening of the metal–imido distance in 7 (1.764 (2) and 1.753 (2) Å) as compared to that of Mo-(NBuᵗ)₂Mes₂[7] (1.708 (7) and 1.733 (7) Å) possibly due to the stronger π-donor properties of the amido ligands. However, the encumbrance due to the bulky diisopropylphenyl groups could also be expected to increase the length of the Mo=N bond. The geometry about N3 and N4 indicates a tendency toward planarity; the distances of the nitrogen atoms from the mean planes defined by Mo, C25, H'1 and Mo, C37, H'2 are 0.174 (3) and 0.142 (3)

(6) Nugent, W. A. *Inorg. Chem.* **1983**, *22*, 965.

(7) Sullivan, A. C.; Wilkinson, G.; Montevalli, M.; Hursthouse, M. B. *J. Chem. Soc., Dalton Trans.* **1988**, 53.

Table II. Final Positional Parameters for
Mo(NAr′)₂(NHAr′)₂

atom	x	y	z	$B,^a$ Å²
Mo	0.07579 (3)	0.10882 (1)	0.25483 (2)	2.501 (8)
N1	0.1009 (3)	0.0671 (1)	0.3300 (2)	2.87 (7)
N2	0.2093 (3)	0.1549 (1)	0.2409 (2)	2.90 (7)
N3	0.0585 (3)	0.0613 (1)	0.1701 (2)	2.85 (7)
N4	-0.0824 (3)	0.1565 (1)	0.2707 (2)	2.92 (7)
C1	0.1710 (4)	0.0469 (2)	0.3895 (2)	3.16 (9)
C2	0.1539 (4)	0.0738 (2)	0.4539 (2)	3.8 (1)
C3	0.2277 (5)	0.0542 (2)	0.5125 (2)	4.7 (1)
C4	0.3150 (5)	0.0082 (2)	0.5077 (2)	5.3 (1)
C5	0.3265 (5)	-0.0181 (2)	0.4464 (2)	4.8 (1)
C6	0.2537 (4)	-0.0005 (2)	0.3849 (2)	3.70 (9)
C7	0.2637 (5)	-0.0311 (2)	0.3172 (2)	4.2 (1)
C8	0.1949 (9)	-0.0867 (2)	0.3197 (3)	9.6 (2)
C9	0.4096 (7)	-0.0381 (5)	0.2969 (4)	15.0 (3)
C10	0.0577 (5)	0.1228 (2)	0.4569 (2)	4.5 (1)
C11	-0.0884 (6)	0.1014 (2)	0.4548 (3)	6.6 (2)
C12	0.0833 (7)	0.1591 (3)	0.5207 (3)	7.3 (2)
C13	0.3141 (4)	0.1932 (2)	0.2391 (2)	2.96 (8)
C14	0.3159 (4)	0.2290 (2)	0.1819 (2)	3.37 (9)
C15	0.4194 (5)	0.2684 (2)	0.1815 (2)	4.1 (1)
C16	0.5162 (5)	0.2723 (2)	0.2341 (3)	4.5 (1)
C17	0.5161 (5)	0.2363 (2)	0.2910 (2)	4.3 (1)
C18	0.4158 (4)	0.1955 (2)	0.2940 (2)	3.60 (9)
C19	0.4145 (5)	0.1566 (2)	0.3549 (2)	4.3 (1)
C20	0.425 (1)	0.1849 (3)	0.4235 (3)	11.7 (3)
C21	0.5285 (9)	0.1152 (3)	0.3520 (5)	16.4 (3)
C22	0.2102 (5)	0.2223 (2)	0.1214 (2)	4.1 (1)
C23	0.2539 (5)	0.1755 (2)	0.0733 (3)	5.6 (1)
C24	0.1831 (6)	0.2760 (2)	0.0791 (3)	7.2 (2)
C25	0.0401 (4)	0.0036 (2)	0.1595 (2)	2.99 (9)
C26	-0.0683 (4)	-0.0247 (2)	0.1876 (2)	3.58 (9)
C27	-0.0802 (5)	-0.0821 (2)	0.1765 (2)	4.7 (1)
C28	0.0096 (6)	-0.1109 (2)	0.1389 (3)	5.3 (1)
C29	0.1161 (5)	-0.0829 (2)	0.1114 (2)	4.5 (1)
C30	0.1341 (4)	-0.0264 (2)	0.1201 (2)	3.31 (9)
C31	0.2475 (4)	0.0047 (2)	0.0877 (2)	4.0 (1)
C32	0.1950 (5)	0.0297 (2)	0.0172 (2)	5.2 (1)
C33	0.3739 (5)	-0.0319 (3)	0.0762 (3)	6.5 (1)
C34	-0.1755 (5)	0.0049 (2)	0.2251 (2)	4.2 (1)
C35	-0.1934 (5)	-0.0183 (2)	0.2993 (2)	5.3 (1)
C36	-0.3116 (5)	0.0040 (2)	0.1819 (3)	5.6 (1)
C37	-0.1063 (4)	0.2091 (2)	0.2350 (2)	3.14 (9)
C39	-0.1653 (4)	0.2090 (2)	0.1670 (2)	3.9 (1)
C39	-0.1824 (5)	0.2595 (2)	0.1322 (2)	4.6 (1)
C40	-0.1430 (5)	0.3085 (2)	0.1634 (3)	5.3 (1)
C41	-0.0870 (5)	0.3084 (2)	0.2306 (3)	5.0 (1)
C42	-0.0655 (4)	0.2588 (2)	0.2673 (3)	4.0 (1)
C43	-0.0008 (5)	0.2599 (2)	0.3406 (3)	5.2 (1)
C44	0.1219 (6)	0.2993 (3)	0.3497 (3)	7.6 (2)
C45	-0.1061 (7)	0.2742 (4)	0.3932 (3)	10.6 (2)
C46	-0.2161 (5)	0.1558 (2)	0.1312 (2)	4.3 (1)
C47	-0.3700 (6)	0.1553 (2)	0.1230 (4)	7.1 (2)
C48	-0.1547 (6)	0.1488 (3)	0.0581 (3)	6.8 (2)

$^a B$ values for anisotropically refined atoms are given in the form of the isotropic equivalent displacement parameter defined as $(4/3) [a^2\beta(1,1) + b^2\beta(2,2) + c^2\beta(3,3) + ab(\cos \delta)\beta(1,2) + ac(\cos \beta)\beta(1,3) + bc(\cos \alpha)\beta(2,3)]$.

Table III. Selected Distances (Å) and Bond Angles (deg)
for Mo(NAr′)₂(NHAr′)₂^a

	Distances			
Mo–N1	1.764 (2)		N2–C13	1.387 (4)
Mo–N2	1.753 (2)		N3–C25	1.410 (4)
Mo–N3	1.987 (2)		N4–C37	1.449 (4)
Mo–N4	1.975 (2)		N3–H′1	1.064
N1–C1	1.389 (4)		N4–H′2	1.099

	Bond Angles			
N1–Mo–N2	114.4 (1)		N3–Mo–N4	115.4 (1)
N1–Mo–N3	110.3 (1)		Mo–N1–C1	155.7 (3)
N1–Mo–N4	106.1 (1)		Mo–N2–C13	172.3 (3)
N2–Mo–N3	105.4 (1)		Mo–N3–C25	134.3 (2)
N2–Mo–N4	105.4 (1)		Mo–N4–C37	123.4 (2)

^{a} Estimated standard deviations are given in parentheses.

Å. Such deviations could result from π-donor effects, but again steric factors cannot be excluded.

Several hydrogen atoms were observed during refinement of the structure; in particular, the amido NH hydrogens were among these with N–H distances of 1.064 and 1.099 Å. We find no evidence in the structure that these hydrogens are hydrogen-bonded either intra- or intermolecularly elsewhere in the lattice.

Recently, Wilkinson and co-workers[5] have isolated and characterized by X-ray crystallography the lithium salt of the dianion [W(NBu^t)₄]²⁻. In this structure, as in the Mo complex described above, only one of the imido groups was found to be strictly linear (176.6 (6)°, while the other three groups are bent between 134.4 and 139.6 (5)°. For 7 as well as [W(NBu^t)₄]²⁻, only one triple bond is actually required to satisfy the 18e rule, since each of the other three ligands can formally contribute 4 electrons. In terms of orbital interactions in this tetrahedral molecule, the overlap of a pπ orbital on each imido ligand with a single empty d orbital on the metal results in a 4-electron/3-center interaction that is somewhat antibonding in character. This can be in part resolved by bending the imido group, which would cause net stabilization of the molecule.

The complex (Ar′N)₂MoCl₂·THF (1) hence promises to be a useful starting material to a variety of derivatives of the bis(arylimido)molybdenum(VI) core, which we are continuing to explore.

Experimental Section

All manipulations were carried out under an inert atmosphere of argon. Solvents were distilled under nitrogen by using standard techniques. Dichlorodioxomolybdenum(VI) was purified by fractional sublimation. All other reagents were used as received without further purification.

Microanalyses were performed by the CNRS Microanalytical Laboratory at Strasbourg, France, or Lyon, France. IR spectra were recorded as Nujol mulls on a Perkin-Elmer 597 spectrometer; absorbances are listed in reciprocal centimeters as weak (w), medium (m), and strong (s). ¹H NMR spectra were recorded on a Bruker SY 200 instrument; chemical shifts (δ) are given in parts per million relative to (CH₃)₄Si in organic solvents. Mass spectra were performed on a Finnigan TFQ 70 instrument, using the chemical ionization technique.

Bis((2,6-diisopropylphenyl)imido)dichloromolybdenum-(VI)-Tetrahydrofuran (1). Under an inert atmosphere 1.52 g of freshly sublimed MoO₂Cl₂ (Strem, 7.64 mmol), 3.10 g of 2,6-diisopropylphenyl isocyanate (Aldrich, 15.27 mmol), and 20 mL of THF were introduced into a thick-walled 100-mL bottle with a screw cap. The closed bottle was then heated at 70 °C. The CO₂ gas formed in the reaction (after ca. 6 h) was allowed to escape by cooling the bottle to room temperature and opening the cap slightly. The cap was then reclosed and the bottle reheated. This process was continued, releasing the pressure each 12 h, until no further gas could be heard to escape upon slow opening of the bottle (2–3 days). Then the solvent was evaporated and the solid residue washed with pentane to give 95% of a red-brown powder. A sample for analysis was prepared by recrystallization from pentane. Anal. Calcd for MoC₂₄H₃₄N₂Cl₂·C₄H₈O: C, 57.05; H, 7.13; N, 4.75. Found: C, 56.45; H, 7.19; N, 4.62. IR (cm⁻¹): 3040 w, 1582 m, 1355 s, 1260 s, 1165 m, 1020 s, 860 vs, 800 m, 760 vs, 755 vs, 370 s. ¹H NMR (ppm, C₆D₆): 1.21 (d, 24 H, CH(CH₃)₂), 1.33 (m, 4 H, (CH₂–CH₂)₂O), 4.02 (sept, 4 H, CH(CH₃)₂), 4.11 (m, 4 H, (CH₂–CH₂)₂O), 6.83–7.01 (m, 6 H, aryl).

Bis((2,6-diisopropylphenyl)imido)dineopentyl-molybdenum(VI) (2). A 630-mg amount of dichloro derivative 1 (1.07 mmol) and 170 mg of LiNp (2.18 mmol) were stirred in pentane for 12 h. Filtration of liberated LiCl and evaporation gave a orange powder. Yield: 95%. Anal. Calcd for MoC₃₄H₅₆N₂: C, 69.39; H, 9.52; N, 4.76. Found: C, 69.25; H, 9.41; N, 5.02. IR (cm⁻¹): 3050 w, 1420 w, 1355 m, 1320 m, 1275 s, 1230 m, 1095 m, 980 m, 798 m, 758 s, 752 s. ¹H NMR (ppm, C₆D₆): 1.15 (d, 24 H, CH(CH₃)₂), 1.27 (s, 18 H, CH₂C(CH₃)₃), 2.29 (s, 4 H,

CH_2C(CH$_3$)$_3$), 3.74 (sept, 4 H, CH(CH$_3$)$_2$), 6.96 (m, 6 H, aryl).

Bis((2,6-diisopropylphenyl)imido)dimesitylmolybdenum-(VI) (3). To 300 mg of 1 (0.51 mmol) in diethyl ether was added 1.30 g of a 1 M solution of MesMgBr in THF (Aldrich, 1.30 mmol), and the mixture was stirred for 48 h. Filtration and evaporation gave a burgandy red solid, which was recrystallized with loss from pentane at –40 °C. Yield: 30%. Anal. Calcd for MoC$_{42}$H$_{56}$N$_2$: C, 73.68; H, 8.19; N, 4.09. Found: C, 73.53; H, 8.47; N, 4.05. IR (cm^{-1}): 1595 m, 1320 m, 1280 s, 1268 s, 1105 m, 979 s, 850 s, 798 s, 750 s, 585 w, 383 w, 350 w. ^{1}H NMR (ppm, C$_6$D$_6$): 0.96 (d, 24 H, CH(CH_3)$_2$), 2.08 (s, 6 H, *p*-CH$_3$(mesityl)), 2.88 (s, 12 H, *o*-CH$_3$(mesityl)), 3.90 (sept, 4 H, CH(CH$_3$)$_2$), 6.79 (s, 4 H, mesityl aryl), 6.96 (s, 6 H, aryl).

Bis((2,6-diisopropylphenyl)imido)bromomesitylmolybdenum(VI) (4). To 300 mg of the dichloro derivative 1 (0.52 mmol) in diethyl ether was added 650 mg of a 1 M solution of MesMgBr in THF (0.65 mmol), and the mixture was stirred for 12 h. Filtration and evaporation gave a bright red solid, which was recrystallized with loss from pentane at –40 °C. Yield: 25%. Anal. Calcd for MoC$_{33}$H$_{45}$N$_2$Br: C, 61.40; H, 7.03; N, 4.34. Found: C, 61.85; H, 7.41; N, 4.26. IR (cm^{-1}): 1600 m, 1370 m, 1330 w, 1290 w, 1280 m, 1265 m, 988 m, 975 m, 860 s, 808 s, 770 s, 765 s, 385 m, 365 m. ^{1}H NMR (ppm, C$_6$D$_6$): 1.08 (d, 24 H, CH(CH_3)$_2$), 2.00 (s, 3 H, *p*-CH$_3$(mesityl)), 2.75 (s, 6 H, *o*-CH$_3$(mesityl)), 3.82 (sept, 4 H, CH(CH$_3$)$_2$), 6.66 (s, 2 H, mesityl aryl), 6.97 (s, 6 H, aryl). The mass spectrum, although not showing the molecular ion peak, clearly exhibits a fragmentation pattern indicating the presence of bromide and not chloride.

Bis((2,6-diisopropylphenyl)imido)chloro(diethylamido)-molybdenum(VI) (5) and Bis((2,6-diisopropylphenyl)imido)bis(diethylamido)molybdenum(VI) (6). A 1.50-g amount of dichloro derivative 1 (2.55 mmol) and 426 mg of lithium diethylamide (Aldrich, 5.39 mmol) were stirred in ether for 12 h. Filtration of liberated LiCl and evaporation gave a deep red oil, which was redissolved in pentane and cooled to –40 °C. The supernatant was decanted, leaving 50 mg (yield 5%) of a deep red solid of 5. Anal. Calcd for MoC$_{28}$H$_{44}$N$_3$Cl: C, 60.70; H, 7.95; N, 7.58. Found: C, 59.25; H, 7.85; N, 7.17. IR (cm^{-1}): 3040 w, 1575 w, 1410 s, 1350 m, 1315 s br, 1265 s br, 1240 m, 995 s, 970 m, 968 s, 928 m, 890 s, 788 s, 740 s br, 600 m, 582 m, 510 w, 420 w, 365 w. ^{1}H NMR (ppm, C$_6$D$_6$): 1.05 (t, 6 H, N(CH$_2$CH_3)$_2$), 1.23 (d, 24 H, CH(CH_3)$_2$), 3.27 (br q, 4 H, N(CH_2CH$_3$)$_2$), 4.07 (sept, 4 H, CH(CH$_3$)$_2$), 6.90–7.03 (m, 6 H, aryl).

Evaporation of the supernatant gave the bis(diethylamido) complex 6 as a deep red oil. Yield: 85%. Anal. Calcd for MoC$_{32}$H$_{54}$N$_4$: C, 64.86; H, 9.12. Found: C, 64.59; H, 9.26. IR (cm^{-1}): 3060 w, 1570 w, 1420 s, 1320 s br, 1265 s br, 1005 s, 973 m, 885 s, 788 s, 750 s, 600 w, 590 w. ^{1}H NMR (ppm, C$_6$D$_6$): 1.17 (t, 12 H, N(CH$_2$CH_3)$_2$), 1.18 (d, 24 H, CH(CH_3)$_2$), 3.84 (q, 8 H, N(CH_2CH$_3$)$_2$), 3.88 (sept, 4 H, CH(CH$_3$)$_2$), 6.85–7.10 (m, 6 H, aryl).

Bis((2,6-diisopropylphenyl)imido)bis((2,6-diisopropylphenyl)amido)molybdenum(VI) (7). A mixture of 371 mg of dichloro derivative 1 (0.97 mmol) and 365 mg of lithium (2,6-diisopropylphenyl)amide monoetherate (2.0 mmol) was stirred in pentane for 12 h. Filtration of liberated LiCl and evaporation gave a deep red powder that was recrystallized from pentane at –40 °C, from which crystals suited for an X-ray analysis were obtained. IR (cm^{-1}): 3323 w, 3040 w, 1580 w, 1350 m, 1315 m, 1265 m, 1252 m, 1235 m, 849 w, 835 w, 789 w, 742 s. ^{1}H NMR of crystals (ppm, C$_6$D$_6$): 1.02 (d, 24 H, CH(CH_3)$_2$), 1.25 (d, 24 H, CH(CH_3)$_2$), 3.40 (sept, 4 H, CH(CH$_3$)$_2$), 3.85 (sept, 4 H, CH(CH$_3$)$_2$), 6.93 (m, 6 H, aryl), 7.08 (m, 6 H, aryl), 8.04 (br s, 2 H, NH).

Crystal Structure Determination. Red crystals were obtained from pentane at –40 °C and were mounted at low temperature under an inert atmosphere. Data were collected on a Philips PW1100/16 diffractometer equipped with a low-temperature device using nickel-filtered Cu Kα radiation (λ = 1.5418 Å) and a θ/2θ flying step-scan method. No significant changes were observed for three standard reflections, which were monitored hourly during the data collection period. The Enraf-Nonius SDP package[8] was used for all calculations, except that a local data reduction program was employed. The initial step-scan data were converted to intensities by the method of Lehmann–Larson[9] and then corrected for Lorentz polarization and absorption factors, the latter computed by the method of Wacker and Stuart.[10] The structure was solved by using the heavy-atom method. Difference Fourier maps revealed maxima at positions expected for hydrogen atoms H'1 and H'2 attached to N3 and N4; these hydrogen atoms were introduced and refined by using a riding model. All other hydrogen atoms were introduced in structure factor calculations by their computed coordinates (C–H = 0.95 Å) and isotropic structure factors such that B(H) = 1 + B_{eqv}(C) Å^2 and were not refined. Final difference maps revealed no significant maxima.

Supplementary Material Available: Tables of bond lengths and angles, anisotropic thermal parameters, and hydrogen atom coordinates (8 pages); a table of observed and calculated structure factors (17 pages). Ordering information is given on any current masthead page.

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Polymeric Mercury Complexes with One-Dimensional Chain Structures. 2. Synthesis and Crystal Structure of [{Hg(CH$_2$P(O)Ph$_2$)$_2$}$_2$BrLi]$_n$

John P. Fackler, Jr.,* and Roman A. Kresinski

Department of Chemistry and Laboratory for Molecular Structure and Bonding, Texas A&M University, College Station, Texas 77843

Summary: An organomercury complex formulated as [{Hg(CH$_2$P(O)Ph$_2$)$_2$}$_2$Br·Li(THF)$_4$$^+$] (I) was obtained by the reaction of LiCH$_2$P(O)Ph$_2$ with HgBr$_2$ in THF. Slow evaporation of a solution of I afforded [{Hg(CH$_2$P(O)-Ph$_2$)$_2$}$_2$BrLi]$_n$ (II), which has a one-dimensional chain structure in the solid state.

There are a number of linear one-dimensional chain complexes of heavy metals such as Au or Hg. In the case of Au, these complexes are usually homometallic[1] and often

owe their chain structure to Au–Au interactions.[2] In the case of Hg(II), which exhibits little homophilicity, the chain structure is usually a result of bridging ligands. These ligands are commonly the conventional inorganic ligands[3] such as OH$^-$, O^{2-}, CN$^-$, or O$_2$$^{2-}$, but the use of some other

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