Four- *versus* Five-co-ordinate Bis(imido) Alkene Complexes of Molybdenum: the Contrasting Effects of *tert*-Butyl- and 2,6-Diisopropylphenyl-imido Substituents‡

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The five-co-ordinate complex $[Mo(NC_6H_3Pr^i_2\text{-}2,6)_2(PMe_3)_2(C_2H_4)]$ **2a** has been prepared *via* treatment of $[Mo(NC_6H_3Pr^i_2\text{-}2,6)_2Cl_2(dme)]$ **1a** (dme = 1,2-dimethoxyethane) with 2 equivalents of EtMgCl in Et_2O in the presence of excess PMe_3; its propene analogue **2b** is unstable at reduced pressure, converting to $[Mo(NC_6H_3Pr^i_2\text{-}2,6)_2(PMe_3)_2]$ **3** with loss of propene. Complex **2a** can also be synthesised, and **2b** generated *in situ*, by treatment of **3** with ethene and propene respectively in hydrocarbon solvent; the analogous η^2-butadiene complex $[Mo(NC_6H_3Pr^i_2\text{-}2,6)_2(PMe_3)_2(\eta^2\text{-}C_4H_6)]$ **2c** has also been generated in solution and identified by NMR spectroscopy. The molecular structure of **2a** shows a distorted trigonal-bipyramidal co-ordination geometry with axial phosphines and equatorial imido ligands: the ethene ligand aligns in the P–Mo–P plane and perpendicular to the N–Mo–N plane. Complex **3** can also be prepared directly through the reaction of **1a** with magnesium turnings in tetrahydrofuran (thf) in the presence of excess PMe_3. By contrast, the reaction of $[Mo(NBu^t)_2Cl_2(dme)]$ **1b** under identical conditions affords the binuclear imido-bridged complex $[\{Mo(NBu^t)(\mu\text{-}NBu^t)(PMe_3)\}_2]$ **4**. Complex **4** does not react with ethene or propene to give the known mononuclear bis(*tert*-butylimido) derivatives, $[Mo(NBu^t)_2(PMe_3)(C_2H_3R)]$ (R = H **5a** or Me **5b**) reflecting the robust nature of the imido bridges.

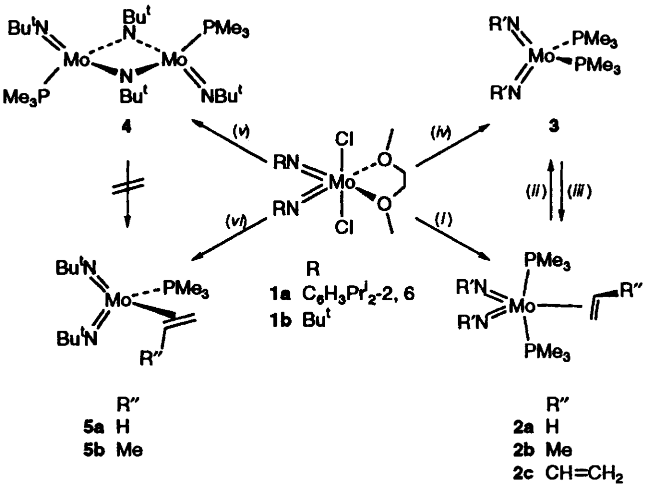
Four-co-ordinate bis(imido) complexes of the Group 6 metals are of interest due to their 'isolobal' relationship to bent metallocenes of the Group 4 metals which find widespread applications as reagents in organic synthesis[1] and as polymerisation catalysts.[2] The relationship can be conveniently probed through derivatives containing 'single-faced' π ligands since such groups adopt a strong orientation preference with respect to other π-donor ligands.[3]

In their studies on $[W(NR)_2]$ complexes containing 2,6-diisopropylphenylimido ligands, Schrock and co-workers[4] found that five-co-ordinate species of the type $[W(NC_6H_3Pr^i_2\text{-}2,6)_2(PMe_2Ph)_2(L)]$ (L = C_2H_4 or C_2H_2) are obtained when L is a relatively small ligand such as ethene or ethyne. Subsequently, we found that a four-co-ordinate manifold can be maintained for molybdenum with small alkenes as co-ligands if the imido groups bear *tert*-butyl substituents, *e.g.* $[Mo(NBu^t)_2(PMe_3)(C_2H_3R)]$ (R = H or Me).[5] We reasoned that these differences arise from the greater steric flexibility of the aryl substituents which can orientate so as to maximise the space available to the other attendant groups. Here, we compare and contrast the properties of a number of simple alkene and phosphine derivatives of molybdenum, including a crystal structure of a five-co-ordinate derivative which offers an opportunity to assess the steric requirements of the $Mo(NC_6H_3Pr^i_2\text{-}2,6)_2$ fragment in comparison to its bis(*tert*-butylimido) counterpart.

Results and Discussion

The chemistry is summarised in Scheme 1. Treatment of $[Mo(NC_6H_3Pr^i_2\text{-}2,6)_2Cl_2(dme)]$ (dme = 1,2-dimethoxyethane) **1a** with 2 equivalents of ethylmagnesium chloride in diethyl ether in the presence of excess trimethylphosphine affords an intense maroon coloured solution, which after work-up and extraction with *n*-pentane gives purple-red needle crystals in moderate yield. Elemental analyses and NMR data are consistent with the five-co-ordinate complex $[Mo(NC_6H_3Pr^i_2\text{-}2,6)_2(PMe_3)_2(C_2H_4)]$ **2a**. Proton and ^{13}C NMR spectra show that the ends of the ethylene ligand and the two phosphines are equivalent down to −80 °C.

A similar reaction ensues with *n*-propylmagnesium chloride, affording a purple-red solution. However, upon removal of the solvent under reduced pressure, the solution changes to dark green and crystals of the four-co-ordinate bis(phosphine)



Scheme 1 *Reagents and conditions:* (*i*) $2PMe_3$, 2RMgCl (R = Et or Pr^n), Et_2O, room temperature (r.t.), 3 h; (*ii*) for R′ = Me only, vacuum, pentane; (*iii*) $R'CH{=}CH_2$, C_6D_6, r.t., 30 min; (*iv*) Mg, $2PMe_3$, thf, r.t., 3 h; (*v*) Mg, $2PMe_3$, thf, r.t., 3 h; (*vi*) see ref. 5. R′ = $C_6H_3Pr^i_2$-2,6

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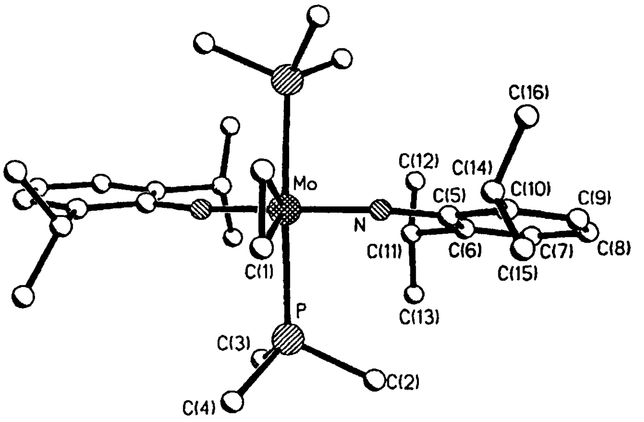

Fig. 1 Molecular structure of **2a**, without H atoms and with key atoms labelled

Table 1 Atomic coordinates (× 10^4) for complex **2a** with estimated standard deviations (e.s.d.s) in parentheses

Atom	*x*	*y*	*z*
Mo	5000	6907.1(4)	7500
C(1)	5222(2)	9032(4)	7837(3)
P	5820.0(5)	6561.7(10)	8725.8(6)
C(2)	6679(2)	6462(5)	8581(3)
C(3)	5693(2)	5019(5)	9248(3)
C(4)	5868(3)	7826(6)	9494(3)
N	5645.3(13)	6320(3)	6959(2)
C(5)	6148(2)	5761(3)	6630(2)
C(6)	6307(2)	4350(3)	6700(2)
C(7)	6842(2)	3879(4)	6372(2)
C(8)	7219(2)	4711(4)	5981(3)
C(9)	7061(2)	6079(4)	5906(2)
C(10)	6529(2)	6621(3)	6216(2)
C(11)	5913(2)	3374(3)	7123(2)
C(12)	5535(2)	2347(4)	6570(3)
C(13)	6339(2)	2610(4)	7780(3)
C(14)	6356(2)	8109(4)	6113(2)
C(15)	6887(3)	9014(6)	6437(6)
C(16)	6140(5)	8466(7)	5281(4)

Table 2 Bond lengths (Å) and angles (°) for complex **2a** with e.s.d.s in parentheses *

Mo–N	1.829(3)	Mo–C(1)	2.207(4)
Mo–P	2.5374(13)	C(1)–C(1a)	1.375(10)
P–C(3)	1.812(4)	P–C(2)	1.818(4)
P–C(4)	1.821(5)	N–C(5)	1.367(4)
C(5)–C(10)	1.421(5)	C(5)–C(6)	1.431(5)
C(6)–C(7)	1.390(5)	C(6)–C(11)	1.517(5)
C(7)–C(8)	1.375(5)	C(8)–C(9)	1.390(5)
C(9)–C(10)	1.392(5)	C(10)–C(14)	1.516(5)
C(11)–C(12)	1.528(5)	C(11)–C(13)	1.531(6)
C(14)–C(15)	1.457(7)	C(14)–C(16)	1.492(8)
N(a)–Mo–N	143.1(2)	N–Mo–C(1)	107.6(2)
N–Mo–C(la)	107.5(2)	C(1)–Mo–C(1a)	36.3(3)
N(a)–Mo–P	87.92(10)	N–Mo–P	87.20(10)
C(1)–Mo–P	79.57(13)	C(1a)–Mo–P	115.87(13)
P–Mo–P(1a)	164.56(5)	C(1a)–C(1)–Mo	71.85(13)
C(3)–P–C(2)	103.8(2)	C(3)–P–C(4)	101.7(3)
C(2)–P–C(4)	101.0(3)	C(3)–P–Mo	114.1(2)
C(2)–P–Mo	115.2(2)	C(4)–P–Mo	118.9(2)
C(5)–N–Mo	172.5(3)	N–C(5)–C(10)	118.5(3)
N–C(5)–C(6)	122.2(3)	C(10)–C(5)–C(6)	119.2(3)
C(7)–C(6)–C(5)	118.3(3)	C(7)–C(6)–C(11)	119.8(3)
C(5)–C(6)–C(11)	121.9(3)	C(8)–C(7)–C(6)	122.7(4)
C(7)–C(8)–C(9)	119.0(3)	C(8)–C(9)–C(10)	121.5(4)
C(9)–C(10)–C(5)	119.2(3)	C(9)–C(10)–C(14)	120.7(3)
C(5)–C(10)–C(14)	120.1(3)	C(6)–C(11)–C(12)	111.8(3)
C(6)–C(11)–C(13)	113.0(3)	C(12)–C(11)–C(13)	109.0(3)
C(15)–C(14)–C(16)	109.2(6)	C(15)–C(14)–C(10)	113.5(4)
C(16)–C(14)–C(10)	112.0(4)		

* Symmetry operator: a $-x + 1, y, -z + \frac{3}{2}$.

complex $[Mo(NC_6H_3Pr^i_2\text{-}2,6)_2(PMe_3)_2]$ **3** are isolated in good yield. The propene ligand of **2b** is evidently highly labile, thwarting its isolation, but it can be generated cleanly *in situ* by treating **3** with excess propene in a sealed NMR tube. The rate of propene exchange was determined to be 1.34 s^{-1} by magnetisation transfer experiments between the methyl resonances of the bound and free propene.

The bis(phosphine) complex **3** can also be prepared independently by treatment of **1a** with 1 equivalent of magnesium in thf in the presence of excess PMe_3. By contrast, the reduction of **1b** under similar conditions affords a species whose elemental analysis is indicative of a 2:1 stoichiometry of imido to phosphine ligands per molybdenum centre, *i.e.* $[Mo(NBu^t)_2(PMe_3)]$. Moreover, ^1H and ^{13}C NMR spectra show that the imido ligands are inequivalent suggesting a binuclear structure (**4**) with terminal and bridging imido units; attempts to obtain crystals suitable for an X-ray diffraction study have so far proved unsuccessful. Complex **4** does not react with ethene or propene to give the known complexes $[Mo(NBu^t)_2(PMe_3)(C_2H_3R)]$ (R = H **5a** or Me **5b**),[5] presumably reflecting the robust nature of the imido bridges.

Molecular Structure of $[Mo(NC_6H_3Pr^i_2\text{-}2,6)_2(PMe_3)_2(C_2H_4)]$ **2a**.—Crystals of **2a** suitable for a molecular structure determination were grown from a saturated *n*-pentane solution at −30 °C. The structure is shown in Fig. 1; atomic parameters and bond lengths and angles are collected in Tables 1 and 2.

The molecular geometry is best described as distorted trigonal bipyramidal with the PMe_3 ligands occupying axial sites and the imido ligands in equatorial positions. A crystallographic C_2 axis bisects the N–Mo–N and P–Mo–P angles and passes through the midpoint of the ethene C=C bond. If the imido ligands are regarded as four-electron donor groups, then **2a** is a 20-electron species and this is expected to have important structural consequences. The N–Mo–N angle of 143.1(2)° is large in comparison with other bis(imido) molybdenum complexes and is approaching the N–Os–N angle in $[Os(NC_6H_3Pr^i_2\text{-}2,6)_2(PMe_2Ph)I_2]$ [151.2(3)°].[6] This has been rationalised in general terms by Schrock and co-workers[6] as a compromise inter-ligand bond angle arising from a need to remove undesirable π-antibonding interactions at the 120 and 180° extremes. While the Mo=N–C (aryl) bond angles are close to linear [172.5(3)°], the Mo–N bond lengths are relatively long [1.829(3)°] in comparison with other bis(arylimido) molybdenum compounds {*e.g.* the arylimido ligand in $[Mo(NBu^t)(NC_6H_3Pr^i_2\text{-}2,6)Cl_2(dme)]$ 1.753(2) Å,[7] $[Mo(NC_6H_3Pr^i_2\text{-}2,6)_2(PMe_3)_2]$ 1.801(13) Å (av.)[8]}, reflecting the unfavourable competition between the two imido ligands for metal d_π symmetry orbitals in this geometry. The P–Mo–P angle of 164.56(5)° arises from bending of the phosphine ligands away from the imido groups. The phenyl rings of the imido ligands are almost co-planar, an unusual orientation for bis(arylimido) metal complexes wherein one of the aryl substituents usually aligns perpendicular to the N–Mo–N plane in order to maximise π overlap between the metal and imido nitrogens and the imido nitrogens and *ipso*-ring carbons. The approximate co-planar orientation observed here is, however, likely to be influenced by the steric interactions between the two PMe_3 groups and the imido substituents.

The ethene ligand occupies an equatorial site with the carbon–carbon bond lying perpendicular to the trigonal plane. The C–C distance of 1.375(10) Å is short compared with the four-co-ordinate complex $[Mo(NBu^t)_2(PMe_3)(C_2H_6)]$ [1.431(7) Å][5] implying relatively little π-back donation from the d^2 centre of **2a**.

In summary, the *tert*-butyl and 2,6-diisopropylphenyl substituents clearly exert dramatically differing influences on the co-ordination number and stability of bis(imido) complexes of molybdenum. The orientational flexibility of the aryl substituent is undoubtedly a key factor, allowing it to accommodate expanded co-ordination numbers compared with its *tert*-butyl counterpart.

Experimental

General.—All manipulations were carried out under an atmosphere of nitrogen using standard Schlenk and cannula techniques or in a conventional nitrogen-filled glove-box. Solvents were refluxed over an appropriate drying agent, and distilled and degassed prior to use. Elemental analyses were performed by the microanalytical services of the Department of Chemistry at Durham. NMR spectra were recorded on a Varian VXR 400 S spectrometer at 400.0 MHz (^{1}H), 100.6 MHz (^{13}C) and 161.9 MHz (^{31}P); chemical shifts are referenced to the residual protio impurity of the deuteriated solvent; IR spectra (Nujol mulls, CsI windows), Perkin-Elmer 577 and 457 grating spectrophotometers; mass spectra, VG 7070E [70 eV (*ca.* 1.12 × 10^{-17} J), 100 μA emission. Trimethylphosphine was prepared by a previously published procedure;[9] [$Mo(NC_6H_3Pr^i{}_2$-2,6)$_2Cl_2$(dme)] **1a** was synthesised *via* a modification of the procedure described by Schrock and co-workers.[10] Anhydrous Na_2MoO_4 was purchased from Aldrich Chemical Co. and used as received. All other chemicals were obtained commercially and used as received unless stated otherwise.

Syntheses.—[$Mo(NC_6H_3Pr^i{}_2$-2,6)$_2Cl_2$(dme)] **1a**. Solutions of triethylamine (27.1 cm^3, 194.4 mmol), chlorotrimethylsilane (68 cm^3, 534.6 mmol) and 2,6-diisopropylaniline (18 cm^3, 97.2 mmol) in 1,2-dimethoxyethane (dme) (*ca.* 20 cm^3 each solution) were added sequentially to a stirred suspension of anhydrous Na_2MoO_4 (10 g, 48.6 mmol) in dme (100 cm^3) at room temperature. The reaction mixture was then heated at 70 °C for 12 h to afford a dark red solution and a large quantity of white precipitate. The solution was filtered from the solid which was then washed with diethyl ether (3 × 50 cm^3). The solvent was then removed from the combined filtrate and washings under reduced pressure to afford analytically pure, red crystalline [$Mo(NC_6H_3Pr^i{}_2$-2,6)$_2Cl_2$(dme)]. Yield 28.9 g (98%) (Found: C, 55.0; H, 7.2; N, 4.5. Calc. for $C_{28}H_{44}Cl_2MoN_2O_2$: C, 55.3; H, 7.3; N, 4.6%).

[$Mo(NBu^t)_2Cl_2$(dme)] **1b**. The procedure for **1b** has been described elsewhere.[5]

[$Mo(NC_6H_3Pr^i{}_2$-2,6)$_2(PMe_3)_2(C_2H_4)$] **2a**. Trimethylphosphine (0.7 cm^3, 7 mmol) was condensed onto a frozen (−196 °C) mixture of **1a** (1.5 g, 2.5 mmol) and diethyl ether (40 cm^3) in a glass ampoule fitted with a Teflon stopcock. After warming the mixture to −78 °C (solid CO_2–acetone slush bath), a nitrogen atmosphere was introduced and EtMgCl (2.5 cm^3, 2 mol dm^{-3} solution in Et_2O) was added by syringe. The reaction mixture was then allowed to warm to room temperature with stirring and occasional venting. After 3 h at room temperature, the volatile components were removed under reduced pressure. Extraction of the residue with pentane (30 cm^3) afforded a dark purple solution which was concentrated and cooled to −30 °C to give dark red crystals of **2a**. Yield 0.93 g (60%) (Found: C, 61.1; H, 4.3; N, 9.4. Calc. for $C_{32}H_{56}MoN_2P_2$: C, 61.3; H, 4.5; N, 9.0%).

Attempted preparation of [$Mo(NC_6H_3Pr^i{}_2$-2,6)$_2(PMe_3)_2(C_2H_6)$] **2b**. Attempts to prepare **2b** were carried out by an analogous procedure to that described above for **2a**, using Pr^nMgCl. Quantities of reagents employed: trimethylphosphine (0.7 cm^3, 7 mmol), **1a** (1.5 g, 2.5 mmol), diethyl ether (40 cm^3), Pr^nMgCl (2.5 cm^3, 2 mol dm^{-3} solution in Et_2O). After stirring for 3 h at room temperature, the reaction mixture was red-purple. However, on removal of the solvent at reduced pressure, the solution turned green and [$Mo(NC_6H_3Pr^i{}_2$-2,6)$_2(PMe_3)_2$] was isolated.

[$Mo(NC_6H_3Pr^i{}_2$-2,6)$_2(PMe_3)_2$] **3**. Trimethylphosphine (0.51 cm^3, 4.92 mmol) was condensed onto a frozen (−196 °C) mixture of **1a** (0.5 g, 0.82 mmol), activated magnesium turnings (0.02 g, 0.90 mmol) and thf (50 cm^3) in a glass ampoule fitted with a Teflon stopcock. After warming the mixture to room temperature, a nitrogen atmosphere was introduced and the mixture was stirred for 3 h. During this time, the mixture changed from dark brown to dark green. The volatile components were then removed under reduced pressure and the dark green residue was extracted with pentane to give a dark green solution which, upon concentration and cooling to −78 °C gave green crystalline **3**. Yield 0.49 g (60%) (Found: C, 60.0; H, 8.7; N, 4.4. Calc. for $C_{30}H_{52}MoN_2P_2$: C, 60.2; H, 8.8; N, 4.7%).

[{$Mo(NBu^t)(\mu$-$NBu^t)(PMe_3)$}$_2$] **4**. Trimethylphosphine (1.0 cm^3, 10 mmol) was condensed onto a frozen (−196 °C) mixture of **1b** (2.0 g, 5.0 mmol), activated magnesium turnings (0.15 g, 5.5 mmol) and thf (50 cm^3) in a glass ampoule fitted with a Teflon stopcock. After warming the mixture to room temperature, a nitrogen atmosphere was introduced and the mixture was stirred for 3 h. During this time, the mixture changed colour from red-brown to dark blue-green. The volatile components were then removed under reduced pressure and the residue was extracted with pentane to give a dark blue-green solution which, upon concentration and cooling to −78 °C afforded dark blue crystalline **4**. Yield 1.89 g (68%) (Found: C, 42.1; H, 8.9; N, 8.9. Calc. for $C_{11}H_{27}MoN_2P$: C, 42.0; H, 8.7; N, 8.9%).

X-Ray Crystallography.—*Crystal data for* **2a**: $C_{32}H_{56}MoN_2P_2$, M = 626.7, monoclinic, space group $C2/c$, a = 20.508(4), b = 9.867(2), c = 17.417(3) Å, β = 98.52(2)°, U = 3485.5(11) Å^3, Z = 4, D_c = 1.194 g cm^{-3}, μ(Mo-Kα) = 0.489 mm^{-1}, λ = 0.710 73 Å, F(000) = 1336, T = 240 K. Measurements were made on a Stoe-Siemens diffractometer with on-line profile fitting for intensities,[11] from a crystal of size 0.46 × 0.18 × 0.08 mm. Of 4599 measured data, 3074 were unique (R_{int} = 0.0258, $2\theta_{max}$ = 50°); semiempirical absorption corrections were applied (transmission factors 0.859–0.921). There was no significant variation for three periodically measured standard reflections.

The structure was solved by direct methods and refined by least-squares techniques on F^2 values for all observed data and a weighting scheme $w^{-1} = \sigma^2(F_o{}^2) + (0.03P)^2 + 7.2147P$, where $P = (F_o{}^2 + 2F_c{}^2)/3$. All non-hydrogen atoms were refined anisotropically, and isotropic H atoms were refined with a riding model except for the freely refined positions of the ethene H atoms. All features in a final difference map were within ±0.47 e Å^{-3}. Final $R' = \{\Sigma[w(F_o{}^2 - F_c{}^2)^2]/\Sigma[w(F_o{}^2)^2]\}^{\frac{1}{2}}$ = 0.0980 for all data, conventional R = 0.0359 on F values of 2267 reflections with $F_o{}^2 > 2\sigma(F_o{}^2)$; goodness of fit = 1.093 on F^2 for 181 refined parameters. Programs: SHELXTL.[12]

Additional material available from the Cambridge Crystallographic Data Centre comprises H-atom coordinates and thermal parameters.

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