The Chemistry of Transition-metal Nitrosyl Compounds. Part VII.[1] The Preparation and Study of Some Cationic Nitrosyl Complexes of Iron

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The cationic iron nitrosyl complexes $[Fe(CO)_2NOL_2]PF_6$ [L = $P(OPh)_3$, $P(OMe)_3$, PPh_3, $PMePh_2$, PMe_2Ph, PEt_3, $P(C_6H_{11})_3$, $AsPh_3$ and $\frac{1}{2}$dppe] have been prepared and some of their properties studied. Certain of these cations [L = $P(OMe)_3$, $PMePh_2$, and PMe_2Ph] react with additional ligand (L) to produce the cationic species $[Fe(CO)(NO)L_3]^+$. With dppe the complex $[Fe(CO)_2NO(dppe)]PF_6$ yields the mononitrosyl cation $[Fe(NO)(dppe)_2]^+$; this cation has also been prepared from the reaction of the hydride $FeHCl(dppe)_2$ with *N*-methyl-*N*-nitrosotoluene-*p*-sulphonamide in the presence of $NaBPh_4$. The cations $[Fe(CO)_2NOL_2]^+$ react with OMe^- to produce the neutral carboxy-compounds $Fe(CO)(NO)(L)_2CO_2Me$ (L = PPh_3 or $\frac{1}{2}$dppe). No evidence for nucleophilic addition of OMe^- to the co-ordinated NO group in any of the cationic species has been found.

This work was initiated as part of a general programme of research directed towards the activation of NO by co-ordination. It was our intention to produce a series of cationic nitrosyl complexes in the hope that the presence of a *formal* positive charge would enhance the reactivity of the co-ordinated NO group towards nucleophiles in the manner previously observed with certain other NO species.[2] In the course of this investigation a convenient route to such compounds, which involves the reaction of the nitrosonium salt, $NOPF_6$, with metal carbonyl derivatives, has been employed. Nitrosonium salts have been used previously in the production of cationic NO derivatives but to a limited extent (see ref. 3). In this paper we report the preparation of a series of cationic complexes $[Fe(CO)_2NOL_2]PF_6$ {L = $P(OMe)_3$, $P(OPh)_3$, PPh_3, $PMePh_2$, PMe_2Ph, PEt_3, $\frac{1}{2}[Ph_2P(CH_2)_2PPh_2]$ (dppe), or $AsPh_3$} by this route and also a study of their reactions.

RESULTS AND DISCUSSION

On treatment of $Fe(CO)_3L_2$ with $NOPF_6$ in dry benzene–methanol carbon monoxide is evolved and the solution changes from yellow to orange. Addition of diethyl ether to this solution causes the precipitation of the product $[Fe(CO)_2NOL_2]PF_6$ which after recrystallisation from tetrahydrofuran–ether, is obtained as a yellow-orange crystalline material. This method is preferred to that reported previously in Part II of this series[4] for the formation of $[Fe(CO)_2NO(PPh_3)_2]^+$ from $Fe(CO)_3(PPh_3)_2$ and NOCl because of the high yields and the purity of product obtained. The triphenylphosphine cation, $[Fe(CO)_2NO(PPh_3)_2]^+$, has also been obtained by the addition of $Fe(CO)_3(PPh_3)_2$ to a solution of isopentyl nitrite in acidified (HPF_6) benzene–methanol presumably *via* the reaction sequence:

$$RONO + HPF_6 \rightleftharpoons ROH + NO^+ + PF_6^-$$

$$(R = \text{isopentyl})$$

$$Fe(CO)_3(PPh_3)_2 + NO^+ \longrightarrow [Fe(CO)_2NO(PPh_3)_2]^+ + CO$$

In contrast, the corresponding reaction of $NOPF_6$ in methanol–benzene with either $Ru(CO)_3(PPh_3)_2$ or $Os(CO)_3(PPh_3)_2$ leads to the hydrido-cations $[HRu(CO)_3(PPh_3)_2]^+$ and $[HOs(CO)_3(PPh_3)_2]^+$ respectively, reflecting the relative basicities of the three $M(CO)_3(PPh_3)_2$ species.[5] Satisfactory analytical data have been obtained for all the salts (Table 1) and they are 1 : 1 electrolytes in nitrobenzene. Their stability in air varies considerably ranging from a few hours with the PMe_2Ph derivative to several days with the PPh_3 complex. In the i.r. spectra (Table 2) the cations exhibit two CO stretching vibrations and one NO stretching vibration in all cases; evidence consistent with the trigonal bipyramidal structure (I)† proposed previously.[4] The ^1H n.m.r. spectra of $[Fe(CO)_2NO(PMePh_2)_2]PF_6$ and $[Fe(CO)_2NO(PMe_2Ph)_2]PF_6$ in CD_2Cl_2 exhibit apparent methyl doublets at τ 7·52 and at 7·81 with $|J_{PH} + J_{PH'}|$ of 8·4 and 10·1 Hz respectively. The fact that both compounds show similar spectra indicates that the two phosphine ligands are in equivalent environments in the molecule in agreement with the proposed structure (I). Further structural assignments cannot be made on the basis of these n.m.r. spectra since the Jenkins–Shaw criteria may not be applicable to this system.

By employing the bidentate phosphine, 1,2-bisdiphenylphosphinoethane, it has been possible to generate the ion $[Fe(CO)_2NO(dppe)]^+$ which must contain the two phosphorus atoms bonded to the metal in a *cis*-arrangement thereby ruling out structure (I). The general profile of the i.r. spectrum of this ion (1800—2100 cm^{-1}) resembles that observed for the other $[Fe(CO)_2NOL_2]^+$ species except that both CO bands are very strong. This change in the relative intensity of the two CO bands suggests a significant decrease in the OC–Fe–CO bond angle and is indicative of a change from an *essentially* trigonal bipyramidal structure (I) towards a square

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† A recent *X*-ray crystal-structure determination of the corresponding osmium complex $[Os(CO)_2NO(PPh_3)_2]ClO_4,CH_2Cl_2$ reveals trigonal bipyramidal geometry with a linear NO group and *trans*-PPh_3 ligands, G. R. Clark, K. R. Grundy, W. R. Roper, J. M. Waters, and K. R. Whittle, *J.C.S. Chem. Comm.*, 1972, 119.

[1] Part VI, R. Davis, B. F. G. Johnson, and K. H. Al-Obaidi, *J.C.S. Dalton*, 1972, 508.

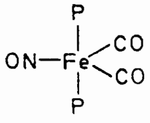
[2] J. H. Swinehart, *Co-ordination. Chem. Rev.*, 1967, **2**, 385, and for example, T. J. Meyer, J. B. Godwin, and N. Winterton, *Chem. Comm.*, 1970, 872.

[3] E. O. Fischer and H. Strametz, *Z. Naturforsch.*, 1968, **23**b, 278; D. J. Hodgson, N. C. Payne, J. A. McGinnity, R. G. Pearson, and J. A. Ibers, *J. Amer. Chem. Soc.*, 1968, **90**, 4486; C. G. Pierpont, D. G. Van Derveer, W. Durland, and R. Eisenberg, *ibid.*, 1970, **92**, 4761; N. G. Connelly and L. F. Dahl, *Chem. Comm.*, 1970, 880.

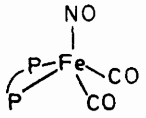
[4] Part II, G. R. Crooks and B. F. G. Johnson, *J. Chem. Soc.* (*A*), 1968, 1238.

[5] B. F. Johnson and J. A. Segal, *J. Organometallic Chem.*, 1971, **31**, C79.

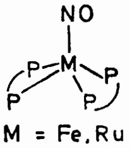
based pyramid, structure (II). No evidence for the formation of other isomeric forms of the ions



(I)



(II)



(III)

$[Fe(CO)_2NOL_2]^+$ has been found. The appearance of the NO band at 1800 cm^{-1} indicates that the NO group is functioning as a (*formal*) three electron-donor (NO^+) and that the Fe–N–O arrangement is linear. Thus in this case, and in contrast to the behaviour exhibited by certain five-co-ordinate cobalt nitrosyl complexes $[Co(NO)X_2(PPh_3)_2]$, there is no evidence that a change in stereochemistry induces a change in the mode of co-ordination of the NO ligand [formally NO^+ (linear) $\rightleftharpoons$ NO^- (bent)].[6]

Addition of free phosphine to solutions of the cations in CD_2Cl_2 causes no immediate change in the ^1H n.m.r. spectra (see above) and no exchange between the co-ordinated and the free phosphine is apparent. Over more extended periods, however, conversion to new cations identified as $[Fe(CO)(NO)L_3]^+$ is observed. These cations [$L = PMe_2Ph$, $PMePh_2$, or $P(OMe)_3$] were conveniently prepared by reaction of the appropriate phosphine with the corresponding $[Fe(CO)_2NOL_2]^+$ species in dichloromethane. Complexes containing bulky phosphines [*e.g.* PCy_3, PPh_3, $P(OPh)_3$, or $AsPh_3$] did not undergo further substitution presumably because of the steric interactions that would be introduced. In the case of triethylphosphine reaction did occur but did not give the expected cationic derivative; instead products such as $Fe(NO)_2(PEt_3)_2$ and $Fe(CO)_3(PEt_3)_2$ were obtained. The orange crystalline salts $[Fe(CO)(NO)L_3]PF_6$ were characterised by their i.r. spectra (Table 2), and analytical data (Table 1). Conductance values in nitrobenzene are in the range expected for 1 : 1 electrolytes (Table 1). Unexpectedly treatment of $Fe(CO)_2[P(OMe)_3]_3$ with $NOPF_6$ gave the cation $[Fe(CO)_2(NO)\{P(OMe)_3\}_2]^+$ and not the monocarbonyl species $[Fe(CO)(NO)\{P(OMe)_3\}_3]^+$.

Further treatment of the cations $[Fe(CO)(NO)L_3]^+$ with an excess of phosphine did not produce the mono-nitrosyl derivatives $[Fe(NO)L_4]^+$, again presumably because of the large steric interactions anticipated for such a species. However reaction of $[Fe(CO)_2NO(dppe)]^+$ with an excess of dppe in dichloromethane gave the cation $[Fe(NO)(dppe)_2]^+$ in excellent yield. In very concentrated dichloromethane solutions an additional product $\{[Fe(CO)(NO)(dppe)]_2dppe\}(PF_6)_2$ was also isolated. This is considered to involve a dppe-bridged system of the type previously observed by Basolo *et al.*[7] in the neutral species $[Fe(CO)(NO)_2]_2dppe$. The cation $[Fe(NO)(dppe)_2]^+$ has also been obtained as its tetraphenylborate salt from an alternative reaction route starting from $FeHCl(dppe)_2$[8] (see Scheme):

$$FeHCl(dppe)_2 \xrightarrow[\text{EtOH}]{\text{MNTS + NaBPH}_4} [FeNO(dppe)_2]BPh_4$$

It exhibits a strong N–O stretching vibration at 1697 cm^{-1} in its i.r. spectrum which is close to the value observed for the analogous ruthenium cation (1673 cm^{-1}). We therefore suggest that the ion $[FeNO(dppe)_2]^+$ is isostructural with this similar ruthenium species which is known to possess a distorted square-based pyramidal structure (III) and an essentially linear Ru–N–O bonding system.[9]

TABLE 1

Analytical and conductivity data for the complexes

Compound	L	Λ_M [a]	Found (%)				Required (%)			
			C	H	N	P	C	H	N	P
$[Fe(CO)_2NOL_2]PF_6$	$P(OPh)_3$	28·0	50·4	3·3	1·6	10·0	50·4	3·3	1·5	10·2
	$P(OMe)_3$	30·7	18·2	3·5	2·5	17·1	18·0	3·4	2·6	17·4
	PPh_3	25·7	56·4	3·8	1·7	11·4	56·2	3·7	1·7	11·5
	$PMePh_2$	27·6	49·2	4·1	2·1	13·3	48·9	3·8	2·0	13·5
	PMe_2Ph	29·4	38·3	4·0	2·4	16·2	38·4	4·0	2·5	16·5
	PEt_2	30·7	32·3	5·6	2·6	17·6	32·1	5·8	2·7	17·8
	PCy_3	26·0	54·1	7·8	1·7	10·7	53·8	7·9	1·7	11·0
	dppe/2 [b]	26·6	49·3	3·8	2·0	13·5	49·1	3·5	2·0	13·6
	$AsPh_3$	25·9	50·6	3·7	1·7	3·7	50·8	3·4	1·6	3·4
$[Fe(CO)NOL_3]PF_6$	$P(OMe)_3$	28·6	19·3	4·4	2·5	19·6	19·0	4·3	2·2	19·6
	$PMePh_2$	25·9	55·9	4·8	1·4	14·3	55·9	4·6	1·6	14·4
	PMe_2Ph	26·4	44·5	4·8	2·1	18·1	44·6	4·9	2·1	18·4
$[FeNO(dppe)_2]PF_6$ [c]		24·8	60·6	4·7	1·5	15·1	60·8	4·7	1·4	15·1
$\{[Fe(CO)NOdppe]_2dppe\}(PF_6)_2$		42·2	55·9	4·4	1·5	14·2	56·1	4·2	1·6	14·5
$[FeNO(dppe)_2]BPh_4$		15·5	75·7	5·7	1·3	10·2	76·0	5·7	1·2	10·3
$Fe(CO)NOL_2(CO_2CH_3)$	PPh_3		67·0	4·8	1·8	9·1	67·2	4·8	2·0	8·9
	dppe/2		61·3	5·0	2·5	10·8	61·0	4·8	2·5	10·1

[a] Molar conductivities (Ω^{-1} cm^2 mol^{-1}) measured in nitrobenzene (10^{-3}M). [b] dppe = 1,2-Bisdiphenylphosphinoethane. [c] Cl (found) 0·0.

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[6] J. P. Collman, P. Farnham, and G. Dolcetti, *J. Amer. Chem. Soc.*, 1971, **93**, 1788.

[7] R. J. Mawby, D. Morris, E. M. Thorsteinson, and F. Basolo, *Inorg. Chem.*, 1966, **5**, 27.

[8] M. Aresta, P. Giannoccaro, M. Rossi, and A. Sacco, *Inorg. Chim. Acta*, 1971, **5**, 115.

[9] C. G. Pierpont, A. Pucci, and R. Eisenberg, *J. Amer. Chem. Soc.*, 1971, **93**, 3050.

Addition of methoxide ion to the cations $[Fe(CO)_2NO(PPh_3)_2]^+$ and $[Fe(CO)_2NO(dppe)]^+$ in anhydrous methanol leads to attack at a carbonyl group and the formation of the neutral carboxy-derivatives, $Fe(CO)(NO)(PPh_3)_2CO_2Me$ and $Fe(CO)(NO)(dppe)CO_2Me$. Such behaviour is commonly observed for cationic carbonyl species and no evidence for addition to the NO group was found. Neither carboxy-compound is particularly stable and in an attempt to obtain their mass spectra only peaks corresponding to the decomposition products $Fe(CO)_3L_2$, $Fe(NO)_2L_2$ ($L = PPh_3$ or $\frac{1}{2}$dppe); $Fe(CO)_4L$ and $Fe(CO)(NO)_2L$ ($L = PPh_3$) were observed. Indeed slow conversion of the PPh_3 species into these products occurs even at room temperature *in vacuo*. The dppe complex, which was recrystallised from benzene–heptane as orange needles, showed a sharp singlet at τ 6·78 in its n.m.r. spectrum ([$^2H_5$]pyridine) integrating correctly for the OCH_3 protons. The triphenylphosphine complex could not be recrystallised without causing extensive decomposition, nevertheless the product obtained directly from the reaction mixture gave satisfactory analysis (Table 1). Other cations of the series also reacted with OMe^- to produce similar but unstable carboxy-derivatives which were identified on the basis of their i.r. spectra only. No variation in ligand reactivity (*i.e.* away from CO to NO) was noted over the whole range of phosphines employed. Such behaviour is perhaps not unexpected since any increase in ν_{NO} (a useful guide to the susceptibility of NO to nucleophilic attack) is also accompanied by a corresponding increase in ν_{CO} (see Table 2). It is also apparent from these studies that the change in structure in going from $[Fe(CO)_2NO(PPh_3)_2]^+$ to $[Fe(CO)_2NO(dppe)]^+$ does not modify the relative reactivities of the CO and NO groups towards nucleophilic addition, again in keeping with the relatively low value observed for ν_{NO}.

TABLE 2

I.r. data[a] (cm^{-1}) for the nitrosyl complexes

Compound	L	ν(CO)	ν(NO)
$[Fe(CO)_2NOL_2]PF_6$	$P(OPh)_3$	2085s, 2030vs	1824vs
	$P(OMe)_3$	2075s, 2018vs	1815vs
	PPh_3	2051s, 1995vs	1792vs
	$PMePh_2$	2050s, 1991vs	1788vs
	PMe_2Ph	2047s, 1987vs	1785vs
	PEt_3	2045s, 1984vs	1783vs
	PCy_3	2037s, 1975vs	1770vs
	dppe/2	2071vs, 2014vs	1800vs
	$AsPh_3$	2052s, 1997vs	1790vs
$[Fe(CO)NOL_3]PF_6$	$P(OMe)_3$	2003vs	1780vs
	$PMePh_2$	1959vs	1745vs
	PMe_2Ph	1950vs	1736vs
$[FeNO(dppe)_2]PF_6$[b]			1681vs
$\{[Fe(CO)NO(dppe)]_2dppe\}(PF_6)_2$[b]		1975vs	1759vs
$[FeNO(dppe)]_2BPh_4$[b]			1697vs
$Fe(CO)(NO)(PPh_3)_2CO_2Me$[b]		1915vs, 1609s[c], 1603s[c]	1693vs
$Fe(CO)(NO)(dppe)CO_2Me$[b]		1952vs, 1942vs, 1605s[c]	1727vs

[a] All measurements in CH_2Cl_2 solution unless stated otherwise. [b] Nujol mull. [c] ν_{CO} In CO_2Me moiety.

The carboxy-compounds reported here are similar to the acyl species $Fe(CO)_2NO(PPh_3)COR$ obtained by Pauson *et al.*[10] from the reaction of $[Fe(CO)_3NO]^-$ with alkyl halides (RX) followed by treatment with triphenylphosphine. However, the complexes obtained in this work react with acid (HPF_6) to regenerate their cationic precursors in virtually quantitative yield. The corresponding reactions with $[Fe(CO)(NO)L_3]^+$ did not lead to *stable* carboxy-derivatives and again there was no evidence for attack on the co-ordinated NO. The fully substituted complex, $[Fe(NO)(dppe)_2]PF_6$, did not react with methoxide ion. This is in keeping with the low value observed for ν_{NO} (1697 cm^{-1}).

$$[Fe(CO)_2NOL_2]^+ \underset{H^+}{\overset{OMe^-}{\rightleftharpoons}} Fe(CO)(NO)L_2CO_2Me$$

EXPERIMENTAL

I.r. spectra were recorded on a Perkin-Elmer 257 spectrophotometer, and ^1H n.m.r. spectra were measured on a Varian HA 100 spectrometer. Microanalytical data were obtained by the Microanalytical Department of these Laboratories. Nitrosonium hexafluorophosphate was acquired from the Ozark Mahoning Company (U.S.A.) and the starting compounds $Fe(CO)_3L_2$ were all prepared by reaction of the free ligand with tricarbonylcyclo-octatetraeneiron in refluxing heptane or toluene.[11] All reactions were carried out under an atmosphere of oxygen-free nitrogen.

Dicarbonylnitrosyl(1,2-*bisdiphenylphosphinoethane*)*iron*(0) *Hexafluorophosphate*.—Nitrosonium hexafluorophosphate (1·47 g, 50% excess) was added, with vigorous stirring, to freshly recrystallised $Fe(CO)_3(dppe)$ (3·0 g) in dry benzene (30 ml) and anhydrous methanol (15 ml). A red solution rapidly formed. This was stirred for 20 min and then a large quantity of diethyl ether was slowly added causing the formation of fine orange crystals (3·46 g) which were washed with ether and could be recrystallised from tetrahydrofuran–ether solution.

The other cations of this type where $L = P(OPh)_3$, $P(OMe)_3$, PPh_3, $AsPh_3$, $PMePh_2$, PMe_2Ph, PEt_3, and PCy_3, were all isolated in 90—100% yields in a similar fashion from the corresponding $Fe(CO)_3L_2$ complexes.

Reaction of $Fe(CO)_3(PPh_3)_2$ *with* HPF_6 *and Isopentyl Nitrite*.—A few drops of 60% hexafluorophosphoric acid was added to isopentyl nitrite (0·15 ml) in benzene (3 ml) and methanol (2 ml). Tricarbonylbis(triphenylphosphine)-iron(0) (200 mg) was then added to the reaction mixture with vigorous stirring and after 15 min a large quantity of diethyl ether was added. The product (232 mg) was filtered off and washed with ether. This yellow solid was shown to be *dicarbonylnitrosylbis*(*triphenylphosphine*)*iron*(0) *hexafluorophosphate* by comparison of its i.r. spectrum with that of an authentic sample.

Carbonylnitrosyltris(*dimethylphenylphosphine*)*iron*(0) *Hexafluorophosphate*.—Dimethylphenylphosphine (30 μl) was added to a solution of $[Fe(CO)_2NO(PMe_2Ph)_2]PF_6$ (60 mg) in CH_2Cl_2 (0·5 ml). The solution was shaken periodically for 45 min and the gas evolved was allowed to escape. Dichloromethane (4 ml) was then added to the mixture which was then filtered through Kieselguhr;

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[10] F. M. Chaudhari, G. R. Knox, and P. L. Pauson, *J. Chem. Soc.* (*C*), 1967, 2255.

[11] T. A. Manuel, *Inorg. Chem.*, 1963, **2**, 854.

addition of diethyl ether gave the product as orange crystals (66 mg) which could be recrystallised suitably from dichloromethane–ether.

Carbonylnitrosyltris(trimethyl phosphite)iron(0) *Hexafluorophosphate* was prepared similarly in very high yield.

Carbonylnitrosyltris(methyldiphenylphosphine)iron(0) *Hexafluorophosphate*.—Methyldiphenylphosphine (1·02 g, 10-fold excess) and $[Fe(CO)_2NO(PMePh_2)_2]PF_6$ (350 mg) were warmed together in refluxing dichloromethane solution (5 ml) for a period of 24 h. Then, after the cooled solution had been filtered through Kieselguhr, diethyl ether was slowly added with vigorous stirring giving an orange precipitate (0·37 g) which was filtered and carefully washed with ether. The cation could be recrystallised from methanol–ether.

Nitrosylbis(1,2-diphenylphosphinoethane)iron(0) *Hexafluorophosphate and the Dicationic Species* $\{[Fe(CO)NO(dppe)]_2(dppe)\}(PF_6)_2$.— 1,2-Bisdiphenylphosphinoethane (0·32 g) and $[Fe(CO)_2NO(dppe)]PF_6$ (1·0 g) were stirred together in refluxing CH_2Cl_2 (1·5 ml) for 2 h. A further portion of dppe (0·16 g) was then added and after a further 3 h yet more dppe (0·20 g) was added. The reaction was monitored by i.r. spectroscopy and, when this indicated the disappearance of the starting cation, the resulting residue was extracted with methanol (50 ml). The material that did not dissolve was then recrystallised from methanol, in which it is very sparingly soluble, to give khaki crystals of $[FeNO(dppe)_2]PF_6$ (0·81 g). The methanol solution obtained as above was evaporated and its residue was recrystallised from dichloromethane–ether to yield the orange product $\{[Fe(CO)NO(dppe)]_2(dppe)\}(PF_6)_2$ (0·39 g).

Nitrosylbis(1,2-bisdiphenylphosphinoethane)iron(0) *Tetraphenylborate*.—Sodium tetraphenylborate (0·20 g) and *N*-methyl-*N*-nitrosotoluene-*p*-sulphonamide (0·16 g) were stirred together with an $FeHCl(dppe)_2$ (0·50 g) suspension in refluxing ethanol (20 ml) for 30 min. The initial red of the hydride slowly decolourised to a light buff hue and after cooling the fine suspension was filtered off, washed with aqueous methanol, and recrystallised from methanol to give dark rhombs (0·44 g).

Methoxycarbonylcarbonylnitrosylbis(triphenylphosphine)iron(0).—A freshly recrystallised pure sample of $[Fe(CO)_2NO(PPh_3)_2]PF_6$ (70 mg) was suspended in carefully dried methanol and NaOMe in methanol solution (0·4M; 5 ml) (previously prepared by the dissolution of clean sodium) was added. The mixture was stirred for 30 min, the yellow suspension gradually acquiring a light buff colour. This was then filtered off, washed on the sinter with methanol (2 × 2 ml), and dried; yield (42 mg).

The dppe analogue was prepared similarly in good yield and could be recrystallised from dry benzene–heptane.

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