Amino-derivatives of Metals and Metalloids. Part IV.[1] Aminosilylation and Aminophosphination of Some Unsaturated Substrates

By R. H. Cragg and M. F. Lappert

The aminometallation of carbon dioxide and disulphide, and phenyl iso- and isothio-cyanate are described, using $Me_2Si(NEt_2)_2$, $Me_2Si(NHEt)_2$, $PhP(NMe_2)_2$, and $PhP(NMe_2)Cl$. The results are correlated with data on other systems. Structures are suggested for the new carbamato-, dithiocarbamato-, ureido-, and thioureido-silanes and -phosphines, on the basis of alcoholyses, infrared spectra, and analogy with previously studied systems.

In Part II,[2] we described the aminostannylation of carbon dioxide, keten, carbon disulphide, phenyl iso- and isothio-cyanate, benzonitrile, di-*p*-tolylcarbodi-imide, sulphur dioxide, and *N*-thionylaniline. The recently recognised[3] aminometallation reactions

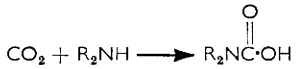
$$M{-}NR_2 + A{=}B \longrightarrow M{-}A{-}B{-}NR_2$$

were then surveyed, and are also considered in a forthcoming Review,[4] in the wider context of the behaviour of derivatives of metals and metalloids as 1,2-dipolarophiles.

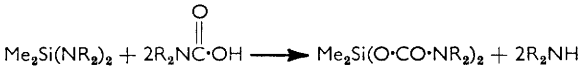
We now report extensions of our studies to derivatives of silicon and phosphorus(III). Since the original Communication,[3] a number of other workers have made contributions in this area. All the data, including some isolated early observations[5-9] and the present results, are summarised in Table 1. The characterisation of the new compounds (I—XII) is indicated in Table 2.

The quoted yields refer to purified materials; the crude compounds were obtained virtually in quantitative amounts.

Mixing the dipolorophile and the dipole at ambient temperature generally caused an exothermic reaction. The $Me_2Si(NMe_2)_2/CO_2$ and $Me_2Si(NMe_2)_2/CS_2$ systems were exceptional. In fact, a trace of amine appeared to be necessary to initiate these reactions, as had earlier been noticed by Breederveld for corresponding experiments with Me_3SiNMe_2.[7,8] A reasonable interpretation (cf., refs. 7 and 8) is that an ionic chain mechanism is involved.



$$CO_2 + R_2NH \longrightarrow R_2N\overset{O}{\overset{\|}{C}}{\cdot}OH$$



$$Me_2Si(NR_2)_2 + 2R_2N\overset{O}{\overset{\|}{C}}{\cdot}OH \longrightarrow Me_2Si(O{\cdot}CO{\cdot}NR_2)_2 + 2R_2NH$$

On the other hand, such catalysis does not appear to be required for the other insertion processes, and we find

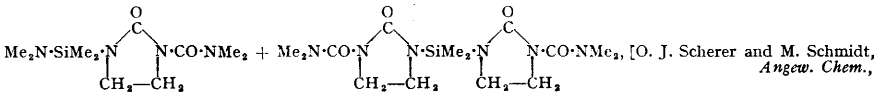
TABLE 1

Products of aminosilylation and aminophosphination

	Dipole					
Dipolarophile	CO_2	CS_2	$R''NCO$ †	$RNCS$	$CH_2{=}C{=}O$	SO_3
$R_3SiNR'_2$	$R_3SiOCO{\cdot}NR'_2$ ^{7,11}	$R_3SiSCS{\cdot}NR'_2$ ^{8}	$R_3SiNR''CO{\cdot}NR'_2$ ^{11,16} *or* $R_3Si[N(R'')CO{\cdot}]_2NR'_2$ ^{13,15}	$R_3SiNR''CS{\cdot}NR'$	$R_3SiCH_2{\cdot}CO{\cdot}NR'_2$ ^{9}	$R_3SiOSO_2{\cdot}NR'_2$
R_3SiNHR'				$R_3SiNR''CS{\cdot}NHR'$ ^{14}		
$R_2Si(NR'_2)_2$	$R_2Si(OCO{\cdot}NR'_2)_2$ *	$R_2Si(SCS{\cdot}NR'_2)NR'_2$ *	$R_2Si(NR''CO{\cdot}NR'_2)_2$ *	$R_2Si(NR''CS{\cdot}NR'_2)_2$ *		
$R_2Si(NHR')_2$	$R_2Si(OCO{\cdot}NHR')_2$ *	$R_2Si(SCS{\cdot}NHR')NHR'$ *	$R_2Si(NR''CO{\cdot}NHR')_2$ *	$R_2Si(NR''CS{\cdot}NHR')_2$ *		
$(R_3Si)_2NH$			$R_3SiNH{\cdot}CO{\cdot}N(R'')SiR_3$ ^{13,15}			
$(R_3Si)_2NR'$			$R_3SiNR'{\cdot}CO{\cdot}N(R'')SiR_3$ ^{14,15} *or* $(R_3SiNR''{\cdot}CO)_2NR'$ ^{14}			
$(R_2SiNH)_{3\text{ or }4}$			$R_2Si(NR''CO{\cdot}NH)_2$ ^{12}			
$RP(NR'_2)_2$	$RP(OCO{\cdot}NR'_2)NR'_2$ ^{11}	$RP(SCS{\cdot}NR'_2)_2$ ^{11} * *or* $RP(SCS{\cdot}NR'^{2})NR'_2$ ^{11}	$RP(NR''CO{\cdot}NR'_2)_2$ *	$RP[NR''CS{\cdot}NR'_2]NR'_2$ ^{11}		
$P(NR_2)_3$	$P(OCO{\cdot}NR_2)_3$ ^{11} *or* $R_2NP(OCO{\cdot}NR_2)_2$ ^{11}	$P(SCS{\cdot}NR_2)_3$ ^{10,11} *or* $R_2NP(SCS{\cdot}NR_2)_2$ ^{11}		$(R_2N)_2PNR''CS{\cdot}NR_2$ ^{11}		
$ROP(NR'_2)_2$	$ROP(OCO{\cdot}NR'_2)NR'_2$ ^{11}	$ROP(SCS{\cdot}NR'_2)_2$ ^{11}		$ROP(NR''CS{\cdot}NR'_2)NR'_2$ ^{11}		
$(RO)_2PNR'_2$	$(RO)_2POCO{\cdot}NR'_2$ ^{11}					
$RP(NR'_2)Cl$			$RP(NR''CO{\cdot}NR'_2)Cl$ *			

* Present work.

† Also $Me_2Si(NMe_2)_2/(CH_2NCO)_2 \longrightarrow$



[O. J. Scherer and M. Schmidt, *Angew. Chem.*, 1965, **77**, 456.]

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[1] Part III, K. Jones and M. F. Lappert, *J. Organometallic Chem.*, 1965, **3**, 295.

[2] Part II, T. A. George, K. Jones, and M. F. Lappert, *J. Chem. Soc.*, 1965, 2157.

[3] K. Jones and M. F. Lappert, *Proc. Chem. Soc.*, 1962, 358.

[4] M. F. Lappert and B. Prokai, *Adv. Organometallic Chem.*, 1966, in the press.

[5] A. Michaelis, *Ber.*, 1898, **31**, 1037.

[6] M. Schmidt and H. Schmidbaur, *Angew. Chem.*, 1958, **70**, 657.

[7] H. Breederveld, *Rec. Trav. chim.*, 1960, **79**, 1126.

[8] H. Breederveld, *Rec. Trav. chim.*, 1962, **81**, 276.

[9] W. W. Limburg and H. W. Post, *Rec. Trav. chim.*, 1962, **81**, 430.

[10] H. J. Vetter and H. Nöth, *Chem. Ber.*, 1963, **96**, 1308.

[11] G. Oertel, H. Malz, and H. Holtschmidt, *Chem. Ber.*, 1964, **97**, 891.

[12] W. Fink, *Chem. Ber.*, 1964, **97**, 1424.

[13] W. Fink, *Chem. Ber.*, 1964, **97**, 1433.

[14] J. F. Klebe, J. B. Bush, and J. E. Lyons, *J. Amer. Chem. Soc.*, 1964, **86**, 4400.

[15] D. Y. Zhinkin, M. M. Morgunova, K. K. Popkov, and K. A. Andrianov, *Doklady Akad. Nauk S.S.S.R.*, 1964, **158**, 641.

no new evidence to change our earlier opinion (suggested for chloro-,[16] amino-,[17,18] and alkoxy [17]-boration) that these reactions proceed through a low-energy 4-centre transition state, and are thermodynamically controlled.

TABLE 2

Carbamato-, dithiocarbamato-, ureido-, and thioureido-silanes and -phosphines

Compound	M. p.	Yield (%)	Principal absorption maxima (cm.$^{-1}$) N–H str.	C=O str.	C=S str.
(I)	(n_D^{20} 1·4458)	90		1688	
(II)	128—130°	82	3300	1620, 1562	
(III)	76—78	85			1205, 1126
(IV)	52—54	82	3295		1195
(V)	50—52			1635	
(VI)	89—91	71	3315	1710	
(VII)	178—180	94		1645	
(VIII)	86—90	59			1206
(IX)	113—115	72	3300, 3240		1202
(X)	57—59	24		1700, 1640	
(XI)	169—170	84			1240
(XII)	67—69	84		1670	

$Me_2Si(O\,CO{\cdot}NEt_2)_2$ (I)

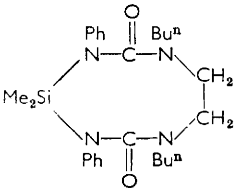
$Me_2Si(O{\cdot}CO{\cdot}NHEt)_2$ (II)

$Me_2Si(S{\cdot}CS{\cdot}NEt_2)NEt_2$ (III)

$Me_2Si(S{\cdot}CS{\cdot}NHEt)NHEt$ (IV)

$Me_2Si(NPh{\cdot}CO{\cdot}NEt_2)_2$ (V)

$Me_2Si(NPh{\cdot}CO{\cdot}NHEt)_2$ (VI)



(VII)

$Me_2Si(NPh{\cdot}CS{\cdot}NEt_2)_2$ (VIII)

$Me_2Si(NPh{\cdot}CS{\cdot}NHEt_2)_2$ (IX)

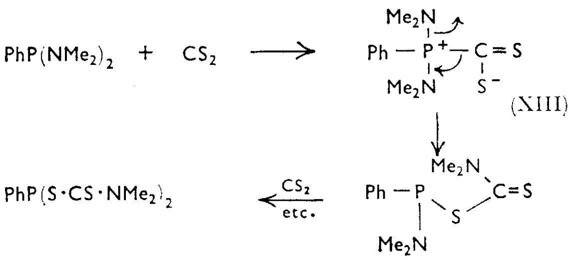
$PhP(NPh{\cdot}CO{\cdot}NMe_2)_2$ (X)

$PhP(S{\cdot}CS{\cdot}NMe_2)_2$ (XI)

$PhP(NPh{\cdot}CO{\cdot}NMe_2)Cl$ (XII)

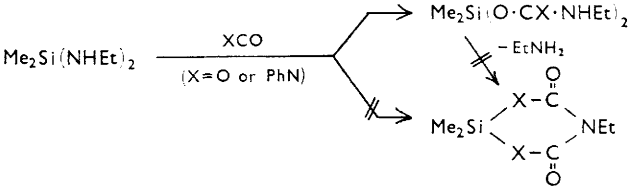
In support, it is found that at least one of these reactions is reversible. It may also be noted that cleavage of M–NR$_2$ (M = Si, P, or B [16,17]) is preferred over that of M–OR, M–Cl, or M–Ph.

The phosphine reactions may well be mechanistically more complex, with the phosphorus atom as the basic site. Thus, the addition of carbon disulphide to bis(dimethylamino)phenylphosphine is immediately accompanied by the development of a deep-red colour, which slowly fades as the mixture is warmed. Since many phosphorus(III) compounds form phosphonium complexes with carbon disulphide,[19] it is probable that the mechanism of the $PhP(NMe_2)_2/CS_2$ reactions involves an intermediate (XIII) of this type. A similar proposal has been made with reference to some reactions of hydrazino-phosphines.[20]

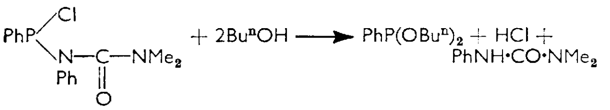


(XIII)

With bis(ethylamino)dimethylsilane, reactions might have been expected to take either of two paths. In contrast to the corresponding aminoboranes,[18] cyclic compounds were not obtained.



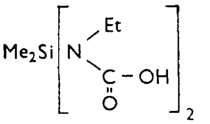
The structures of the products in Table 2 were established by alcoholyses, infrared spectra, and by analogy with products of similar reactions of amino-derivatives of tin(IV) [2] and boron.[17] Alcoholysis is especially important in order to establish relative migratory aptitudes. Thus, the structure of $PhP(NPh{\cdot}CO{\cdot}NMe_2)Cl$ is in large measure established by the identification of the products of the following reaction. Isomeric compounds $Me_2NP(NPh{\cdot}CO{\cdot}Ph)Cl$ or $PhP(NPh{\cdot}CO{\cdot}Cl)NMe_2$ would yield different derivatives.



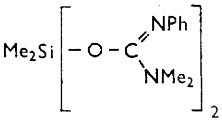
$$PhP(Cl)(NPh{\cdot}CO{\cdot}NMe_2) + 2Bu^nOH \longrightarrow PhP(OBu^n)_2 + HCl + PhNH{\cdot}CO{\cdot}NMe_2$$

The formation of the 9-membered cyclic bisureidosilane (VII) is interesting. A similar reaction pattern has been established for corresponding boron compounds.[17]

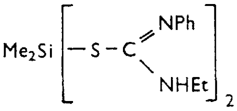
The structurally significant spectral assignments are indicated in Table 2. The presence of absorption bands arising from N–H stretching in the derivatives obtained



(XIV)



(XV)



(XVI)

from $Me_2Si(NHEt)_2$ excludes the possibility that reactions involve insertion into the N–H rather than the Si–N bond. For example, the product obtained with carbon dioxide is not (XIV). The spectra of CO$_2$

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[16] M. F. Lappert and B. Prokai, *J. Chem. Soc.*, 1963, 4223.

[17] R. H. Cragg, M. F. Lappert, and B. P. Tilley, *J. Chem. Soc.*, 1964, 2108.

[18] R. H. Cragg and M. F. Lappert, *Adv. Chem. Ser.*, 1964, No. 42, 220.

[19] K. Issleib and W. Seidel, *Chem. Ber.*, 1959, **92**, 2681.

[20] R. P. Nielsen, J. F. Vincent, and H. H. Sisler, *Inorg. Chem.*, 1963, **2**, 760.

adducts were carefully compared with those of corresponding CS_2 adducts, and likewise PhNCO with PhNCS derivatives. These comparisons provide a small number of mutally exclusive bands and proved a basis for the assignments indicated in Table 2. As a result, structures such as (XV) and (XVI) are almost certainly eliminated.

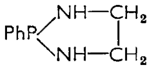
EXPERIMENTAL

General Procedures and Starting Materials.—Toluene, benzene, and diethyl ether were stored over sodium and distilled before use. n-Butanol was dried by distillation from calcium. Light petroleum had b. p. 40—60°. Microanalyses (C, H, S) were carried out by Mr. B. Manohin and his staff, and we offer them our best thanks. Bis(diethylamino)dimethylsilane[21] and bis(ethylamino)dimethylsilane[22] were prepared by published procedures. The usual precautions were taken to deal with air-sensitive materials.[1,2,16-18] Infrared spectra were recorded as mulls on a Perkin-Elmer model 21 (2—15 μ) spectrophotometer.

The Preparation of Aminophosphines.[23]—Dichlorophenylphosphine (11·3 g., 1 mol.) in light petroleum (50 ml.) was added to a solution of dimethylamine (10·7 g., 4 mol.) in the same solvent (40 ml.) at −78°. The mixture was heated under reflux for 2½ hr. After cooling, dimethylammonium chloride (10·3 g., 100%) was filtered off. The combined washings and filtrate afforded bis(dimethylamino)phenylphosphine (8·5 g., 65%), b. p. 57—59°/0·1 mm., n_D^{20} 1·5476, identified by comparison with an authentic sample.[24]

Dichlorophenylphosphine (16·77 g., 1 mol.) in benzene (100 ml.) was cooled to 0°. Dimethylamine (4·03 g., 1 mol.) was added and, after ~1 hr. at 20°, then dropwise triethylamine (8·68 g., 1 mol.) in benzene (50 ml.). After 1 hr. under reflux, filtration afforded triethylammonium chloride (11·82 g., 100%), and from the filtrate there was obtained chlorodimethylaminophenylphosphine[25] (13·9 g., 79·4%), b. p. 118°/0·1 mm., n_D^{20} 1·5440 (Found: C, 47·0; H, 5·3; Cl, 17·2; N, 6·8. Calc. for $C_8H_{11}ClNOP$: C, 47·2; H, 5·4; Cl, 17·4; N, 6·9%). This experiment was carried out by Dr. P. A. Barfield.

Ethylenediamine and bis(dimethylamino)phenylphosphine were refluxed in 1 : 1 molar ratio until a quantitative yield of dimethylamine was collected in a trap at −78°. It was hoped in this way to prepare the heterocycle (XVII). However, the residue could not be distilled and appeared to be polymeric.



(XVII)

Interaction of Carbon Dioxide and Bis(diethylamino)dimethylsilane.—Solid carbon dioxide was washed with light petroleum and added to bis(diethylamino)dimethylsilane (2·20 g., 1 mol.) at 20°. Initially there was no reaction, but on addition of a trace of diethylamine there was heat of mixing and *bis-*(NN*-diethylcarbamato)dimethylsilane* (2·85 g.) (I) (Found: C, 49·6; H, 8·5; N, 9·9. $C_{12}H_{26}N_2O_4Si$ requires C, 49·7; H, 8·85; N, 9·65%) was obtained. Upon attempted distillation, the product decomposed, with elimination of carbon dioxide. However, it has been reported[26] as a liquid, b. p. 105—107°/0·2 mm., n_D^{20} 1·4370.

Interaction of Carbon Dioxide and Bis(ethylamino)dimethylsilane.—Solid carbon dioxide was washed with light petroleum and added to bis(ethylamino)dimethylsilane (2·2 g., 1 mol.). The reaction was exothermic. On filtering the mixture, *bis-*(N*-ethylcarbamato)dimethylsilane* (II) (2·35 g.) (Found: C, 40·8; H, 7·6; N, 12·2. $C_8H_{18}N_2O_4Si$ requires C, 41·1; H, 7·7; N, 11·9%) was obtained.

Interaction of Carbon Disulphide and Bis(diethylamino)dimethylsilane.—Excess of carbon disulphide was added to bis(diethylamino)dimethylsilane (1·85 g., 1 mol.) at 20°, and upon addition of a trace of diethylamine the mixture became warm. After 1 hr., the excess of carbon disulphide was removed under vacuum, leaving *diethylamino-*(NN*-diethyldithiocarbamato)dimethylsilane* (III) (2·16 g.) (Found: C, 44·7; H, 10·1; N, 9·9. $C_{11}H_{26}N_2S_2Si$ requires C, 45·2; H, 9·8; N, 10·1%).

Interaction of Carbon Disulphide and Bis(ethylamino)dimethylsilane.—Similarly, from an excess of carbon disulphide and bis(ethylamino)dimethylsilane (3·4 g., 1 mol.) there was obtained under vacuum *ethylamino-*(NN*-ethyldithiocarbamato)dimethylsilane* (IV) (3·9 g.) (Found: C, 38·8; H, 7·6; N, 12·4. $C_7H_{18}N_2S_2Si$ requires C, 38·0; H, 8·1; N, 12·6%).

Interaction of Phenyl Isocyanate and Bis(diethylamino)dimethylsilane.—Phenyl isocyanate (4·4 g., 2 mol.) and bis(dimethylamino)dimethylsilane (3·7 g., 1 mol.) were heated under reflux in light petroleum (30 ml.) for 1 hr. On removal of the solvent, a viscous liquid remained. After 5 days at 20°, the residue afforded *dimethylbis-*(N*-phenyl-*N′N′*-diethylureido)silane* (V) (5·9 g.) (Found: C, 64·0; H, 7·4; N, 13·0. $C_{24}H_{36}N_4O_2Si$ requires C, 63·6; H, 7·1; N, 12·8%).

Interaction of Phenyl Isocyanate and Bis(ethylamino)dimethylsilane.—Phenyl isocyanate (5·4 g., 2 mol.) was added to bis(ethylamino)dimethylsilane (3·3 g., 1 mol.) in benzene (25 ml.). The reaction was exothermic. After ½ hr. under reflux, the solvent was removed and *dimethylbis-*(N*-phenyl-*N′*-ethylureido)silane* (VI) (6·2 g.) (Found: C, 62·0; H, 7·5; N, 14·2. $C_{20}H_{28}N_4O_2Si$ requires C, 62·4; H, 7·3; N, 14·6%) was obtained.

Preparation of 1,3*-Di-n-butyl-*2,2*-dimethyl-*1,3,2*-diazasilolidine and Its Reaction with Phenyl Isocyanate.*—Bis(diethylamino)dimethylsilane (6·0 g., 1 mol.) and *NN′*-di-n-butylethylenediamine (7·60 g., 1 mol.) were heated under reflux in toluene (25 ml.) for 3 hr. Diethylamine (3·8 g., 87%) (authenic infrared spectrum) was eliminated and trapped at −78°. Upon removal of the solvent, a viscous residue remained, which afforded the *diazasilolidine* (7·4 g., 80%), b. p. 64°/0·2 mm. (Found: C, 62·4; H, 11·9; N, 12·5. $C_{12}H_{28}N_2Si$ requires C, 63·0; H, 12·3; N, 12·3%).

Phenyl isocyanate (3·5 g., 2 mol.) was added to the silolidine (3·6 g., 1 mol.) in diethyl ether (25 ml.) at −78°. A white solid crystallised gradually, and was identified as the *cyclic bisureidosilane* (VII) (6·7 g.) (Found: C, 68·0; H, 8·3; N, 13·7. $C_{26}H_{24}N_4O_2Si$ requires C, 67·0; H, 8·2; N, 12·0%).

Interaction of Phenyl Isothiocyanate and Bis(diethylamino)dimethylsilane.—Phenyl isothiocyanate (1·07 g., 2 mol.) and bis(diethylamino)dimethylsilane (0·8 g., 1 mol.) were heated

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[21] H. H. Anderson, *J. Amer. Chem. Soc.*, 1952, **74**, 1421.

[22] E. Larsson and B. Smith, *Acta Chem. Scand.*, 1949, **3**, 48.

[23] D. S. Payne, A. L. Porte, and A. P. Lane, *J. Chem. Soc.*, 1962, 3984.

[24] P. A. Barfield, Ph.D. Thesis, Manchester, 1965.

[25] H. Nöth and H. J. Vetter, *Chem. Ber.*, 1963, **96**, 1109.

[26] G. Oertel, H. Holtschmidt, and H. Malz, G.P. 1,157,226 (1964) (*Chem. Abs.*, 1964, **60**, 6868).

under reflux in benzene (40 ml.) for 3 hr. After 12 hr. at 20°, the yellow solid *dimethylbis-*(N-*phenyl-*N'N'*-diethylthioureido)silane* (VIII) (1·1 g.) (Found: C, 49·9; H, 5·2; N, 11·3. $C_{20}H_{26}N_4S_2Si$ requires C, 50·9; H, 5·5; N, 11·9%) crystallised out.

Interaction of Phenyl Isothiocyanate and Bis(*ethylamino*)*-dimethylsilane.*—Phenyl isothiocyanate (5·0 g., 2 mol.) was added to bis(ethylamino)dimethylsilane (2·7 g., 1 mol.) in benzene (30 ml.). The reaction was exothermic. After 1 hr. under reflux, the solvent was removed, leaving *dimethylbis-*(N-*phenyl-*N'*-ethylthioureido)silane* (IX) (5·6 g.) (Found: C, 56·8; H, 6·1; N, 13·2. $C_{18}H_{28}N_4S_2Si$ requires C, 57·2; H, 6·7; N, 13·4%).

Interaction of Phenyl Isocyanate and Bis(*dimethylamino*)*-phenylphosphine.*—Phenyl isocyanate (3·8 g., 2 mol.) and bis(dimethylamino)phenylphosphine (3·05 g., 1 mol.) were heated under reflux in benzene (15 ml.) for 3 hr. After removal of the solvent, a viscous liquid remained, which afforded *phenylbis-*(N-*phenyl-*N'N'*-dimethylureido)phosphine* (X) (1·6 g.) (Found: C, 65·9; H, 5·4; N, 12·6. $C_{24}H_{27}N_4O_2P$ requires C, 66·5; H, 6·2; N, 12·9%), after shaking with diethyl ether–light petroleum (15 ml.).

Interaction of Carbon Disulphide and Bis(*dimethylamino*)*-phenylphosphine.*—Excess of carbon disulphide was added to bis(dimethylamino)phenylphosphine (0·8 g., 1 mol.). Upon addition of carbon disulphide, the mixture became warm and a transient blood-red colour developed, which rapidly disappeared. On removal of the excess of carbon disulphide (20°/10 mm.), *bis-*(NN*-dimethyldithiocarbamato)phenylphosphine* (XI) (1·2 g.) (Found: C, 42·5; H, 4·7; N, 8·2. $C_{12}H_{17}N_2PS_4$ requires C, 41·6; H, 4·9; N, 8·1%) was obtained.

Interaction of Phenyl Isocyanate and Chlorodimethylaminophenylphosphine.—Phenyl isocyanate (2·65 g., 1 mol.) and chlorodimethylaminophenylphosphine (4·20 g., 1 mol.) were heated under reflux in benzene (20 ml.) for 3 hr. After removal of the solvent, a white solid was obtained. This was washed with diethyl ether and identified as *chloro-*(N-*phenyl-*N'N'*-dimethylureido)phenylphosphine* (XII) (5·7 g.) (Found: C, 59·6; H, 6·0; N, 9·2. $C_{15}H_{16}ClN_2OP$ requires C, 58·8; H, 5·2; N, 9·15%).

n-Butanol (2 mol.) was added to the urea (1 mol.). Upon refluxing (1 hr.), hydrogen chloride was evolved, but no dimethylamine. The residue was washed with ether and on filtering and removal of the solvent afforded bis-(n-butoxy)phenylphosphine in good yield; it was characterised by its infrared spectrum. *NN*-Dimethyl-*N'*-phenylurea was the solid residue.

We thank Dr. J. C. McCoubrey and Albright and Wilson (Mfg.), Ltd., for a gift of dichlorophenylphosphine, Dr. P. A. Barfield for preparing some of the phosphines, and the D.S.I.R. for a grant (to R. H. C.).

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