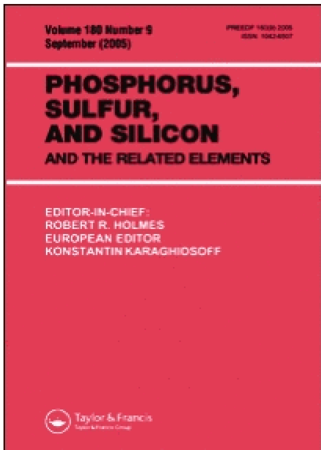


Phosphorus, Sulfur, and Silicon and the Related Elements
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713618290

Studies on Organophosphorus Compounds: The Synthesis of [1,3,2]-Diazaphospholes and [1,3,2]-Oxaazaphospholes

O. A. Omran [a]; H. M. Moustafa [a]
[a] Chemistry Department, Sohag, Egypt

Online Publication Date: 01 November 2006

To cite this Article: Omran, O. A. and Moustafa, H. M. (2006) 'Studies on Organophosphorus Compounds: The Synthesis of [1,3,2]-Diazaphospholes and [1,3,2]-Oxaazaphospholes', Phosphorus, Sulfur, and Silicon and the Related Elements, 181:11, 2519 — 2528

To link to this article: DOI: 10.1080/10426500600754794
URL: http://dx.doi.org/10.1080/10426500600754794

Phosphorus, Sulfur, and Silicon, 181:2519–2528, 2006

ISSN: 1042-6507 print / 1563-5325 online
DOI: 10.1080/10426500600754794

Studies on Organophosphorus Compounds: The Synthesis of [1,3,2]-Diazaphospholes and [1,3,2]-Oxaazaphospholes

O. A. Omran
H. M. Moustafa
Chemistry Department, Sohag, Egypt

*A number of spiro[cyclopentane (cyclohexane, and cycloheptane)-1,4′-perhydro-[1,3,2]diazaphosphole] derivatives (**3a–c**, **5a–c**, **11a–c**, and **12a–c**) and spiro-[cyclopentane (cyclohexane, and cycloheptane)-1,4′-perhydro[1,3,2]oxaazaphos-phole] derivatives (**7a–c**) were prepared via an interaction of 2,4-bis-(4-methoxy-phenyl)-1,3,2,4-dithiaphosphetane-2,4-disulphide (1) with substances containing two functional groups.*

Keywords 1-Phenylamino-1-cyanocycloalkanes; 2,4-bis-(4-methoxyphenyl)-1,3,2,4-dithiaphosphetane-2,4-disulphide

INTRODUCTION

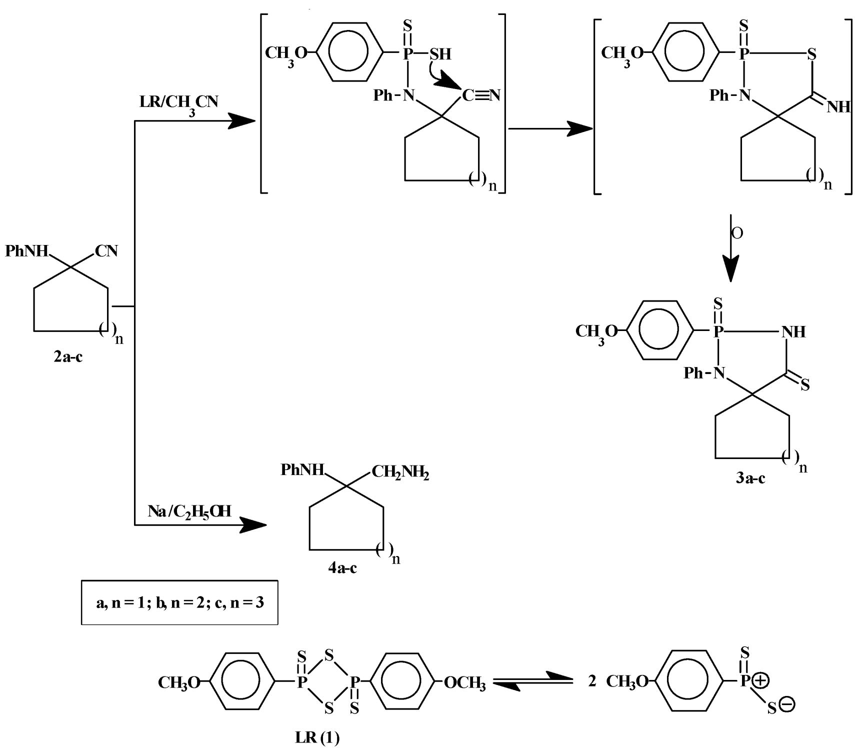
It is well known that 2,4-bis-(4-methoxyphenyl)-1,3,2,4-dithiaphosphetane-2,4-disulphide (Lawesson's Reagent, [LR]) is a most effective and versatile thiation reagent for different cabonyl compounds.[1–5] The nucleophiles attack LR at the phosphorus atom and form phosphorus heterocycles.[6–9] In view of the latest development and also in continuation of our study of phosphorus hetero-cycles,[10–13] In this article it was of interest to synthesize spiro[1,3,2]diazaphospholes and spiro[1,3,2]oxaazaphospholes from the reaction of LR with other classes of substrates with two functional groups.

RESULTS AND DISCUSSION

The reaction of 1-phenylamino-1-cyanocyclopentane(cyclohexane, and cyclo-heptane)[14] (**2a–c**) with LR (**1**) in boiling acetonitrile afforded 2′-(4-methoxy-phenyl)-4′-phenylspiro[cyclopentane (cyclohexane, and

Received February 26, 2006; accepted April 16, 2006.
Address correspondence to H. M. Moustafa, Chemistry Department, Faculty of Science, Sohag, Egypt. E-mail: hassa20002000@yahoo.com

cycloheptane)-1,4′-perhy-dro[1,3,2]diazaphosphole]-2′,5′-disulfides (**3a–c**). The reaction pathway was assumed to proceed via a nucleophilic attack of the amino group on the phosphorus of LR followed by a P-SH addition to the nitrile group and subsequent rearrangement,[16] which yielded compounds **3a–c** (*cf.* Scheme 1). The IR spectra of compounds **3a–c** showed the absence of bands corresponding to cyano groups while exhibiting the characteristic absorption band at 651, 642, and 659 cm^{-1} for P=S, respectively. ^{1}H NMR spectra of compounds **3a–c** exhibited a singlet at 3.9 ppm corresponding to OCH_3 (*cf.* Table I).



SCHEME 1

The treatment of compounds **2a–c** with sodium metal in ethanol afforded the corresponding amines **4a–c** in good yield. IR spectra of compounds **4a–c** showed the absence of absorption bands corresponding to CN groups and the appearance of new bands corresponding to NH_2 groups (3163–3412 cm^{-1}). The ^{1}H NMR ($CDCl_3$) spectra of compounds **4a–c** are in agreement with the proposed structure (*cf.* Table I). Compounds **4a–c** were allowed to react with LR (**1**) in anhydrous benzene at 80°C to give 2′-(4-methoxyphenyl)-3′-phenylspiro[cyclopentane

TABLE I Analytical and Spectral Data of the New Compounds

Compound no.	M.P.(°C) Cryst. solvent	Yield (%)	Mol. Form. (Mol. Wt.)	Analytical data Cacd./found C	H	N	S	IR (Cm^{-1})	^{1}HNMR ∂ (ppm)
3a	160	86	$C_{19}H_{21}N_2OPS_2$	58.74	5.45	7.21	16.50	3212 (NH), 1246	11.2 (br, 1H, NH), 7.8–6.9 (m, 9H,
	ethanol		(388.48)	58.50	5.27	7.03	16.23	(C—O), 651(P=S)	arom.); 3.9 (s, 3H, OCH_3), 1.9–1.4 (m, 8H, cyclic CH_2)
3b	170	83	$C_{20}H_{23}N_2OPS_2$	59.68	5.75	6.95	15.93	3266 (NH), 1253	12.9 (br, 1H, NH), 7.8–6.7 (m, 9H,
	ethanol		(402.51)	59.30	5.58	6.73	15.72	(C—O), 642 (P=S)	arom.); 3.9 (s, 3H, OCH_3), 1.9–1.2 (m, 10H, cyclic CH_2)
3c	191	79	$C_{21}H_{25}N_2OPS_2$	60.55	6.05	6.72	15.93	3259 (NH), 1239	11.9 (br, 1H, NH), 7.8–7.0 (m, 9H,
	ethanol		(416.53)	60.21	5.87	6.58	15.73	(C—O), 659 (P=S)	arom.); 3.9 (s, 3H, OCH_3), 1.9–1.2 (m, 12 H, cyclic CH_2)
4a	Liquid	77	$C_{12}H_{18}N_2$	75.74	9.53	14.72	—	3359, 3261, 3203	11.1 (br, 1H, NH), 7.5–6.9 (m, 5H,
			(190.29)	75.27	9.34	14.39		(NH_2 + NH)	arom.); 4.3 (br, 2H, NH_2); 2.8 (s, 2H, CH_2), 1.9–1.2 (m, 8 H, cyclic CH_2)
4b	Liquid	79	$C_{13}H_{20}N_2$	76.42	9.86	13.71	—	3412, 3336, 3230	13.0 (br, 1H, NH), 7.5–6.9 (m, 5H,
			(204.31)	76.01	9.61	13.49		(NH_2 + NH)	arom.); 4.9 (br, 2H, NH_2); 2.8 (s, 2H, CH_2), 1.9–1.1 (m, 10 H, cyclic CH_2)
4c	Liquid	71	$C_{14}H_{22}N_2$	77.01	10.15	12.82	—	3389, 3298, 3163	11.8 (br, 1H, NH), 7.5–6.9 (m, 5H,
			(218.34)	76.60	9.97	12.51		(NH_2+ NH)	arom.); 4.4 (br, 2H, NH_2); 2.8 (s, 2H, CH_2), 1.9–1.0 (m, 12 H, cyclic CH_2)
5a	216	61	$C_{19}H_{23}N_2OPS$	63.66	6.46	7.81	8.94	3286 (NH), 661 (P=S)	13.0 (br, 1H, NH), 7.5–6.8 (m, 9 H,
	ethanol		(358.44)	63.31	6.29	7.57	8.74		arom.); 3.9 (s, 3H, OCH_3), 2.9 (s, 2H, CH_2), 1.9–1.3 (m, 8 H, cyclic CH_2)
5b	233	59	$C_{20}H_{25}N_2OPS$	64.49	6.76	7.52	8.60	3299 (NH), 703	10.6 (br, 1H, NH), 7.5–6.8 (m, 9 H,
	ethanol		(372.46)	64.07	6.61	7.33	8.43	(P=S)	arom.); 3.9 (s, 3H, OCH_3), 2.9 (s, 2H, CH_2), 1.9–1.2 (m, 10 H, cyclic CH_2)
5c	240	68	$C_{21}H_{27}N_2OPS$	65.26	7.04	7.24	8.29	3271 (NH), 655 (P=S)	10.6 (br, 1H, NH); 7.5–6.9 (m, 9H,
	ethanol		(386.49)	64.89	6.90	7.05	8.08		arom.), 3.9 (s, 3H, OCH_3), 2.9 (s, 2H, CH_2), 1.9–1.1 (m, 12 H, cyclic CH_2)

(Continued on next page)

TABLE I Analytical and Spectral Data of the New Compounds

Compound No.	M.P.(°C) Cryst. Solvent	Yield (%)	Mol. Form. (Mol. Wt.)	Analytical Data Cacd./Found C	H	N	S	IR (Cm^{-1})	^{1}HNMR ∂ (ppm)
6a	Liquid	65	$C_{12}H_{17}NO$ (191.27)	75.35 74.94	8.95 8.77	7.32 7.17	—	3412 (OH), 3223 (NH)	10.9 (br, H, NH); 7.4–6.8 (m, 5H, arom.), 4.0 (s, 1H, OH), 3.1 (s, 2H, CH_2), 1.9–1.2 (m, 8 H, cyclic CH_2)
7a	163 ethanol	61	$C_{19}H_{20}NO_2PS$ (389.47)	58.59 58.21	4.91 4.77	3.59 3.48	16.46 16.25	1244 (C—O), 661 (P=S)	7.6–7.0 (m, 9H, arom.), 4.1 (s, 2H, OCH_2), 3.9 (s, 3H, OCH_3),1.9–1.3 (m, 8 H, cyclic CH_2)
7b	198 ethanol	59	$C_{20}H_{22}NO_2PS$ (403.49)	59.53 59.17	5.49 5.27	3.47 3.38	15.89 15.69	1233 (C—O, 669 (P=S)	7.6-7.0 (m, 9H, arom.), 4.2 (s, 2H, OCH_2), 3.9 (s, 3H, OCH_3),1.9-1.2 (m, 10 H, cyclic CH_2)
7c	186 ethanol	68	$C_{21}H_{24}NO_2PS$ (417.52)	60.41 60.05	5.79 5.61	3.35 3.26	15.35 15.16	1249 (C—O), 655 (P=S)	7.6–7.0 (m, 9H, arom.), 4.2 (s, 2H, OCH_2), 3.9 (s, 3H, OCH_3),1.9–1.2 (m, 12 H, cyclic CH_2)
8a	199 ethanol	65	$C_{19}H_{23}N_3O$ (309.41)	73.75 73.29	7.49 7.33	13.58 13.30	—	3276, 3209, 3148 (3 NH), 1649 (C=O)	11.2, 10.8, 10.5(br, 3H, 3 NH), 7.3–6.7 (m, 10 H, arom.) 3.3 (s, 2H, CH_2), 1.9–1.2 (m, 12 H, cyclic CH_2)
8b	186 dioxan	60	$C_{20}H_{25}N_3O$ (323.44)	74.27 73.89	7.79 7.63	12.99 12.80	—	3295, 3200, 3147 (3 NH), 1644 (C=O)	11.9, 10.78, 10.1(br, 3H, 3NH), 7.3–6.7 (m, 10 H, arom.) 3.3 (s, 2H, CH_2), 1.9–1.1 (m, 10 H, cyclic CH_2)
8c	173 ethanol	67	$C_{21}H_{27}N_3O$ (337.46)	74.74 74.37	8.06 7.96	12.45 12.21	—	3293, 3222, 3129 (3 NH), 1650 (C=O)	12.7, 10.8, 10.5(br, 3H, 3 NH), 7.3–6.7 (m, 10 H, arom.) 3.3 (s, 2H, CH_2), 1.9–1.2 (m, 12 H, cyclic CH_2)
9a	138 DMF	61	$C_{19}H_{23}N_3S$ (325.47)	70.11 69.75	7.12 6.93	12.91 12.71	9.85 9.69	3271, 3139 (2 NH), 1410 (SH)	12.0, 10.3 (br, 2H, 2 NH), 7.3–6.7 (m, 8 H, arom.), 4.5 (s, 1H, SH); 3.3 (s, 2H, CH_2), 1.9–1.2 (m, 8 H, cyclic CH_2)
9b	132 ethanol	59	$C_{20}H_{25}N_3S$ (339.50)	70.75 70.44	7.42 7.24	12.37 12.18	9.44 9.29	3175, 3112 (2 NH), 1432 (SH)	12.9, 11.7 (br, 2H, 2 NH), 7.3–6.7 (m, 8 H, arom.), 4.5 (s, 1H, SH); 3.3 (s, 2 H, CH_2), 1.9–1.2 (m, 10 H, cyclic CH_2)

9c	141 ethanol	68	$C_{21}H_{27}N_3S$ (353.52)	71.34 71.01	7.69 7.52	11.88 11.71	9.06 8.89	3307, 3169 (2 NH), 1427 (SH)	11.1, 9.9 (br, 2H, 2 NH), 7.3–6.7 (m, 8 H, arom.), 4.3 (s, 1H, SH); 3.3 (s, 2H, CH_2), 1.9–1.2 (m, 12 H, cyclic CH_2)
10a	203 ethanol	65	$C_{19}H_{21}N_3$ (291.41)	78.31 77.94	7.26 7.09	14.42 14.23	—	3209 (NH), 1621 (C=N)	9.7 (br, 1H, NH), 7.5–7.0 (m, 10H, arom.), 3.4 (s, 2H, CH_2), 1.9–1.2 (m, 8 H, cyclic CH_2)
10b	211 dioxan	79	$C_{20}H_{23}N_3$ (305.44)	78.65 78.26	7.59 7.41	13.76 13.56	—	3267 (NH), 1617 (C=N)	11.1(br, 1H, NH), 7.5–7.0 (m,10H, arom.), 3.4 (s, 2H, CH_2), 1.9–1.2 (m, 10 H, cyclic CH_2)
10c	216 ethanol	67	$C_{21}H_{25}N_3$ (319.46)	78.95 78.56	7.89 7.73	13.15 13.01	—	3211 (NH), 1609 (C=N)	9.9 (br, 1H, NH), 7.5–7.0 (m, 10H, arom.), 3.4 (s, 2H, CH_2), 1.9–1.2 (m, 12 H, cyclic CH_2)
11a	249 DMF	61	$C_{26}H_{28}N_3O_2PS$ (477.56)	65.39 65.03	5.91 5.78	8.79 8.60	6.71 6.59	3286 (NH), 1649 (C—O), 661 (P=S)	9.9 (br, H, NH); 7.5–6.9 (m, 14H, arom.); 3.9 (s, 3H, OCH_3); 3.5 (s, 2H, CH_2); 1.9–1.2 (m, 8 H, cyclic CH_2)
11b	221 ethanol	59	$C_{27}H_{30}N_3O_2PS$ (491.59)	65.97 65.60	6.15 6.00	8.54 8.41	6.52 6.38	3213 (NH), 1646 (C=O), 669 (P=S)	10.9 (br, H, NH); 7.5–6.9 (m, 14H, arom.); 3.9 (s, 3H, OCH_3); 3.5 (s, 2H, CH_2); 1.9–1.2 (m, 10 H, cyclic CH_2)
11c	272 ethanol	68	$C_{28}H_{32}N_3O_2PS$ (505.61)	66.51 66.15	6.38 6.21	8.33 8.16	6.34 6.17	3287 (NH), 1652 (C=O), 653 (P=S)	10.3 (br, H, NH); 7.5–6.9 (m, 14H, arom.); 3.9 (s, 3H, OCH_3); 3.5 (s, 2H, CH_2); 1.9–1.1 (m, 12 H, cyclic CH_2)
12a	231 ethanol	65	$C_{26}H_{28}N_3OPS_2$ (493.62)	63.26 62.85	5.71 5.56	8.51 8.37	12.99 12.70	1410 (SH), 658 (P=S)	7.3–6.9 (m, 14H, arom.), 4.5 (s, 1H, SH); 3.9 (s, 3H, OCH_3); 3.5 (s, 2H, CH_2), 1.9–1.2 (m, 8 H, cyclic CH_2)
12b	227 dioxan	79	$C_{27}H_{30}N_3OPS_2$ (507.65)	63.88 63.47	5.95 5.81	8.27s 8.09	12.63 12.40	1429(SH), 670 (P=S)	7.5–7.0 (m, 14H, arom.), 4.3 (s, 1H, SH), 3.9 (s, 3H, OCH_3), 3.6 (S, 2H, CH_2); 1.9–1.2 (m, 10 H, cyclic CH_2)
12c	242 ethanol	67	$C_{28}H_{32}N_3OPS_2$ (521.67)	64.46 64.12	6.18 6.03	8.05 7.90	12.29 12.09	1420 (SH), 673 (P=S)	7.5–7.0 (m, 14H, arom.), 4.3 (s, 1H, SH), 3.9 (s, 3H, OCH_3), 3.5 (S, 2H, CH_2); 1.9–1.1 (m, 12 H, cyclic CH_2)

[a]Uncorrected.
[b]Satisfactory microanalysis obtained C, −0.47; H, −0.25; N, −0.39; S, −0.35.
[c]Measured by Nicolet FT-IR 710 spectrophotometer.
[d]Measured by ^{1}HNMR LA 400 MHz (Jeol) Assiut University.

(cyclohexane, and cycloheptane)-1,4′-perhydro[1,3,2]diazaphosphole]-2′-disulfi-des (**5a–c**). As to the mechanism for the formation P-heterocycles **5a-c**, it is suggested that a nucleophilic attack on LR to give the intermediate, which at elevated temperature looses H_2S to give, compounds **5a-c** (*cf.* Scheme 2). The structures of these products were based on spectroscopic data and elemental analysis (*cf.* Table I).

Compounds **6b**,**c** were synthesized by Desai,[15] and this synthesis can be carried out in another route via the diazotization of compounds **4b**,**c**. Also, the diazotization of compound **4a** gave 1-anilinocyclopentanemethanol (**6a**). Compounds **6a–c** were then submitted to the reaction with LR in anhydrous benzene to give 2′-(4-methoxyphenyl)-3′-phenylspiro[cyclopentane (cyclohexane, and cycloheptane)-1,4′-perhydro[1,3,2]oxaazaphosphole]-2′-disulfides (**7a–c**) (*cf.* Scheme 2). The structures of the new products have been elucidated via analytical results and spectroscopic data (*cf.* Table I).

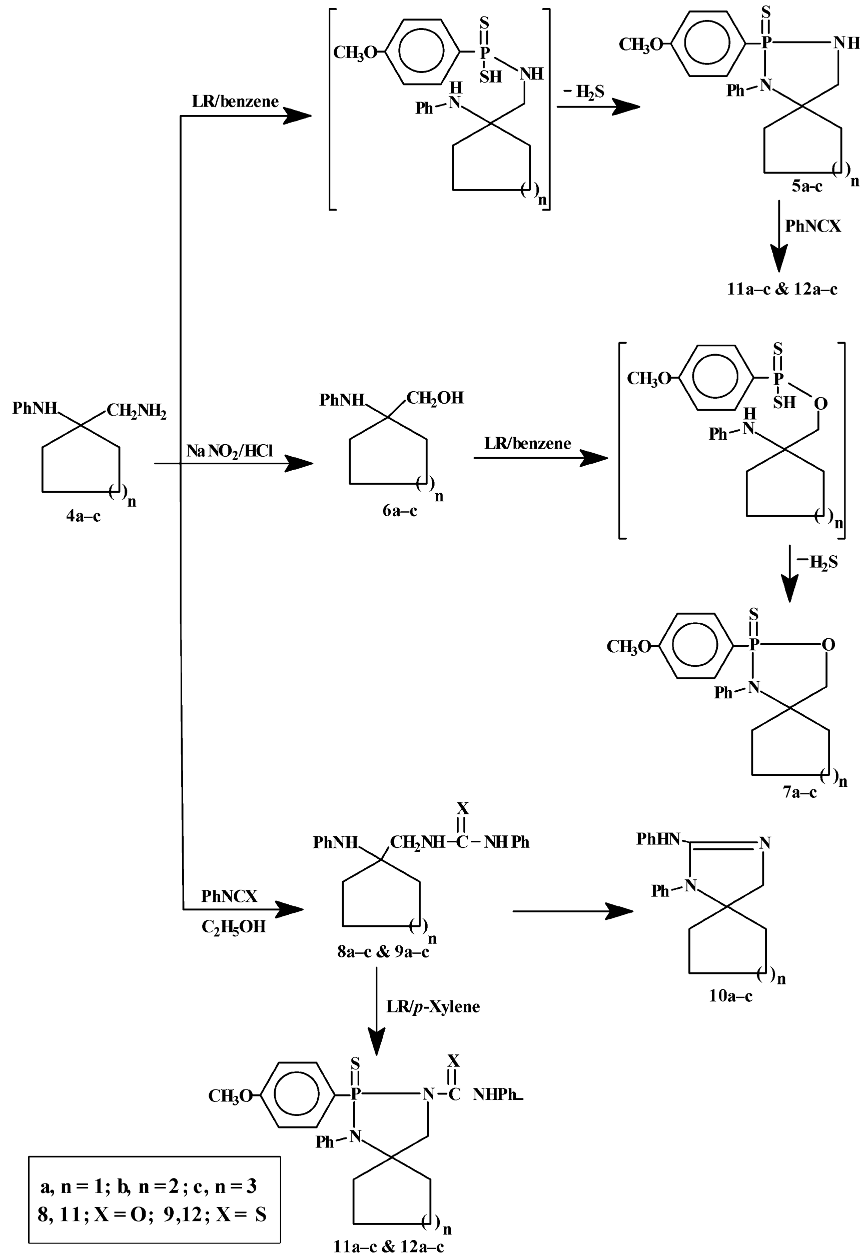
Moreover, the addition of compounds **4a–c** to phenyl isocyanate and/or phenyl isothiocyanate in ethanol at 26°C afforded the opened products **8a–c** and **9a-c**, respectively, via an addition of the amino groups of compounds **4a–c** to the isocyanate or isothiocyanate (*cf.* Scheme 2). The cyclized products **10a–c** were obtained by refluxing compounds **8a–c** and/or **9a–c** in N,N-dimethylaniline. The IR and ^{1}H NMR spectra of these compounds confirm the proposed structures (*cf.* Table I).

The reaction of compounds **8a–c** and/or **9a–c** with LR in refluxing *p*-xyl-ene afforded 2′(4-methoxyphenyl)-3′-phenylspiro[cyclopentane (cyclohexane and cycloheptane)-1,4′-perhydro[1,3,2]diazaphosphole]-1-ylphenylaminomethane-1-ones (**11a–c**) and 2′(4-methoxyphenyl)-3′-phenylspiro[cyclopentane (cyclohexane and cycloheptane)-1,4′-perhydro[1,3,2]diazaphosphole]-1-ylphenylaminometha-ne-1-thiones (**12a–c**), respectively. The same products **11a–c** and **12a–c** were also obtained through the reaction of compounds **5a–c** with phenyl isocyanate and/or phenyl isothiocyanate in boiling benzene. The structure of compounds **11a–c** and **12a–c** were confirmed on the basis of their elemental and spectral analyses (*cf.* Table I).

EXPERIMENTAL

The Synthesis of 2′-(4-Methoxyphenyl)-4′-phenylspiro [cyclopentane (Cyclohexane and Cycloheptane)-1,4′-Perhydro[1,3,2]diazaphosphole]-2′,5′-disulfides (3a–c): General Procedure

2,4-bis-(4-methoxyphenyl)-1,3,2,4-dithiaphosphetane-2,4-disulphide (LR, 1) (2.02 g; 0.005 mole) and 1-phenylamino-1-cyanocyclopentane


LR/benzene
CH3O
S
P
SH
NH
H
Ph−N
( )n
− H2S
NH
Ph−N
5a-c
PhNCX
11a–c & 12a–c
PhNH
CH2NH2
4a–c
Na NO2/HCl
CH2OH
6a–c
O
−H2S
7a–c
X
CH2NH−C−NH Ph
PhHN
N
10a–c
PhNCX
C2H5OH
8a–c & 9a–c
LR/p-Xylene
N−C NHPh
11a–c & 12a–c
a, n = 1; b, n = 2 ; c, n = 3
8, 11 ; X = O; 9,12 ; X = S

SCHEME 2

(cyclohexane, and/or cycloheptane) (**2a–c**) (0.01 mol) were dissolved in acetonitrile (80 mL). The reaction mixture was refluxed for 6 h, concentrated and cooled the precipitate was filtered off, dried, and recrystallized from the suitable solvent to give compounds **3a–c**.

The Synthesis of 1-Anilinocyclopentane(cyclohexane, and Cycoheptane)methyl-amine l4a–c: General Procedure

To a solution of the proper nitrile **2a–c** (1.0 g) in absolute ethanol (10 mL) was added 1.5 g of clean sodium. When all the sodium reacted (10–15 min.), the reaction mixture was cooled to about 20°C, and 15 mL of conc HCl was added. The reaction mixture was concentrated and cooled and 20 mL of 40% NaOH was added dropwise with shaking to the reaction mixture. The formed amine was extracted with chloroform and purified by distillation.

The Synthesis of 1-Anilinocyclopentane (Cyclohexane and Cycloheptane)methanol 6a–c: General Procedure

To a stirred ice-cooled solution of the proper amine (**4a–c**) (0.01 mole) in 8 mL of conc HCl was added portionwise (0.83 g, 0.012 mole) of $NaNO_2$. The mixture was kept at r.t. for 30 min and then poured onto crushed ice. The obtained solution was neutralized with NH_4OH, and the separated oil was extracted with chloroform and purified by distillation to give compounds **6a–c**.

The Synthesis of 2′-(4-Methoxyphenyl)-3′-phenylspiro [cyclopentane(cyclohexane and cycloheptane)-1,4′-Perhydro-[1,3,2]diazaphosphole]-2′-disulfides (5a-c) and 2′-(4-methoxyphenyl)-3′-phe-nylspiro[cyclopentane(cyclohexane and Cycloheptane)-1,4′-perhydro[1,3,2]oxaazaphosphole]-2′-disulfides (7a–c): General Procedure

A mixture of the proper amine (**4a–c**) (0.01 mole) and/or the proper hydroxylderivative (**6a–c**) (0.01 mole) and 2,4-bis-(4-methoxyphenyl)-1,3,2,4-dithiaphosphetane-2,4-disulphide (LR, **1**) (2.02 g, 0.005 mole) in dry benzene (80 mL) was refluxed for 5 h, concentrated, and cooled. The formed precipitate was filtered off, dried, and recrystallized from the suitable solvent to give compounds **5a–c** and **7a–c**, respectively.

The Synthesis of Compounds 8a–c and 9a–c: General Procedure

To a stirred solution of the proper amine **4a–c** (0.01 mole) in 30 mL of absolute ethanol was added phenyl isocyanate (1.19 g, 0.01 mole) or phenyl isothiocyanate (1.35 g, 0.01 mole). The reaction mixture was stirred at r.t. for 1 hour. The formed precipitate was filtered off, dried, and recrystallized from the suitable solvent to give compounds **8a–c** and **9a–c**, respectively.

The Synthesis of 3′-Phenyl-2′-phenylaminospiro [cyclopentane(cyclohexane and Cycloheptane)-1,4′-(3′,5′-dihydroimidazoles)] (10a–c): General Procedure

Compound **8a–c** and/or **9a–c** (0.01 mol) was refluxed in N,N-dimethylaniline (10 mL) for 12 h. The solvent was concentrated, cooled, and filtered off. The solid product was recrystallized from the suitable solvent (*cf.* Table I).

The Synthesis of 2′-(4-Methoxyphenyl)-3′-phenylspiro[cyclopentane(cyclohexane and Cycloheptane)-1,4′-Perhydro[1,3,2]diazaphosphole]-1-ylphenylamino-methane-1-ones (11a–c) or –1-thiones (12a–c): General Procedure

2,4-bis-(4-methoxyphenyl)-1,3,2,4-dithiaphosphetane-2,4-disulphide (LR, 1) (2.02 g; 0.005 mole) and the proper compound **8a–c** and/or **9a–c** (0.01 mol) were dissolved in p-xylene (60 mL). The reaction mixture was refluxed for 6 h, concentrated, and cooled; the precipitate was filtered off, dried, and recrystallized from the proper solvent to give compounds **11a–c** and **12a–c**, respectively.

The Alternate Synthesis of Compounds 11a–c and 12a–c: General Procedure

Compound **5a–c** (0.01 mol), phenyl isocyanate and/or pheny isothiocyanate (0.01 mol) and dry benzene (70 mL) were refluxed together for 3 h when colorless crystals separated out, which were filtered and washed with warm benzene to afford compounds **11a–c** and **12a–c**.

REFERENCES

[1] J. C. Thomas, *J. Org. Chem.,* **67,** 6461 (2002).
[2] M. Ori and T. Nishio, *Heterocycles,* **52,** 111 (2000).
[3] T. Nishio and H. Sekiguchi, *Tetrahedron,* **55,** 5017 (1999).
[4] T. Nishio, *J. Org. Chem.,* **62,** 1106 (1997).
[5] A. M. Polozov, S. E. Cremer, P. E. Fanwick, and E. Phillip, *Can. J. Chem.,* **77,** 1274 (1999).

[6] B. Mohamed, E. M. Lotfi, and Z. Hedi, *Phosphorus, Sulfur, and Silicon,* **157,** 145 (2000).
[7] L. S. Boulous and H. A. Abd El-Malek, *Heteroatom Chem.,* **10,** 488 (1999).
[8] A. A. El-Barbary and S.-O. Lawesson, *Tetrahedron,* **37,** 2641 (1981).
[9] C. S. Sarma and J. C. S. Kataky, *Indian J. Chem.,* **38B,** 464 (1999).
[10] H. M. Moustafa, *Phosphorus, Sulfur, and Silicon,* **148,** 131 (1999).
[11] H. M. Moustafa, *Phosphorus, Sulfur, and Silicon,* **164**, 11 (2000).
[12] A. B. A. G. Ghattas, O. A. Abd Allah, and H. M. Moustafa, *Phosphorus, Sulfur, and Silicon,* **157,** 1 (2000).
[13] H. M. Moustafa, *Phosphorus, Sulfur, and Silicon,* **178,** 1397 (2003).
[14] M. S. Chande and S. K. Balel, *Indian Journal of Chemistry*, **35B,** 377 (1996).
[15] P. R. Desai, R. D. Desai, G. S. Saharia, and B. R. Sharma, *J. Indian Chem. Soc.,* **40,** 525 (1963).
[16] B. S. Pedersen and S.-O. Lawesson, *Tetrahedron,* **35,** 2433 (1979).