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

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

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*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]oxaazaphosphole] 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 carbonyl compounds.<sup>1–5</sup> The nucleophiles attack LR at the phosphorus atom and form phosphorus heterocycles.<sup>6–9</sup> In view of the latest development and also in continuation of our study of phosphorus hetero-cycles,<sup>10–13</sup> 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.

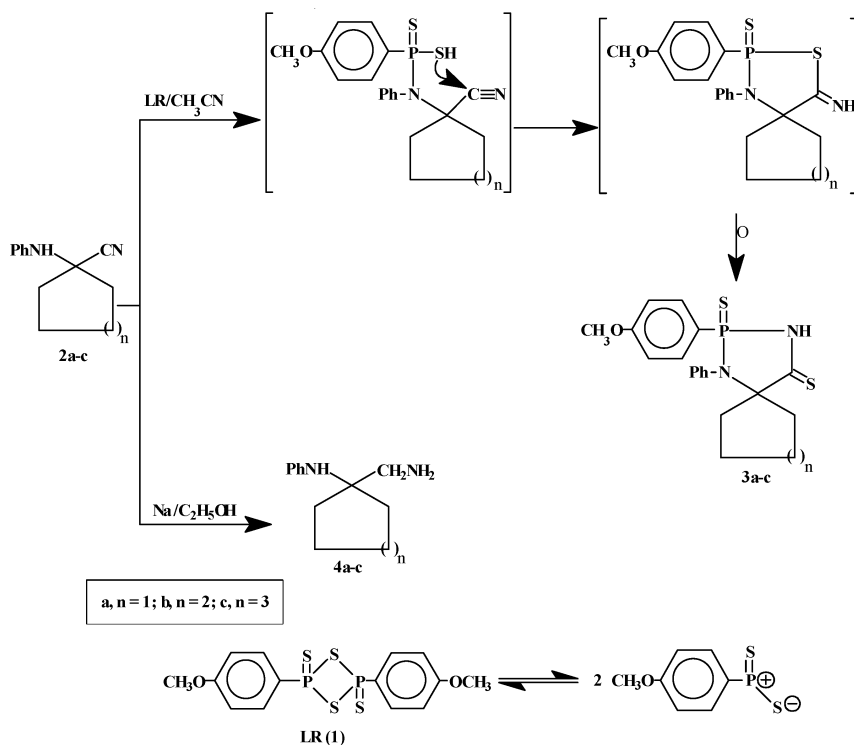
## RESULTS AND DISCUSSION

The reaction of 1-phenylamino-1-cyanocyclopentane(cyclohexane, and cyclo-heptane)<sup>14</sup> (**2a-c**) with LR (**1**) in boiling acetonitrile afforded 2'-(4-methoxy-phenyl)-4'-phenylspiro[cyclopentane (cyclohexane, and

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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,<sup>16</sup> 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  $\text{cm}^{-1}$  for P=S, respectively.  $^1\text{H}$  NMR spectra of compounds **3a-c** exhibited a singlet at 3.9 ppm corresponding to  $\text{OCH}_3$  (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  $\text{NH}_2$  groups ( $3163\text{--}3412\text{ cm}^{-1}$ ). The  $^1\text{H}$  NMR ( $\text{CDCl}_3$ ) 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  $80^\circ\text{C}$  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 Calcd./found				IR (Cm <sup>-1</sup> )	<sup>1</sup> HNMR $\delta$ (ppm)
				C	H	N	S		
3a	160 ethanol	86	C <sub>19</sub> H <sub>21</sub> N <sub>2</sub> OPS <sub>2</sub> (388.48)	58.74 58.50	5.45 5.27	7.21 7.03	16.50 16.23	3212 (NH), 1246 (C—O), 651(P=S)	11.2 (br, 1H, NH), 7.8–6.9 (m, 9H, arom.); 3.9 (s, 3H, OCH <sub>3</sub> ), 1.9–1.4 (m, 8H, cyclic CH <sub>2</sub> )
3b	170 ethanol	83	C <sub>20</sub> H <sub>23</sub> N <sub>2</sub> OPS <sub>2</sub> (402.51)	59.68 59.30	5.75 5.58	6.95 6.73	15.93 15.72	3266 (NH), 1253 (C—O), 642 (P=S)	12.9 (br, 1H, NH), 7.8–6.7 (m, 9H, arom.); 3.9 (s, 3H, OCH <sub>3</sub> ), 1.9–1.2 (m, 10H, cyclic CH <sub>2</sub> )
3c	191 ethanol	79	C <sub>21</sub> H <sub>25</sub> N <sub>2</sub> OPS <sub>2</sub> (416.53)	60.55 60.21	6.05 5.87	6.72 6.58	15.93 15.73	3259 (NH), 1239 (C—O), 659 (P=S)	11.9 (br, 1H, NH), 7.8–7.0 (m, 9H, arom.); 3.9 (s, 3H, OCH <sub>3</sub> ), 1.9–1.2 (m, 12 H, cyclic CH <sub>2</sub> )
4a	Liquid	77	C <sub>12</sub> H <sub>18</sub> N <sub>2</sub> (190.29)	75.74 75.27	9.53 9.34	14.72 14.39	—	3359, 3261, 3203 (NH <sub>2</sub> + NH)	11.1 (br, 1H, NH), 7.5–6.9 (m, 5H, arom.); 4.3 (br, 2H, NH <sub>2</sub> ); 2.8 (s, 2H, CH <sub>2</sub> ), 1.9–1.2 (m, 8 H, cyclic CH <sub>2</sub> )
4b	Liquid	79	C <sub>13</sub> H <sub>20</sub> N <sub>2</sub> (204.31)	76.42 76.01	9.86 9.61	13.71 13.49	—	3412, 3336, 3230 (NH <sub>2</sub> + NH)	13.0 (br, 1H, NH), 7.5–6.9 (m, 5H, arom.); 4.9 (br, 2H, NH <sub>2</sub> ); 2.8 (s, 2H, CH <sub>2</sub> ), 1.9–1.1 (m, 10 H, cyclic CH <sub>2</sub> )
4c	Liquid	71	C <sub>14</sub> H <sub>22</sub> N <sub>2</sub> (218.34)	77.01 76.60	10.15 9.97	12.82 12.51	—	3389, 3298, 3163 (NH <sub>2</sub> + NH)	11.8 (br, 1H, NH), 7.5–6.9 (m, 5H, arom.); 4.4 (br, 2H, NH <sub>2</sub> ); 2.8 (s, 2H, CH <sub>2</sub> ), 1.9–1.0 (m, 12 H, cyclic CH <sub>2</sub> )
5a	216 ethanol	61	C <sub>19</sub> H <sub>23</sub> N <sub>2</sub> OPS (358.44)	63.66 63.31	6.46 6.29	7.81 7.57	8.94 8.74	3286 (NH), 661 (P=S)	13.0 (br, 1H, NH), 7.5–6.8 (m, 9 H, arom.); 3.9 (s, 3H, OCH <sub>3</sub> ), 2.9 (s, 2H, CH <sub>2</sub> ), 1.9–1.3 (m, 8 H, cyclic CH <sub>2</sub> )
5b	233 ethanol	59	C <sub>20</sub> H <sub>25</sub> N <sub>2</sub> OPS (372.46)	64.49 64.07	6.76 6.61	7.52 7.33	8.60 8.43	3299 (NH), 703 (P=S)	10.6 (br, 1H, NH), 7.5–6.8 (m, 9 H, arom.); 3.9 (s, 3H, OCH <sub>3</sub> ), 2.9 (s, 2H, CH <sub>2</sub> ), 1.9–1.2 (m, 10 H, cyclic CH <sub>2</sub> )
5c	240 ethanol	68	C <sub>21</sub> H <sub>27</sub> N <sub>2</sub> OPS (386.49)	65.26 64.89	7.04 6.90	7.24 7.05	8.29 8.08	3271 (NH), 655 (P=S)	10.6 (br, 1H, NH); 7.5–6.9 (m, 9H, arom.); 3.9 (s, 3H, OCH <sub>3</sub> ), 2.9 (s, 2H, CH <sub>2</sub> ), 1.9–1.1 (m, 12 H, cyclic CH <sub>2</sub> )

(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 Calcd./Found				IR (Cm <sup>-1</sup> )	<sup>1</sup> HNMR $\delta$ (ppm)
					C	H	N	S		
6a		Liquid	65	C <sub>12</sub> H <sub>17</sub> 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 <sub>2</sub> ), 1.9–1.2 (m, 8 H, cyclic CH <sub>2</sub> )
7a	163	ethanol	61	C <sub>19</sub> H <sub>20</sub> NO <sub>2</sub> PS (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 <sub>2</sub> ), 3.9 (s, 3H, OCH <sub>3</sub> ), 1.9–1.3 (m, 8 H, cyclic CH <sub>2</sub> )
7b	198	ethanol	59	C <sub>20</sub> H <sub>22</sub> NO <sub>2</sub> PS (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 <sub>2</sub> ), 3.9 (s, 3H, OCH <sub>3</sub> ), 1.9–1.2 (m, 10 H, cyclic CH <sub>2</sub> )
7c	186	ethanol	68	C <sub>21</sub> H <sub>24</sub> NO <sub>2</sub> PS (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 <sub>2</sub> ), 3.9 (s, 3H, OCH <sub>3</sub> ), 1.9–1.2 (m, 12 H, cyclic CH <sub>2</sub> )
8a	199	ethanol	65	C <sub>19</sub> H <sub>23</sub> N <sub>3</sub> O (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 <sub>2</sub> ), 1.9–1.2 (m, 12 H, cyclic CH <sub>2</sub> )
8b	186	dioxan	60	C <sub>20</sub> H <sub>25</sub> N <sub>3</sub> O (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 <sub>2</sub> ), 1.9–1.1 (m, 10 H, cyclic CH <sub>2</sub> )
8c	173	ethanol	67	C <sub>21</sub> H <sub>27</sub> N <sub>3</sub> O (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 <sub>2</sub> ), 1.9–1.2 (m, 12 H, cyclic CH <sub>2</sub> )
9a	138	DMF	61	C <sub>19</sub> H <sub>23</sub> N <sub>3</sub> S (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 <sub>2</sub> ), 1.9–1.2 (m, 8 H, cyclic CH <sub>2</sub> )
9b	132	ethanol	59	C <sub>20</sub> H <sub>25</sub> N <sub>3</sub> S (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 <sub>2</sub> ), 1.9–1.2 (m, 10 H, cyclic CH <sub>2</sub> )

<b>9c</b>	141	68	C <sub>31</sub> H <sub>27</sub> N <sub>3</sub> S (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 <sub>2</sub> ), 1.9–1.2 (m, 12 H, cyclic CH <sub>2</sub> )
<b>10a</b>	203	65	C <sub>19</sub> H <sub>21</sub> N <sub>3</sub> (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 <sub>2</sub> ), 1.9–1.2 (m, 8 H, cyclic CH <sub>2</sub> )
<b>10b</b>	211	79	C <sub>20</sub> H <sub>23</sub> N <sub>3</sub> (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 <sub>2</sub> ), 1.9–1.2 (m, 10 H, cyclic CH <sub>2</sub> )
<b>10c</b>	216	67	C <sub>21</sub> H <sub>25</sub> N <sub>3</sub> (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 <sub>2</sub> ), 1.9–1.2 (m, 12 H, cyclic CH <sub>2</sub> )
<b>11a</b>	249	61	C <sub>26</sub> H <sub>28</sub> N <sub>3</sub> O <sub>3</sub> PS (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 <sub>3</sub> ); 3.5 (s, 2H, CH <sub>2</sub> ); 1.9–1.2 (m, 8 H, cyclic CH <sub>2</sub> )
<b>11b</b>	221	59	C <sub>27</sub> H <sub>30</sub> N <sub>3</sub> O <sub>3</sub> PS (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 <sub>3</sub> ); 3.5 (s, 2H, CH <sub>2</sub> ); 1.9–1.2 (m, 10 H, cyclic CH <sub>2</sub> )
<b>11c</b>	272	68	C <sub>28</sub> H <sub>32</sub> N <sub>3</sub> O <sub>3</sub> PS (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 <sub>3</sub> ); 3.5 (s, 2H, CH <sub>2</sub> ); 1.9–1.1 (m, 12 H, cyclic CH <sub>2</sub> )
<b>12a</b>	231	65	C <sub>26</sub> H <sub>28</sub> N <sub>3</sub> OPS <sub>2</sub> (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 <sub>3</sub> ); 3.5 (s, 2H, CH <sub>2</sub> ); 1.9–1.2 (m, 8 H, cyclic CH <sub>2</sub> )
<b>12b</b>	227	79	C <sub>27</sub> H <sub>30</sub> N <sub>3</sub> OPS <sub>2</sub> (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 <sub>3</sub> ); 3.6 (s, 2H, CH <sub>2</sub> ); 1.9–1.2 (m, 10 H, cyclic CH <sub>2</sub> )
<b>12c</b>	242	67	C <sub>28</sub> H <sub>32</sub> N <sub>3</sub> OPS <sub>2</sub> (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 <sub>3</sub> ); 3.5 (s, 2H, CH <sub>2</sub> ); 1.9–1.1 (m, 12 H, cyclic CH <sub>2</sub> )

<sup>a</sup>Uncorrected.<sup>b</sup>Satisfactory microanalysis obtained C, –0.47; H, –0.25; N, –0.39; S, –0.35.<sup>c</sup>Measured by Nicolet FT-IR 710 spectrophotometer.<sup>d</sup>Measured by <sup>1</sup>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<sub>2</sub>S 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,<sup>15</sup> 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).

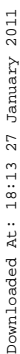
Moreover, the addition of compounds **4a-c** to phenyl isocyanate and/or phenyl isothiocyanate in ethanol at 26°C afforded the opened products **8a-c** and **9a-c**, respectively, via an addition of the amino groups of compounds **4a-c** to the isocyanate or isothiocyanate (*cf.* Scheme 2). The cyclized products **10a-c** were obtained by refluxing compounds **8a-c** and/or **9a-c** in N,N-dimethylaniline. The IR and <sup>1</sup>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-ylphenylaminomethane-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



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(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 4a–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<sub>2</sub>. The mixture was kept at r.t. for 30 min and then poured onto crushed ice. The obtained solution was neutralized with NH<sub>4</sub>OH, 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 phenyl 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**.

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