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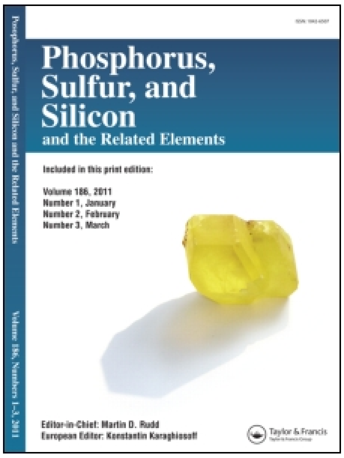

Phosphorus, Sulfur, and Silicon and the Related Elements
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1,2-DIHYDRO-2-(4-METHOXYPHENYL)-2-SULPHIDE-1,3,2-DIAZAPHOSPHORIN-4(3H)-THIONES FROM 3-AMINOPROPENENITRILE DERIVATIVES AND LA WESSON'S REAGENT

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To cite this Article Testa, M. G. , Perrini, G. , Chiacchio, U. and Corsaro, A.(1994) '1,2-DIHYDRO-2-(4-METHOXYPHENYL)-2-SULPHIDE-1,3,2-DIAZAPHOSPHORIN-4(3H)-THIONES FROM 3-AMINOPROPENENITRILE DERIVATIVES AND LA WESSON'S REAGENT', Phosphorus, Sulfur, and Silicon and the Related Elements, 86: 1, 75 — 80

To link to this Article: DOI: 10.1080/10426509408018390

URL: http://dx.doi.org/10.1080/10426509408018390

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Phosphorus, Sulfur, and Silicon, 1994, Vol. 86, pp. 75–80
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1,2-DIHYDRO-2-(4-METHOXYPHENYL)-2-SULPHIDE-1,3,2-DIAZAPHOSPHORIN-4(3H)-THIONES FROM 3-AMINOPROPENENITRILE DERIVATIVES AND LAWESSON'S REAGENT†

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(Received November 15, 1993; in final form January 4, 1994)

The title compounds are obtained from structures incorporating the 3-aminopropenenitrile moiety by treatment with the Lawesson's reagent in refluxing toluene.

Key words: 3-Aminopropenenitrile; Lawesson's reagent; 1,3,2-diazaphosphorines; 1,3,2-thiazaphosphorines.

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Structures containing the 3-aminopropenenitrile moiety have found a broad application as building blocks for the synthesis of a variety of heterocycles.[1,2] The sole known reactions which lead to phosphorus cycles are those of some open chain derivatives with phosphorus halides affording 1,3,2-diazaphosphorines.[3] In these reactions the enaminonitrile system shows a rare reactivity acting as double 1,5-nucleophilic reagent like 4-amino-1-azabutadienes.[4]

More common are the heterocyclization reactions in which 3-aminopropenenitriles undergo electrophilic-nucleophilic processes to give products incorporating their NCCC framework.[1] This latter type of process has been found to be involved also in their reactions with the Lawesson's reagent which, performed under the usual conditions,[5] afford 1,3,2-diazaphosphorin-4-thiones **5**. Unisolable intermediate compounds of these reactions leading to **5** are the 6-imino-1,3,2-thiazaphosphorines **4**, for the formation of which a rationalization analogous to that formulated by Lawesson for the formation of 1,3,2-oxazaphosphorines from 3-oxonitriles[6] is proposed (Scheme).

The dipolar species **2**, which is in equilibrium with the Lawesson's reagent,[5] attacks the enaminic nitrogen atom to give thiophosphoric derivative **3**. This undergoes a cyclization involving an exo-addition of the sulphydryl group to the nitrile function to give **4**, which rapidly rearranges to **5**.

The intermediacy of iminothiazaphosphorines **4** finds a confirmation in the isolation of the thionation product of **4m**, the 1,3,2-thiazaphosphorin-4-thione **6m**, from the reaction mixture of heterocyclic enaminonitrile **1m**. It is surprising, however, that only thiazaphosphorine **4m** undergoes the thionation process by Lawesson's reagent in preference to the Dimroth type rearrangement, which latter proved to be faster in all the other cases.

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† Dedicated to Prof. G. Purrello on the occasion of his 70th birthday.

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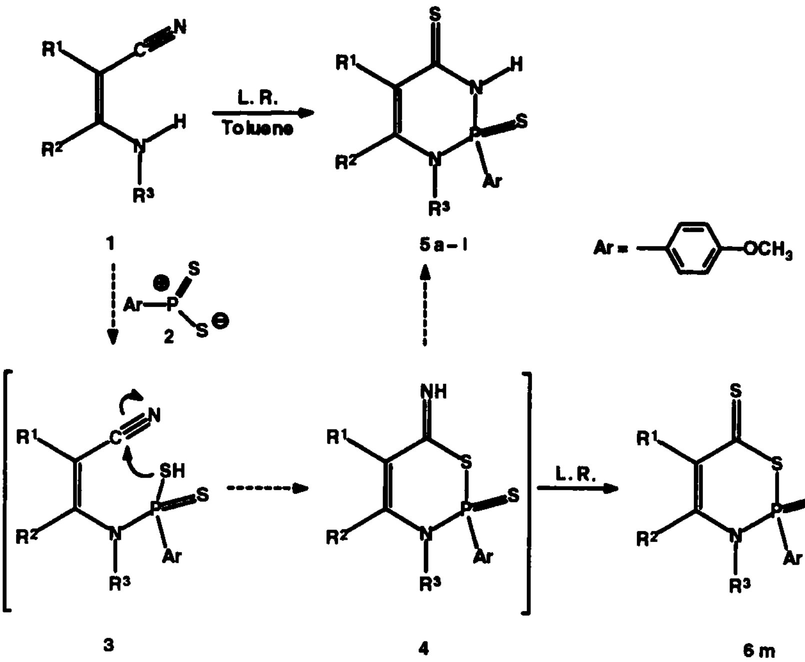


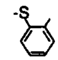
1,5,6	R¹	R²	R³
a	H	C_6H_5	H
b	H	C_6H_5	C_6H_5
c	H	$-(CH_2)_3-$	
d	H	$-(CH_2)_4-$	
e	H	$-(CH_2)_5-$	
f	$-(CH)_4=$		H
g			H
h	$-(CH_2)_3-$		H
i	$-CH_2-CH(CH_3)-N(CO_2C_2H_5)-$		H
l	$-CH_2-CH(CH_3)-S-$		H
m	$-CH_2-CH(CH_3)-O-$		H

SCHEME

The structures of the new diazaphosphorines **5a–l** and thiazaphosphorine **6m** were established by means of their spectroscopic data and microanalyses. In the I.R. spectra of **5a–l** the presence of associated NH groups is shown by a broad band in the region 3020–3070 cm^{-1} which is accompanied in the cases of **5a, f–l** by a sharper band in the region 3190–3260 cm^{-1} (Table I). No band was observed in the region 2600–2550 cm^{-1} for the SH moiety suggesting the thionic form for all the isolated compounds. The thionic form was confirmed also by the ^{13}C NMR

TABLE I

Yields, physical and IR data of **5a–l** and **6m**

Compd. No	R^1	R^2	R^3	Yields[a] (%)	M.p. (°C)	Molecular Formula	M^+	ν_{max} (cm^{-1})
5a	H	C_6H_5	H	62	112-114	$C_{16}H_{15}N_2OPS_2$	346	3191, 3020[c]
5b	H	C_6H_5	C_6H_5	34	173-174	$C_{22}H_{19}N_2OPS_2$	422	3040[c]
5c	H	$-(CH_2)_3-$		40	210	$C_{13}H_{15}N_2OPS_2$	310	3050[c]
5d	H	$-(CH_2)_4-$		50	190	$C_{14}H17N_2OPS_2$	324	3040[c]
5e	H	$-(CH_2)_5-$		46	162	$C_{15}H_{19}N_2OPS_2$	338	3040[c]
5f	$-(CH)_4=$		H	42	165-167[d]	$C_{14}H_{13}N_2OPS_2$	320	3270,3150[c]
5g			H	44	168-170	$C_{16}H_{13}N_2OPS_3$	376	3260,3050[c]
5h	$-(CH_2)_3-$		H	54	225-227	$C_{13}H_{15}N_2OPS_3$	310	3180, 3070[c]
5i	$-CH_2-CH(CH_3)-N-(CO_2C_2H_5)-$		H	48	183-185	$C_{18}H_{20}N_3O_3PS_2$	397	3210, 3060[c]
5l	$-CH_2-CH(CH_3)-S-$		H	55	188-190	$C_{13}H_{15}N_2OPS_3$	342	3260, 3060[c]
6m	$-CH_2-CH(CH_3)-O-$		H	31	151-153	$C_{13}H_{14}NO_2PS_3$	343	3200

[a] Based on the starting enaminonitrile. [b] Microanalyses were in satisfactory agreement with the calculated values (C,H,N ±0.4). [c] Broad band. [d] Different from Literature[12] m.p. 95°C.

spectra (Table III) which contain signals in the region 184.00–194.28 ppm for the thiocarbonyl carbons in line with the values found for the corresponding oxazaphosphorine analogues.[6] In the ^{1}H-NMR spectra recorded in deuterochloroform solutions (Table II) the NH protons show very broad signals; in some cases they are covered by aromatic multiplets. The resonances of the NH protons appear well apart and shift to lower fields (9.44–11.88 ppm) when the spectra were recorded in deuterated dimethylsulphoxide. In all cases the resonances appear as doublets because of the P—H coupling. In the EI-mass spectra all isolated compounds show very stable molecular ions. The primary fragmentation process is represented by the loss of the sulphydryl radical. The peak due to the elimination of the methyl radical from molecular ions of β-heterocyclic derivatives **5i, l** and **6m** is of equal intensity to that of the loss of the sulphydryl radical.

The Lawesson's reagent reacts with structurally diferent enaminonitriles such as open chain and bridged derivatives of 3-aminopropenenitriles (**1a, b** and **1c–e**, respectively), aromatic and not aromatic cyclic enaminonitriles (**1f, g** and **1h–m**, respectively).

The formation of the corresponding 1,2-dithiolo-3-thiones, which were expected as possible by-products assuming for **1a–m** a similar behavior to that of 3-oxonitriles,[6] was not observed even when the reactions were carried out in the presence of two equivalents of sulphur.

The yields were not optimized, but apparently do not depend on the geometry of the starting enaminonitrile probably because of the equilibrium of the two geometrical isomers under the reaction conditions. The use of basic solvents such as pyridine instead of aromatic hydrocarbons, however, appears to suppress the production of diazaphosphorine **5a** in the case of **1a** to favour the formation of

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other products among them the searched dithiolothione. This result has prompted us to study the solvent effect on the reaction of β-enaminonitriles with the Lawesson's reagent. These results will be reported later.

TABLE II

^{1}H NMR data of **5a–e** and **6m**

Comp. No	δ_H ($CDCl_3$)[a]
5a	3.83 (s, 3H, methoxyl H), 6.16 (s, 1H, methinic H), 7.40-7.16 (m, 2H, aromatic H), 7.44-7.69 (m, 5H, aromatic H) 7.80-7.92 (m, 2H, aromatic H) [9.88 (d, 1H, J_{PH} 12.5 Hz, phosphaminic H), 11.15 (d, 1H, J_{PH} 13.9 Hz, thioamidic H)]
5b	3.83 (s, 3H, methoxyl H), 6.57 (s, 1H, methinic H), 6.90-6.96 (m, 2H, aromatic H), 7.19 (brs, 5H, aromatic H), 7.20-7.31 (m, 5H, aromatic H), 7.85-7.95 (m, 2H, aromatic H), 8.18 (brd, 1H, thioamidic H)
5c	1.91-2.17 (m, 2H, methylene 4-H), 2.76-2.89 (m, 2H, methylene 3-H), 3.18-3.32 (m, 2H, methylene 5-H), 3.85 (s, 3H, methoxyl H), 6.02 (s, 1H, methynic H), 6.90-7.02 (m, 2H, aromatic H), 7.52 (brd, J_{PH} 12.9 Hz , phosphaminic H), 7.79-7.91 (m, 2H, aromatic H)
5d	1.62-1.85 (m, 4H, methylene 4- and 5-H), 2.37-2.55 (m, 2H, methylene 3-H), 2.95-3.70 (m, 2H, methylene 6-H), 3.92 (s, 3H, methoxyl H), 5.97 (s, 1H, methynic H), 6.95-7.12 (m, 2H, aromatic H), 7.80-8.12 (m, 2H, aromatic H)[b]
5e	1.62-1.75 (m, 6H, methylene 4, 5- and 6-H), 2.47-2.52 (m, 2H, methylene 3-H), 3.50-3.65 (m, 2H, methylene 7-H), 3.88 (s, 3H, methoxyl H), 6.03 (s, 1H, methynic H), 6.95-7.02 (m, 2H, aromatic H), 7.31 (brd, 1H, J_{PH} 13.0 Hz, phosphaminic H), 7.80-7.92 (m, 2H, aromatic H)
5f	3.82 (s, 3H, methoxyl H), 6.93-7.13 (m, 4H, aromatic H), 7.50 (t, 1H, J 6 Hz aromatic H), 7.75- 7.87 (m, 2H, aromatic H), 8.41 (d, 1H, J 8.3 Hz, aromatic H), [9.58 (d, 1H, J_{PH} 11.7 Hz, phosphaminic H), 11.88 (,d, 1H J_{PH}11.77 Hz, thioamidic H)]
5g	[3.83 (s, 3H, methoxyl H), 7.12-7.18 (m, 2H, aromatic H), 7.39-7.60 (m, 3H, aromatic H), 7.83-7.97 (m, 3H, aromatic H), 10.51 (brd, 1H, J_{PH} 12.0 Hz, phosphaminic H), 11.60 (brd, 1H, J_{PH} 14.0 Hz, thioamidic H)]
5h	[1.76-1.89 (m, 2H, cyclopentene H), 2.56-2.68 (m, 4H, cyclopentene H), 3.83 (s, 3H, methoxyl H), 7.09-7.15 (m, 2H, aromatic H); 7.72-7.83 (m, 2H, aromatic H), 10.01 (brd, J_{PH} 14.0 Hz, phosphaminic H), 10.61 (brd, 1H, J_{PH} 14.0 Hz thioamidic H)]
5i	1.32 (t, 3H, J 7.3 Hz, methyl H), 1.46 (d, 3H, J 6.7 Hz, 5-methyl H), 2.59 (dd, 1H, J 3.3 and 14.3 Hz, pyrrole 4-H), 3.31 (dd, 1H, J 9.9 and 14.3 Hz, pyrrole 4-H), 3.87 (s, 3H, methoxyl H), 4.28 (q, 2H, J 13.0 Hz, methylene-H), 4.32 (m, 1H, pyrrole 5-H); 6.83-7.05 (m, 2H, aromatic H), 7.74-8.05 (m, 2H, aromatic H)
5l	1.47 (d, 3H, J 7.1 Hz, methyl H), 3.15 (dd, 1H, J 6.0 and 16.4 Hz, thiophene 4-H); 3.47 (dd, 1H, J 9.0 and 16.4 Hz, thiophene 4-H); 3.86 (s, 3H, methoxyl H), 3.90 (m, 1H, thiophene 5-H), 7.75-7.87 (m, 2H, aromatic)
6m	1.47 (d, 3H, J 6.8 Hz, methyl H), 3.00 (dd, 1H, J 8.5 and 14.9 Hz furan 4-H), 3.55 (dd, 1H, J 8.1 and 14.9 Hz furan 4-H); 3.87 (s, 3H, methoxyl H), 3.94 (m, 1H, furan 5-H), 6.93-6.99 (m, 2H, aromatic H), 7.55-7.88 (m, 2H, aromatic H), [10.46 (brd, 1H, J_{PH} 15.2 Hz, phosphaminic H)]

[a] Values in square brakets are taken from the spectra registered in DMSO-d_6. [b] The NH signal is covered by the aromatic multiplet.

TABLE III

^{13}C NMR data of **5a–l** and **6m**

Compd. No	δ_C($CDCl_3$)[a]
5a	55.58 (methoxyl C), 106.94 (diazaphosphorine 4-C), 113.78, 114.10, 125.00, 127.22, 128.80, 131.12, 133.31, 133.61, 133.82, 133.99, 162.64 (aromatic C), 148.47 (diazaphosphorine 6-C), 191.53 (diazaphosphorine 4-C)
5b	55.46 (methoxyl C), 106.50 (diazaphosphorine 5-C), 114.05, 114.47, 116.48, 122.25, 125.00, 127.20, 128.32, 128.67, 128.83, 129.94, 133.22, 133.46, 135.10, 139.00, 163.33 (aromatic C), 148.69 (diazaphosphorine 6-C), 192.10 (diazaphosphorine 4-C)
5c	21.57, 26.66, 48.49 (pyrrolidine 4-, 3- and 5-C, respectively), 55,59 (methoxyl C), 102.45 (diazaphosphorine 5-C), 114.10, 114.43, 123.00, 125.30, 133.18, 133.65, 163.65, (aromatic C), 155.33 (diazaphosphorine 6-C), 190.15 (diazaphosphorine 4-C)
5d	18.37, 22.17, 29.24 and 41.29 (piperidine 4-, 3-, 5- and 6-C, respectively), 55.57 (methoxyl C), 107.23 (diazaphosphorine 5-C), 114.07, 114.39, 123.00, 125.10, 133.67, 164.00 (aromatic C), 152.80 (diazaphosphorine 6-C), 190.00 (diazaphosphorine 4-C)
5e	26.28, 28.78, 29.50, 35.96 and 45.92 (azacycloeptane 5-, 4-, 6-, 3- and 7-C, respectively), 55.55 (methoxyl C), 108.42 (diazaphosphorine 5-C), 113.99, 114.26, 123.22, 125.64, 133.12, 133.64, 163.66 (aromatic C), 155.76 (diazaphosphorine 6-C), 190.24 (diazaphosphorine 4-C)
5f	55.62 (methoxyl C), 113.91, 114.23, 118.20, 120.37, 120.91, 125.00, 127.50, 133.19, 133.51, 135.08, 162.85 (aromatic C and diazaphosphorine 5-C), 150.00 (diazaphosphorine 6-C), 194.28 (diazaphosphorine 4-C)
5g	55.65 (methoxyl C), 113.96, 114.30, 123.25, 123.33, 124.06, 124.25, 125.09, 127.00, 129.18, 131.40, 133.53, 133.82, 136.12, 141.67, 163.10 (aromatic C and diazaphosphorine 5-C), 150.17 (diazaphosphorine 6-C), 188.04 (diazaphosphorine 4-C)
5h	19.49, 31.45, 34.64 (cyclopentene C), 55.63 (methoxyl C), 113.81, 114.13, 133.14, 133.64, 162.81 (aromatic C), 125.44, 151.35, 188.80 (diazaphosphorine 5-, 6- and 4-C, respectively)
5i	14.07 (methyl C), 21.13 (5-methyl C), 34.31 (pyrrole 4-C), 55.63 (methoxyl C), 62.75 (methylene C), 72.75 (pyrrole 5-C), 113.63, 113.95, 1133.42, 133.69, 162.84 (aromatic C), 124.50, 152.63, 185.56 (diazaphosphorine 5-, 6- and 4-C, respectively)
5l	22.12 (methyl C) 43.53 (thiophene 4-C), 55.59 (methoxyl C) 77.00 (thiophene 5-C), 114.07,114.40, 133.16, 133.44, 163.50 (aromatic C), 124.20, 153.52, 184.45 (diazaphosphorine 5-, 6- and 4-C)
6m	22.69 (methyl C), 42.54 (furan 4-C), 55.84 (methoxyl C), 75.29 (furan 5-C), 113.80, 114.22, 127.50, 132.98, 133.25, 162.83 (aromatic C), 125.00, 153.81. 183.99 (thiazaphosphorine 5-, 4- and 6- C, respectively)

[a] The spectra of **5 d,e** were registered in DMSO-d_6 solution.

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TABLE IV

Microanalyses of **5a–l** and **6m**

Compd. no.	Molecular formula	Calc. (found) C	H	N	S
5a	$C_{16}H_{15}N_2OPS_2$	55.48 (55.68)	4.36 (4.28)	8.09 (8.19)	18.51 (18.29)
5b	$C_{22}H_{19}N_2OPS_2$	62.54 (62.58)	4.53 (4.43)	6.63 (6.80)	15.1 (15.38)
5c	$C_{13}H_{15}N_2OPS_2$	50.31 (50.02)	4.87 (4.90)	9.03 (9.12)	20.66 (20.65)
5d	$C_{14}H_{17}N_2OPS_2$	51.83 (51.78)	5.28 (5.30)	8.64 (8.50)	19.77 (19.92)
5e	$C_{15}H_{19}N_2OPS_2$	53.23 (53.01)	5.66 (5.77)	8.28 (8.47)	18.95 (18.93)
5f	$C_{14}H_{13}N_2OPS_2$	52.49 (52.58)	4.09 (4.11)	8.74 (8.77)	20.02 (19.91)
5g	$C_{16}H_{13}N_2OPS_3$	51.05 (50.96)	3.48 (3.49)	7.44 (7.54)	25.55 (25.70)
5h	$C_{13}H_{15}N_2OPS_2$	50.31 (50.12)	4.87 (4.78)	9.03 (9.20)	20.66 (20.85)
5i	$C_{16}H_{20}N_3O_3PS_2$	48.35 (48.39)	5.07 (5.05)	10.57 (10.46)	16.13 (16.00)
5l	$C_{13}H_{15}N_2OPS_3$	45.60 (45.75)	4.42 (4.39)	8.18 (8.08)	28.09 (27.85)
6m	$C_{13}H_{14}NO_2PS_3$	45.47 (45.17)	4.11 (4.18)	4.08 (4.12)	28.01 (28.09)

EXPERIMENTAL

All melting points were determined on a Kofler hot-stage apparatus and are uncorrected. Elemental analyses were performed on a Carlo Erba 1106 elemental analyzer. ^1H and ^{13}C NMR spectra were recorded on a Bruker WP 80 spectrometer using tetramethylsilane as internal standard and deuterochloroform and or dimethylsulphoxide-d_6 as solvents. I.R. spectra (potassium bromide discs) were recorded on a Perkin Elmer 281 spectrophotometer, and mass spectra on a VB ZAB-2SE spectrometer. Analytical TLC was performed on aluminum plates precoated with Merck silica gel 60-F_{254}. Crude products were separated by flash chromatography using Merck silica gel 60 and a mixture of cyclohexane-ethyl acetate (8:2, v/v) as eluent.

Starting materials. Enaminonitriles **1a–m** were prepared following literature procedures.[7–11] The Lawesson's reagent was purchased from Aldrich and used as received. Toluene was distilled from sodium benzophenone ketyl under nitrogen prior to use.

General procedure for the reaction of enaminonitriles **1** *with the Lawesson's reagent.* A mixture of enaminonitrile **1** (1 mmol) and Lawesson's reagent (1.2 mmol) in anhydrous toluene (50 ml) was refluxed for 1 hour. After cooling to room temperature, the solid was filtered off and then extracted twice with 30 ml of hot toluene. The filtrates were combined and evaporated in vacuo. The resultant residue was subjected to flash chromatography to obtain compounds **5a–l** and **6m**. Yields and other physical data of all isolated compounds are compiled in Table I; their ^1H and ^{13}C NMR data are reported in Tables II and III, respectively. The microanalyses data are listed in Table IV.

ACKNOWLEDGEMENT

The authors are grateful to the Italian M.U.R.S.T. which partially supported this work.

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