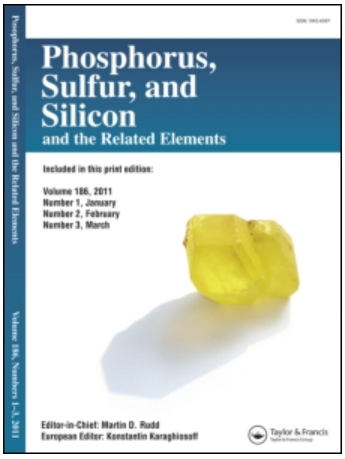

Phosphorus, Sulfur, and Silicon and the Related Elements
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713618290

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To cite this Article Reddy, B. Surender , Reddy, R. B. , Mouli, G. V. P. Chandra and Reddy, Y. D.(1991) 'SYNTHESIS OF POLYNUCLEAR HETEROCYCLES: 2-ARYLIDENE THIAZOLO(3,2-a)IMIDAZOLE RINGS FUSED TO BENZOTHIADIAZOLE AND PHENAZINE SYSTEMS', Phosphorus, Sulfur, and Silicon and the Related Elements, 63: 1, 143 — 146
To link to this Article: DOI: 10.1080/10426509108029437
URL: http://dx.doi.org/10.1080/10426509108029437

Phosphorus, Sulfur, and Silicon, 1991, Vol. 63, pp. 143–146
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SYNTHESIS OF POLYNUCLEAR HETEROCYCLES: 2-ARYLIDENE THIAZOLO(3,2-a)IMIDAZOLE RINGS FUSED TO BENZOTHIADIAZOLE AND PHENAZINE SYSTEMS

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(Received March 14, 1991; in final form May 15, 1991)

2-Arylidene thiazolo(3,2-a)imidazo(4,5-b)phenazine-3(2H)-ones and 2-arylidene thiazolo(3,2-a)imidazo(4,5-e)-(2,1,3)benzothiadiazol-3 (2H)-ones were prepared from the reaction of 2-mercapto imidazophenazine/2-mercapto imidazo-(2,1,3)benzothiadiazole with chloro acetic acid and aromatic aldehydes in the presence of acetic anhydride, acetic acid and sodium acetate. They were characterised by their analytical and spectral data.

Key words: Imidazobenzothiadiazole; imidazo phenazine; benzal proton; antifungal; antibacterial.

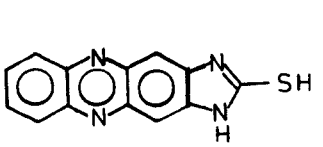
Thiazolo(3,2-a)benzimidazole derivates are reported to exhibit significant anti-inflammatory,[1] virucidal,[2] antiulcer[3] and herbicidal[4] activities. Thiazolo(3,2-a)benzimidazol-3-(2H)-ones are claimed to possess insecticidal, pesticidal, antifungal and antibacterial activities.[5,6] Incorporation of arylidene moiety[7] into heterocyclic systems lead to compounds of increased antifungal activity. We reported[8] earlier that fused heterocyclic rings containing nitrogen and sulphur exhibited remarkable antimicrobial properties. The varied biological properties of thiazolobenzimidazoles stimulated us to synthesize the title compounds and to screen them for their antimicrobial activity.

The cyclisation of 2-mercapto imidazophenazine (**I**)[9] and 2-mercapto imidazo(2,1,3)benzothiadiazole (**II**)[10] is carried out with different aromatic aldehydes and chloro acetic acid in the presence of acetic anhydride, acetic acid and sodium acetate to give 2-arylidene thiazolo(3,2-a)imidazo-(4,5-b)phenazine-3 (2H)-ones (**III**) and 2-arylidene thiazolo-(3,2-a)imidazo(4,5-e)(2,1,3)benzothiadiazol-3 (2H)-ones (**IV**) respectively. All these compounds have been identified by their analytical and spectral data.

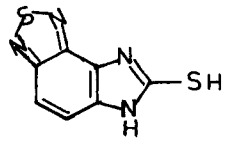
All these compounds displayed a strong carbonyl absorption around 1700 cm^{-1} in their IR spectra. The other IR bands appeared at 1590 cm^{-1} (C=N), 1540 cm^{-1} (C=C).

The chemical shifts in PMR spectra of compounds of both types (**III** and **IV**) indicated the presence of aromatic protons. They appeared as a broad peak from δ7.8 to δ8.6. The benzal proton appeared around δ4.4 to δ4.6. But the splitting pattern was not clear due to the poor solubilities of these compounds in NMR solvents.

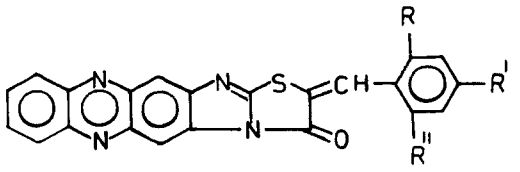
The structures of the above compounds were also confirmed by their mass spectra. The molecular ion of **IIIa** was recorded as a base peak at *m/z* 414 with M +



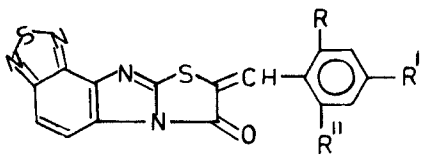
I



II



III



IV

TABLE I

Physical and analytical data of compounds **III** and **IV**

Compd. No.	R[a], R′, R″	Mol. formula	M.P. °C	Yield %	Analyses[b] Found (Calcd.) C	H	N	S
III a	R=R″=H, R′=Cl	$C_{22}H_{11}ON_4S\,Cl$	325–26	76	63.77	2.66	13.53	7.73
b	R=Cl, R′=R″=H	$C_{22}H_{11}ON_4S\,Cl$	316–317	60	63.77	2.66	13.53	7.73
c	R=R″=H, R′=Br	$C_{22}H_{11}ON_4S\,Br$	320–321	72	57.52	2.40	12.2	6.97
d	R=H, R′=R″=Cl	$C_{22}H_{10}ON_4S\,Cl_2$	322–323	69	58.92	2.23	12.5	7.14
e	R=R″=Cl, R′=H	$C_{22}H_{10}ON_4S\,Cl_2$	318–319	64	58.92	2.23	12.5	7.14
f	R=R″=H, R′=CH_3	$C_{23}H_{14}ON_4S$	299–300	71	70.05	3.55	14.22	8.12
g	R=R″=H, R′=OCH_3	$C_{23}H_{14}O_2N_4S$	309–310	58	67.32	3.42	13.66	7.80
h	R=R″=H, R′=NO_2	$C_{22}H_{11}O_3N_5S$	323–24	74	62.12	2.59	16.47	7.59
i	R=NO_2, R′=R″=H	$C_{22}H_{11}O_3N_5S$	312–313	50	62.12	2.59	16.47	7.59
j	R=R″=H, R′=OH	$C_{22}H_{12}O_2N_4S$	324–325	52	66.67	3.03	14.14	8.08
IV a	R=R″=H, R′=Cl	$C_{16}H_7ON_4S_2Cl$	310–311	79	51.89	1.89	15.14	17.30
b	R′=Br, R=R″=H	$C_{16}H_7ON_4S_2Br$	309–310	74	46.26	1.69	13.49	15.42
c	R″=H, R′=R=Cl	$C_{16}H_6ON_4S_2Cl_2$	321–322	65	47.52	1.49	13.86	15.84
d	R=R″=Cl, R′=H	$C_{16}H_6ON_4S_2Cl_2$	287–288	62	47.52	1.49	13.86	15.84
e	R=R′=R″=H	$C_{16}H_8ON_4S_2$	180–181	70	57.14	2.38	16.67	19.05
f	R=R″=H, R′=OCH_3	$C_{17}H_{10}O_2N_4S_2$	299–300	56	55.74	2.73	15.3	17.49
g	R=R″=H, R′=NO_2	$C_{16}H_7O_3N_5S_2$	320–321	68	50.39	1.84	18.37	16.8
h	R′=R″=H, R=NO_2	$C_{16}H_7O_3N_5S_2$	318–319	51	50.39	1.84	18.37	16.8

[a]Compounds recrystallised from chloroform, DMSO, pyridine and dioxane.
[b]Analysis for halogens also found satisfactory.

2 peak due to the isotopic contributions of chlorine and sulphur. The molecular ion suffers a loss of CO to give a peak at *m/z* 386 which further ejects chlorine to record at *m/z* 351. The ion at *m/z* 351 splits away to give up HNCS and appears at *m/z* 292. The other peaks for fragments appear at *m/z* values of 264, 220, 153 etc.

The mass spectrum of **IVb** displays a molecular ion at *m/z* 414 which is also a base peak. It loses CO to record at *m/z* 387 and it ejects bromine to give a peak at *m/z* 307. The molecular ion also loses p-bromo phenyl ketene to give a peak at *m/z* 207. The other major peaks at *m/z* values of 275, 262, 180 are also observed.

BIOLOGICAL SCREENING

A few compounds of the type **III** and **IV** were evaluated against bacteria such as *Bacillus polymixa*, *Bacillus subtilis* (gram +ve) and *Proteus vulgaris* (gram −ve) by using the filter paper disc diffusion technique.[11] Compounds **IIIa** and **IVb** showed feeble activity against gram +ve bacteria but are non-toxic to gram −ve bacteria. All other compounds have registered no activity against the bacteria tested.

A few of the title compounds were also screened against fungi such as *Dreschlera specifera* and *Fusarium oxysporum* by adopting food poisoning technique.[12] Compounds **IIIb** and **IVb** are moderately active against *D. specifera* at 840 μg/ml concentration level, while compounds **IIIa** and **IIIc** are more active against the same fungi at the same dose level. Compounds **IIIa**, **IIIb** and **IVa** registered 100% spore germination inhibition in *F. oxysporum* at 360 μg/ml. Compounds **IIIc** and **IVd** have shown 100% inhibition in the same fungi at 600 μg/ml. The rest of the compounds are found to have moderate to feeble activity.

EXPERIMENTAL

All the melting points are uncorrected. IR spectra were recorded on Schimadzu Spectrometer in KBr pellets/Nujol. NMR spectra were recorded on a Varian EM-360L spectrometer using TMS as internal standard (chemical shifts in δppm). Mass spectra were taken on JMS-D-300 Joel mass spectrometer at 70 eV.

*2-Arylidene thiazolo(3,2-a)imidazo(4,5-b)phenazine-3-(2H)-ones(**III**) or 2-Arylidene thiazolo(3,2-a)imidazo(4,5-e)(2,1,3)-benzothiadiazole-3 (2H)-ones (**IV**).* A mixture of 2-mercapto-1H-imidazo phenazine (0.01 mole, I) or 2-mercapto imidazo(2,1,3)benzothiadiazole (0.01 mol, II), fused sodium acetate (2g), chloro acetic acid (1.43 g; 0.015 mol) and an aromatic aldehyde (0.01 mol) in glacial acetic acid (25 ml) and acetic anhydride (20 ml) was refluxed for 24 hrs and cooled. The precipitated solid was filtered off, dried and recrystallised from suitable solvents.

ACKNOWLEDGEMENTS

The authors are thankful to the Principal, Regional Engineering College, Warangal, for providing facilities, especially GVPC for providing financial assistance from College Research Grants. One of the authors (BSR) is thankful to CSIR, New Delhi (India) for the award of a Senior Research Fellowship. We are also thankful to Dr. S. M. Reddy and Mr. Lakshma Reedy, Dept. of Botany, Kakatiya University, for carrying out the biological activity investigations.

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