Synthesis of Substituted 5-Hydrothiazolo[4,3-*b*]-1,3,4-oxa(thia)diazoles and 5-Hydrothiazolo[3,4-*b*]-1,2,4-triazoles as Potential Antifungal Agents

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2-Aryl-3-(3-arylthioureido)-4-thiazolidinones (IIIa–d) were prepared by addition–condensation of aldehyde 4-arylthiosemicarbazones (IIa–d) and mercaptoacetic acid. 5-Aryl-2-(arylamino)-5-hydrothiazolo-[4,3-*b*]-1,3,4-thiadiazoles (IVa–d), the corresponding thiazolo[4,3-*b*]-1,3,4-oxadiazoles (Va–d), and 1,5-diaryl-5-hydro-2-mercaptothiazolo[3,4-*b*]-1,2,4-triazoles were obtained by chemoselective intramolecular heterocyclization of IIIa–d with concentrated H_2SO_4, CH_3I, and NaOH, respectively. Compounds IIIa–d—VIa–d were compared with Dithane M-45, a commercial fungicide, for their antifungal action against *Aspergillus flavus* and *Fusarium solani*. The screening results have been correlated with the structural features of the tested compounds.

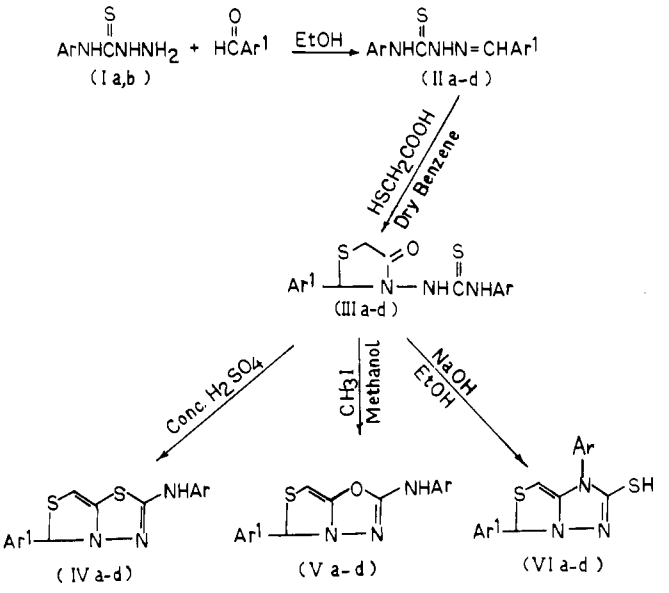
INTRODUCTION

Penicillins, which are among the most important antibiotics in clinical use, contain a thiazole nucleus fused with a β-lactam ring. Likewise, thiazolotriazole derivatives have been reported to display useful pesticidal activities (Singh et al., 1985; Shigematsu et al., 1980; Nomura et al., 1977). In addition a large number of nitrogen-bridged heterocyclic systems incorporating 1,3,4-oxa(thia)diazole and 1,2,4-triazole rings have been reported as effective fungicidal (Singh et al., 1987; Shau and Nayak, 1990), bactericidal (Sinnur et al., 1986; Patel et al., 1990; Khalil et al., 1990), and herbicidal (Astles et al., 1991; Kumura et al., 1990) agents. In view of the above papers and our desire to develop new antifungal agents of high potency, we fused a biolabile thiazole nucleus with biologically versatile 1,3,4-oxa(thia)diazole and 1,2,4-triazole rings to probe to what extent these combinations could be successful. The investigation appeared to be interesting as the compounds IVa–d—VIa–d reported herein are hitherto unknown bicyclic nitrogen-bridged heterocycles.

The synthetic approach to the title compounds is outlined in Scheme I. Addition condensation of aldehyde 4-arylthiosemicarbazones (IIa–d) to mercaptoacetic acid furnished the substituted 4-thiazolidinones (IIIa–d). IIIa–d were cyclized chemoselectively with concentrated H_2SO_4, CH_3I, and NaOH to give the corresponding thiazolo[4,3-*b*]-1,3,4-thiadiazoles (IVa–d), thiazolo[4,3-*b*]-1,3,4-oxadiazoles (Va–d), and thiazolo[3,4-*b*]-1,2,4-triazoles (VIa–d), respectively.

The structural assignments of the synthesized compounds were based on elemental analyses, IR, ^1H NMR, and mass spectra (Tables I and II). The IR spectra of 3-(3-arylthioureido)-4-thiazolidinones (IIIa–d) revealed a strong band around 1690 cm^{-1} (νC=O), a medium band near 3330 cm^{-1} (νN—H), and a band around 1210 cm^{-1} (νC=S). The disappearance of νC=O and νC=S bands in the IR spectra of compounds IV–VI is compatible with their assigned structures. Further, compounds IV and V exhibited a weak band around 3310 cm^{-1} attributable to νN—H. Compounds VI were devoid of this band but showed a weak band near 2550 cm^{-1} due to νS—H. Of the tested compounds IIIa–d—VIa–d, compounds IVd, Vd, and VId displayed antifungal activity of the order of Dithane M-45 [a commercial fungicide, mangnous ethylene bis(dithiocarbamate) with zinc ions] at 1000 ppm concentration against *Aspergillus flavus* and *Fusarium solani*.

Scheme I



II a,b-VI a,b: Ar = 4-$CH_3C_6H_4$; Ar^1 = 2-ClC_6H_4, 4-$OCH_3C_6H_4$

II c,d-VI c d: Ar = 4-ClC_6H_4; Ar^1 = 4-$CH_3C_6H_4$, 4-$OCH_3C_6H_4$

EXPERIMENTAL PROCEDURES

Melting points were determined in open glass capillaries and are uncorrected. IR spectra in KBr were recorded on a Perkin-Elmer 157 infrared spectrophotometer (ν_{max} cm^{-1}). ^1H NMR spectra were recorded on a Varian EM-360L (60 MHz) NMR spectrometer in $CDCl_3$ plus DMSO-d_6 using TMS as internal reference; chemical shifts are expressed in δ. Mass spectra were recorded on a JEOL JMS-D300 instrument.

4-Arylthiosemicarbazides (Ia,b). These were prepared by following the method of Kazakov and Postovskii (1961). Thus, aromatic amines (0.1 mol) in ammonia (25 mL, sp. gr., 0.88) were treated with CS_2 (10 mL), sodium chloroacetate (0.1 mol), and hydrazine hydrate (10 mL, 50%) successively. It was warmed and then cooled and kept overnight. The 4-arylthiosemicarba-

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Table I. Analytical Data of Compounds IIIa–d—VIa–d

compd	yield	mp, °C	molecular formula	found (calcd), % C	H	N
IIIa	60	215–216	$C_{17}H_{16}ClN_3OS_2$	53.83 (54.04	4.08 4.24	11.32 11.13)
IIIb	66	177–178	$C_{18}H_{19}N_3O_2S_2$	58.08 (57.91	4.86 5.09	11.22 11.26)
IIIc	63	227–229	$C_{17}H_{16}ClN_3OS_2$	54.08 (54.04	4.02 4.24	11.18 11.13)
IIId	62	205–206	$C_{17}H_{16}ClN_3O_2S_2$	51.92 (51.84	3.99 4.07	10.75 10.67)
IVa	80	238–239	$C_{17}H_{14}ClN_3S_2$	56.92 (56.75	4.13 3.89	11.84 11.68)
IVb	83	205–206	$C_{18}H_{17}N_3OS_2$	61.13 (60.85	4.71 4.79	11.72 11.83)
IVc	74	244–245	$C_{17}H_{14}ClN_3S_2$	56.68 (56.75	3.96 3.89	11.85 11.68)
IVd	78	166–167	$C_{17}H_{14}ClN_3OS_2$	54.48 (54.33	3.79 3.73	11.02 11.19)
Va	64	195–196	$C_{17}H_{14}ClN_3OS$	59.48 (59.39	4.24 4.08	12.38 12.23)
Vb	69	110–111	$C_{18}H_{17}N_3O_2S$	63.54 (63.72	4.92 5.02	12.58 12.39)
Vc	59	175–176	$C_{17}H_{14}ClN_3OS$	59.59 (59.39	4.17 4.08	12.43 12.23)
Vd	66	118–120	$C_{17}H_{14}ClN_3O_2S$	56.53 (56.75	3.74 3.89	11.81 11.68)
VIa	72	173	$C_{17}H_{14}ClN_3S_2$	56.71 (56.75	3.72 3.89	11.54 11.68)
VIb	79	163–164	$C_{18}H_{17}N_3OS_2$	60.73 (60.85	4.88 4.79	12.02 11.83)
VIc	68	188–189	$C_{17}H_{14}ClN_3S_2$	56.58 (56.75	3.82 3.89	11.76 11.68)
VId	73	170–171	$C_{17}H_{14}ClN_3OS_2$	54.14 (54.33	3.90 3.73	11.28 11.19)

zides thus formed were recrystallized from ethanol. Ia (Ar = 4-$CH_3C_6H_4$) and Ib (Ar = 4-ClC_6H_4) agreed well with their analytical data already reported in the literature (Tisler, 1956; Ramachander and Srinivasan, 1962).

Aldehyde 4-Arylthiosemicarbazones (IIa–d). These were prepared by the usual condensation of appropriate aromatic aldehydes (0.1 mol) with 4-arylthiosemicarbazides (0.1 mol) and were recrystallized from ethanol as shining yellowish needles. Compounds IIa–d are already reported in the literature (Tisler, 1956; Srinivasan and Ramachander, 1961).

2-Aryl-3-(3-arylthioureido)-4-thiazolidinones (IIIa–d). These compounds were prepared by the addition–condensation of thiosemicarbazides and mercaptoacetic acid following the method of Astik et al. (1975). Thus, to a well-stirred solution of II (0.05 mol) in dry benzene (50 mL) was added mercaptoacetic acid (0.075 mol). The mixture was refluxed for 6 h on a water bath; the clear solution thus obtained was allowed to cool and then poured into water. The crystalline product thus obtained was washed with water and recrystallized from ethanol.

5-Aryl-2-(arylamino)-5-hydrothiazolo[4,3-*b*]-1,3,4-thiadiazoles (IVa–d). These were prepared according to the method of Maffii et al. (1959). 4-Thiazolidinones IIIa–d (0.01 mol) were treated dropwise with concentrated H_2SO_4 (5 mL) between 0 and 15 °C. The mixture in each case was cooled and poured on crushed ice. On neutralization with ammonia the desired product (IV), which precipitated out, was recrystallized from ethanol.

5-Aryl-2-(arylamino)-5-hydrothiazolo[4,3-*b*]-1,3,4-oxadiazoles (Va–d). An equimolar mixture of III and methyl iodide was refluxed for 6 h in methanol and excess solvent distilled off. The residue was treated with 5% NaOH, and the product thus obtained was washed with water and recrystallized from ethanol to furnish V.

1,5-Diaryl-5-hydro-2-mercaptothiazolo[3,4-*b*]-1,2,4-triazoles (VIa–d). A solution of III (0.01 mol) in NaOH (aqueous 4%, 33 mL) and ethanol (20 mL) was refluxed for 2 h. The resulting mixture was cooled and filtered. The filtrate was adjusted to pH 5–6 with 5 N HCl, and the precipitated solid was filtered and recrystallized from ethanol.

Analytical data of compounds IIIa–d—VIa–d are given in Table I, and spectral data are recorded in Table II.

Antifungal Screening. The antifungal activity of compounds IIIa–d—VIa–d was evaluated against *A. flavus* and *F. solani*. The pure cultures of the tested fungi, the pathogenicity of which was already verified, were obtained from the Division of Mycology and Plant Pathology, Indian Agricultural Research Institute, New Delhi. Agar (bacteriological grade), supplied by Sharabhai M. Chemicals, was used as such. Antifungal screening was done by the usual agar plate technique (Horsfall, 1945) at 1000, 100, and 10 ppm concentrations using Czapek's agar medium (Raper and Thom, 1968). The compounds were applied as suspensions in an acetone–water mixture (20:80 v/v). One milliliter of the test suspension was thoroughly mixed with 9 mL of the medium by rotating the plates on table top, and then the mixture was allowed to set. A fungal disk of 5-mm diameter was cut out with the help of a sterilized cork borer from the periphery of 1-week-old culture of the test fungus already planted on the Czapek's agar medium and was inoculated in the center of each Petri plate in an inverted position to bring the mycelia in direct contact with the medium. Petri plates containing 9 mL of Czapek's agar medium and 1 mL of acetone–water mixture (20:80 v/v) served as controls. The number of replicate assays in each case was three, whereas six replications of controls were provided. The plates were incubated at 28 °C (±1 °C) for 96 h. After 96 h, four diameters of the fungal colony, intersecting one another at about 45°, were measured with a millimeter scale and percent inhibition of mycelial growth was calculated by

$$\% \text{ inhibition} = [(C - T) \times 100]/C$$

where C is the average diameter of fungal colony (mm) in control plates and T is the average diameter of fungal colony (mm) in treated plates.

Dithane M-45, a commercial fungicide, was also tested under similar conditions for comparison. The antifungal screening results of synthesized compounds are summarized in Table III.

RESULTS AND DISCUSSION

The formation of substituted 5-hydrothiazolo[4,3-*b*]-1,3,4-oxa(thia)diazoles (IV and V) and 5-hydrothiazolo-[3,4-*b*]-1,2,4-triazoles (VI) involves the chemoselective

Table II. Spectral Data of Compounds IIIa–d—VIa–d

compd	IR (KBr) ν, cm^{-1}			^1H NMR ($CDCl_3$ + DMSO-d_6), δ	MS/M^+, m/z
	νC=N, νC=S*	νC=O	νN—H, νS=H$^+$		
IIIa	1210*	1685	3330	9.16 (br s, 2 H, 2 × N*H*) 6.78–7.92 (m, 9 H, Ar-*H* and SC*H*Ar) 2.64 (s, 2 H, SCH_2CO) 2.28 (s, 3 H, CH_3)	377, 379
IIIb	1205*	1680	3315	9.13 (br s, 2 H, 2 × N*H*) 6.73–7.96 (m, 9 H, Ar-*H* and SC*H*Ar) 3.80 (s, 3 H, OCH_3) 2.67 (s, 2 H, SCH_2CO) 2.33 (s, 3 H, CH_3)	373
IIIc	1210*	1690	3325	9.20 (br s, 2 H, 2 × N*H*) 6.67–7.92 (m, 9 H, Ar-*H* and SC*H*Ar) 2.73 (s, 2 H, SCH_2CO) 2.30 (s, 3 H, CH_3)	377, 379
IIId	1210*	1685	3325	9.26 (br s, 2 H, 2 × N*H*) 6.80–7.98 (m, 9 H, Ar-*H* and SC*H*Ar) 3.78 (s, 3 H, OCH_3) 2.76 (s, 2 H, SCH_2CO)	393, 395
IVa	1625		3315	9.14 (br s, 1 H, N*H*) 6.76–7.94 (m, 10 H, C_5-*H*, C_7-*H*, and Ar-*H*) 2.32 (s, 3 H, CH_3)	359, 361
IVb	1620		3320	9.18 (br s, 1 H, N*H*) 6.73–7.98 (m, 10 H, C_5-*H*, C_7-*H*, and Ar-*H*) 3.78 (s, 3 H, OCH_3) 2.30 (s, 3 H, CH_3)	355
IVc	1625		3310	9.22 (br s, 1 H, N*H*) 6.70–7.94 (m, 10 H, C_5-*H*, C_7-*H*, and Ar-*H*) 2.32 (s, 3 H, CH_3)	359, 361
IVd	1620		3320	9.28 (br s, 1 H, N*H*) 6.84–7.98 (m, 10 H, C_5-*H*, C_7-*H*, and Ar-*H*) 3.76 (s, 3 H, OCH_3)	375, 377
Va	1610		3315	9.16 (br s, 1 H, N*H*) 6.78–7.94 (m, 10 H, C_5-*H*, C_7-*H*, and Ar-*H*) 2.34 (s, 3 H, CH_3)	343, 345
Vb	1620		3315	9.10 (br s, 1 H, N*H*) 6.72–7.94 (m, 10 H, C_5-*H*, C_7-*H*, and Ar-*H*) 3.78 (s, 3 H, OCH_3) 2.30 (s, 3 H, CH_3)	339
Vc	1625		3320	9.20 (br s, 1 H, N*H*) 6.66–7.92 (m, 10 H, C_5-*H*, C_7-*H*, and Ar-*H*) 2.32 (s, 3 H, CH_3)	343, 345
Vd	1625		3310	9.27 (br s, 1 H, N*H*) 6.76–7.96 (m, 10 H, C_5-*H*, C_7-*H*, and Ar-*H*) 3.76 (s, 3 H, OCH_3)	359, 361
VIa	1630		2560$^+$	6.70–7.88 (m, 10 H, C_5-*H*, C_7-*H*, and Ar-*H*) 2.30 (s, 3 H, CH_3)	359, 361
VIb	1620		2550$^+$	6.67–7.93 (m, 10 H, C_5-*H*, C_7-*H*, and Ar-*H* 3.78 (s, 3 H, OCH_3) 2.28 (s, 3 H, CH_3)	355
VIc	1625		2555$^+$	6.62–7.90 (m, 10 H, C_5-*H*, C_7-*H*, and Ar-*H*) 2.28 (s, 3 H, CH_3)	359, 361
VId	1630		2560$^+$	6.70–7.94 (m, 10 H, C_5-*H*, C_7-*H*, and Ar-*H*) 3.78 (s, 3 H, OCH_3)	375, 377

intramolecular heterocyclization of 2-aryl-3-(3-arylthioureido)-4-thiazolidinones (IIIa–d). Chemoselectivity in the intramolecular cyclization of III to IV–VI may be rationalized by the "hard and soft acids and bases, HSAB, principle". In the case of cyclization of IIIa–d with concentrated H_2SO_4, the protonation of carbonyl oxygen leads to the corresponding IVa–d via intramolecular nucleophilic attack by thionic S of the thiourea moiety (Maffii et al., 1959). Methyl iodide, a soft Lewis acid, methylates the thionic S of the thiourea function of III followed by cyclization to V with the elimination of methanethiol, which could be easily detected. The reaction of III with NaOH furnishes N,S-ambident anion, the terminal nitrogen of which attacks the carbonyl carbon to yield the corresponding VI (Postovskii and Vereshchagina, 1956).

It appears from the screening data (Table III) that most of the compounds showed significant antifungal activity at 1000 ppm against *A. flavus* and *F. solani*, but their activity decreased markedly on dilution. Compounds IVd, Vd, and VId displayed antifungal activity of the order of commercial fungicide, Dithane M-45, at 1000 ppm and inhibited more than 48% growth of both test fungi even at 10 ppm concentration. In general, compounds IVa–d—VIa–d were invariably more potent than their parent 4-thiazolidinones (IIIa–d). Further, thiazolothiadiazoles (IVa–d) and thiazolotriazoles (VIa–d) were slightly better than thiazolooxadiazole analogues (Va–d).

Although some of the compounds were considerably toxic against both fungal species at higher concentration (1000 ppm), the overall results are not so encouraging as one would expect from the combined performance of the fused biolabile nuclei. This might be attributed to the partial saturation in the thiazole nucleus resulting in the loss of planarity of thiazolooxa(thia)diazole (IV and V) and thiazolotriazole (VI) N-bridged heterocyclic systems. This presumption is supported by earlier observations that compact size and planarity of a molecule often enhance its pesticidal activity (Chatt et al., 1956; Fischer and Summers, 1976; Rothwell and Wain, 1963).

It is, however, noteworthy that the introduction of a

Table III. Antifungal Screening Results of Compounds IIIa–d—VIa–d

compd	av % inhibition in vitro after 96 h against *A. flavus* 1000 ppm	100 ppm	10 ppm	*F. solani* 1000 ppm	100 ppm	10 ppm
IIIa	55	28	18	53	33	12
IIIb	49	23	10	47	20	10
IIIc	53	26	13	49	30	11
IIId	69	39	21	69	40	27
IVa	80	59	43	78	55	42
IVb	58	29	19	60	29	19
IVc	78	53	40	81	52	41
IVd	93	68	53	96	70	51
Va	71	42	26	72	41	28
Vb	54	27	19	58	28	18
Vc	65	31	20	63	30	21
Vd	92	61	49	93	59	49
VIa	86	69	49	87	57	44
VIb	76	49	33	78	51	33
VIc	81	57	35	85	56	34
VId	95	70	56	99	71	55
Dithane M-45	96	82	66	100	86	73

chloro or methoxy group in the aryl moiety of these compounds tends to augment the antifungal action. From the comparison of the fungitoxicity data (Table III) of compounds IIIb and IIId, IVb and IVd, Vb and Vd, and VIb and VId it seems that the introduction of a chloro group is comparable to that of a methyl group and that the former seems to be better than the latter. The antifungal action varied but marginally with the fungal species.

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Registry No. IIIa, 141585-98-0; IIIb, 141585-99-1; IIIc, 141586-00-7; IIId, 141586-01-8; IVa, 141586-02-9; IVb, 141586-03-0; IVc, 141586-04-1; IVd, 141586-05-2; Va, 141586-06-3; Vb, 141586-07-4; Vc, 141586-08-5; Vd, 141586-09-6; VIa, 141586-10-9; VIb, 141586-11-0; VIc, 141586-12-1; VId, 141586-13-2.