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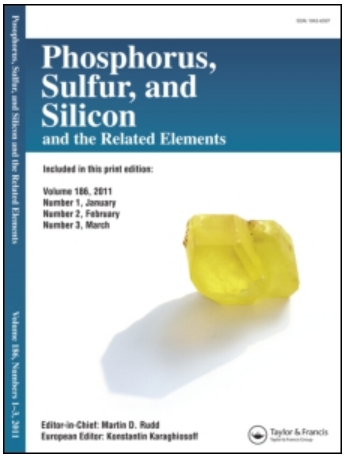

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REACTIONS OF PHENYL ISOTHIOCYANATE AND SULFUR WITH DIMERIC ADDUCTS: NOVEL SYNTHESIS OF THIAZOLES, THIAZOLO-[4,5-d]PYRIMIDINE AND THIAZOLO[4,5-d]-PYRIDINE DERIVATIVES

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REACTIONS OF PHENYL ISOTHIOCYANATE AND SULFUR WITH DIMERIC ADDUCTS: NOVEL SYNTHESIS OF THIAZOLES, THIAZOLO-[4,5-d]PYRIMIDINE AND THIAZOLO[4,5-d]-PYRIDINE DERIVATIVES

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Dimeric adducts **1–3** react with phenyl isothiocyanate and sulfur to afford the thiazole derivatives **4**, **12** and **14**. The latter products react with different chemical reagents to afford new thiazoles and their fused derivatives which show high fungicidal and bactericidal activities.

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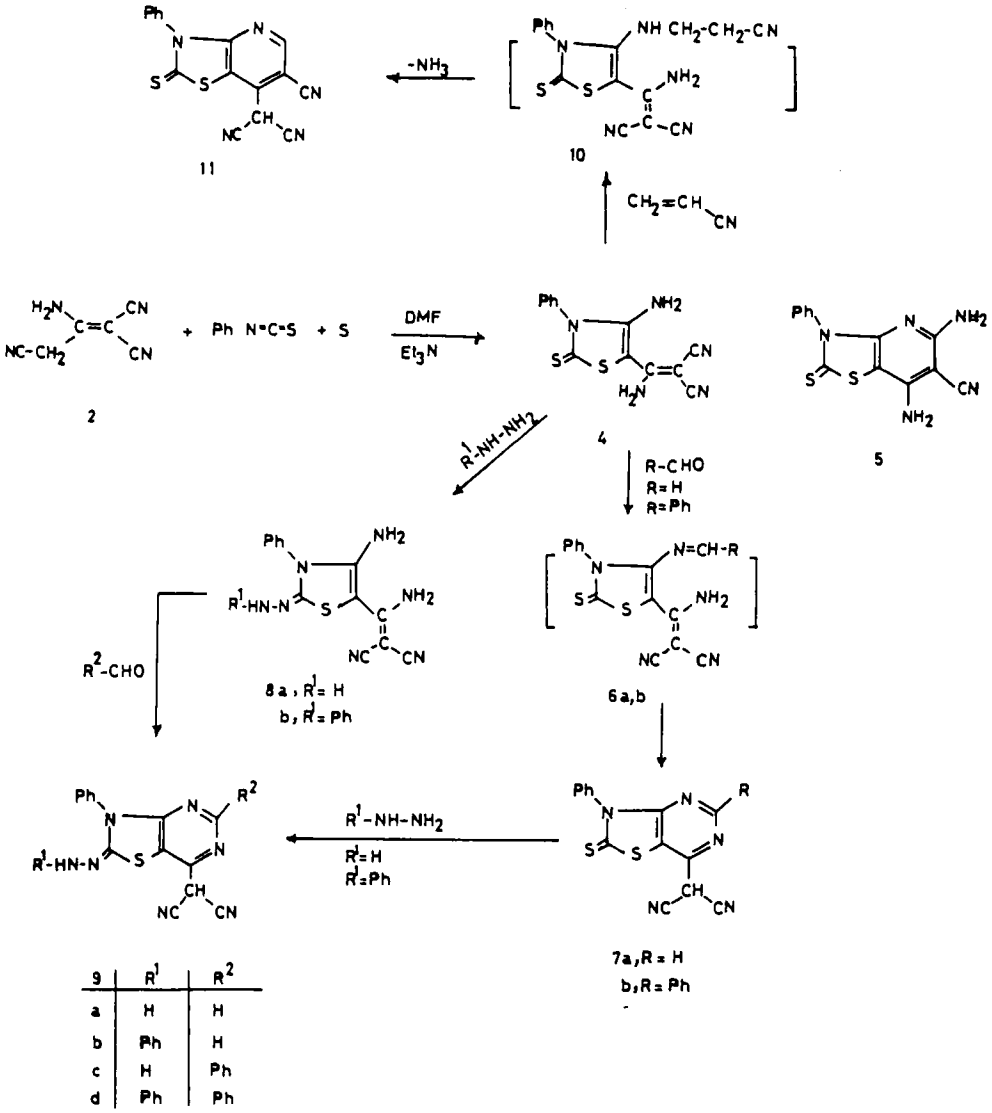
INTRODUCTION

Recently we were involved in studying the reaction of phenyl isothiocyanate with active methylene reagents followed by cyclization with α-halocarbonyl compounds to afford thiazolidinone and 2,3-dihydrothiazole derivatives[1–3] which are of potential anesthetic,[4] hypoglycemic[5] and fungicidal[6] activities. In this article we describe our trials to make an extension of such synthetic routes using dimeric adducts of malononitrile, ethyl cyanoacetate and acetonitrile as active methylene reagents to react with phenyl isothiocyanate and sulfur element[7,8] to afford thiazoles and their fused derivatives together with studying the biological activities of the newly synthesized products.

RESULTS AND DISCUSSION

Although 3-amino-2,4-dicyano-crotononitrile (**1**),[9] 3-amino-2,4-diethoxycarbonyl-crotononitrile (**2**)[10] and 3-iminobutyronitrile (**3**)[11] were recently used in heterocyclic synthesis,[12–15] to our knowledge no attention was paid for their use to afford thiazole derivatives. Thus, we now report that **1** reacts with phenyl isothiocyanate and sulfur in dimethylformamide solution and in the presence of triethylamine to afford a single product with molecular formula $C_{13}H_9N_5S_2$. Two possible isomeric structures **4** and **5** were considered. The possibility of structure **5** is ruled out on the basis of spectral and chemical evidences. The IR spectrum of the reaction product revealed the presence of two CN groups stretching at 2220, 2210 cm^{-1}. The ^1H NMR



9	R^1	R^2
a	H	H
b	Ph	H
c	H	Ph
d	Ph	Ph

SCHEME I

spectrum revealed the presence of two singlets at δ = 2.84 and 3.48 ppm for two NH_2 protons (D_2O) exchangeable signals) and a multiplet at δ = 7.10–7.65 ppm for C_6H_5 protons. Such data are in agreement with structure **4**. Reaction of **4** with both benzaldehyde and formaldehyde in refluxing dimethylformamide and in the presence of a catalytic amount of triethylamine afforded the thiazolo[4,5-d]pyrimidine derivatives **7a,b**. Formation of the latter products are explained in terms of the intermediate formation of the expected Schiff's base **6a,b** followed by cyclization and oxidation[16]. Structures of compounds **7a,b** are established based on IR and ^1H NMR spectral data (cf. Table II).

Reaction of **4** with hydrazine hydrate and phenylhydrazine afford the thiazol-2-hydrazone derivatives **8a,b**. The latter products reacted with each of formaldehyde

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and benzaldehyde to afford the thiazolo[4,5-d]pyrimidine derivatives **9a–d**. Structures of compounds **8a,b** and **9a–d** were confirmed based on analytical and spectral data (cf. Tables I and II). Moreover, the structures of **9a–d** were established based on their synthesis via another reaction route. Thus, reaction of **7a,b** with hydrazine hydrate and phenylhydrazine afforded the same products **9a–d** (m.p., mixed m.p. and IR spectrum).

TABLE I

Physical and analytical data of the newly prepared compounds

Compd. (colour)	solvent	m.p. (°C)	yield (%)	Mol. formula (M. wt.)	Analysis (Calcd./Found) % C	H	N	S
4	EtOH	201	90	$C_{13}H_9N_5S_2$	52.0	3.0	23.4	21.4
(brown)				(299.37)	52.0	3.0	23.6	21.8
7a	EtOH	282–5	78	$C_{14}H_7N_5S_2$	54.3	2.2	22.6	20.7
(brown)				(309.37)	54.1	2.5	22.4	20.2
7b	EtOH	225–7	72	$C_{20}H_{11}N_5S_2$	62.3	2.8	18.2	16.6
(yellow)				(385.47)	62.0	3.0	18.0	16.5
8a	EtOH	273–7	81	$C_{13}H_{11}N_7S$	52.5	3.7	32.9	10.7
(brown)				(297.34)	52.3	3.4	32.5	10.4
8b	EtOH	195	69	$C_{19}H_{15}N_7S$	61.1	4.0	26.2	8.5
(yellow)				(373.35)	60.8	4.3	26.0	8.4
9a	DMF	216–8	77	$C_{14}H_9N_7S$	54.7	2.9	31.9	10.4
(orange)				(307.34)	54.5	3.1	31.6	10.3
9b	DMF	226–9	70	$C_{20}H_{13}N_7S$	62.6	3.3	25.5	8.3
(orange)				(383.35)	62.2	3.0	25.2	8.1
9c	MeOH	235–7	62	$C_{20}H_{13}N_7S$	62.6	3.3	25.5	8.3
(yellow)				(383.35)	62.2	3.2	25.8	8.4
9d	AcOH	170	67	$C_{26}H_{17}N_7S$	67.9	3.7	21.3	6.9
(red)				(459)	67.6	3.9	21.0	7.2
11	EtOH	168	78	$C_{16}H_7N_5S_2$	57.6	2.1	21.0	19.1
(yellow)				(333.41)	57.6	2.5	20.7	18.9
12	EtOH	210–12	81	$C_{15}H_{13}N_3O_3S_2$	51.8	3.7	12.1	18.4
(green)				(347.40)	51.5	3.9	11.7	18.2
13a	EtOH	145	73	$C_{15}H_{15}N_5O_3S$	52.1	4.5	20.2	9.2
(brown)				(345.37)	52.3	4.6	20.0	9.5
13b	MeOH	141	77	$C_{21}H_{19}N_5O_3S$	59.8	4.6	16.6	7.6
(brown)				(421.44)	59.6	4.8	16.3	7.9
14	MeOH	55	70	$C_{11}H_{10}N_2OS_2$	52.8	4.0	11.2	25.6
(yellow)				(250.33)	52.7	4.3	11.5	25.3
15	EtOH	143	83	$C_{18}H_{14}N_2OS_2$	63.9	4.1	8.2	18.9
(orange)				(338.45)	63.7	4.2	8.5	18.5
16	EtOH	232–5	79	$C_{11}H_{12}N_4OS$	53.2	4.8	22.5	12.9
(orange)				(248.30)	53.0	4.6	22.1	13.2
17	AcOH	66	69	$C_{14}H_9N_3S_2$	59.3	3.8	14.7	22.4
(yellow)				(283.41)	58.9	4.1	14.7	22.2

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TABLE II

I. R. and ^{1}H NMR data of the newly prepared compounds

Compd. No.	I.R. cm^{-1} selected bands	^{1}H NMR (δ ppm)
4	3420-3250 (2 NH_2), 2220, 2210 (2 CN), 1650 (C=C).	2.84 (s, 2H, NH_2), 3.48 (s, 2H, NH_2), 7.10-7.65 (m, 5H, C_6H_5).
7a	2220, 2215 (2 CN), 1210 (C=S).	3.28 (s, 1H, CH), 7.22-7.43 (m, 6H, C_6H_5, pyrimidine H-2).
7b	2220, 2215 (2 CN), 1630, 1655 (C=C, C=N).	3.44 (s, 1H, CH), 7.10-7.61 (m, 10H, 2 C_6H_5).
8a	3420-3330 (3 NH_2), 2220, 2210 (2 CN), 1620 (C=C).	2.45, 2.86, 3.28 (3s, 6H, 3 NH_2), 7.0-7.58 (m, 5H, C_6H_5).
8b	3410-3320 (NH_2, NH), 2220, 2210 (2 CN), 1625 (C=C).	2.84, 3.54 (2s, 4H, 2 NH_2), 7.10-7.62 (m, 10H, 2 C_6H_5), 9.21 (s, br, 1H, NH).
9a	3425, 3350 (NH_2), 2225, 2210 (2 CN), 1650 (C=N).	3.58 (s, 2H, NH_2), 4.21 (s, 1H, CH), 7.33-7.38 (m, 6H, C_6H_5, pyrimidine H-2).
9b	3520-3500 (NH), 2220, 2210 (2 CN), 1655 (C=N).	4.20 (s, 1H, CH), 7.32-7.40 (m, 11H, 2 C_6H_5, pyrimidine H-2), 9.10 (s, br, 1H, NH).
9c	3420, 3325 (NH_2), 2220, 2215 (2 CN), 1650 (C=N).	3.56 (s, 2H, NH_2), 4.10 (s, 1H, CH), 7.32-7.35 (m, 10H, 2 C_6H_5).
9d	3420 (NH), 2220, 2210 (2 CN), 1655 (C=N).	4.23 (s, 1H, CH_2), 7.31-7.39 (m, 15H, 3 C_6H_5), 8.56 (s, 1H, NH).
11	2225-2220 (3 CN), 1630, 1610 (C=N, C=C).	3.31 (s, 1H, CH), 7.21-7.39 (m, 6H, C_6H_5, pyridine H-2).
12	3580-3200 (OH, NH_2), 2220 (CN), 1680 (C=O), 1200 (C=S).	1.25 (t, 3H, J= 7.77 Hz, CH_3), 3.35 (s, 2H, NH_2), 4.25 (q, 2H, J= 7.77 Hz, CH_2), 7.23-7.50 (m, 5H, C_6H_5), 9.08 (s, br, 1H, OH).
13a	3450-3190 (2 NH_2, OH), 2220 (CN), 1650, 1630 (C=N, C=C).	1.23 (t, 3H, J= 7.86 Hz, CH_3), 2.84, 3.41 (2s, 4H, 2 NH_2), 4.22 (q, 2H, J= 7.86 Hz, CH_2), 7.32-7.38 (m, 5H, C_6H_5), 9.49 (s, 1H, OH).
13b	3520-3200 (NH_2, OH), 2220 (CN), 1655, 1630 (C=N, C=C).	1.25 (t, 3H, J= 7.91 Hz, CH_3), 3.51 (s, 2H, NH_2), 4.19 (q, 2H, J= 7.91 Hz, CH_2), 7.10-7.36 (m, 10H, 2 C_6H_5), 8.21 (s, 1H, NH), 9.34 (s, 1H, OH).
14	3400, 3620 (NH_2), 1680 (C=O), 1210 (C=S).	2.32 (s, 3H, CH_3), 4.21 (s, 2H, NH_2), 7.30-7.36 (m, 5H, C_6H_5).
15	3050 (aromatic CH), 2950 (CH_3), 1710 (C=O), 1210 (C=S).	2.21 (s, 3H, CH_3), 6.90 (s, 1H, CH), 7.21-7.40 (m, 10H, 2 C_6H_5).
16	3400-3200 (2 NH_2), 2950 (CH_3), 1680 (C=O).	2.22 (s, 3H, CH_3), 4.22 (s, 2H, NH_2), 5.31 (s, 2H, NH_2), 7.30-7.35 (m, 5H, C_6H_5).
17	3045 (CH aromatic), 2980 (CH_3), 2210 (CN), 1660 (C=N), 1210 (C=S).	1.99 (s, 3H, CH_3), 7.30-7.38 (m, 6H, C_6H_5), pyridine H-2).

Reaction of **4** with acrylonitrile afforded the thiazolo[4,5-b]pyridine derivative **11** which is formed via intermediate formation of the expected cyanoethylated product **10** followed by cyclization through loss of an ammonia molecule, then oxidation.

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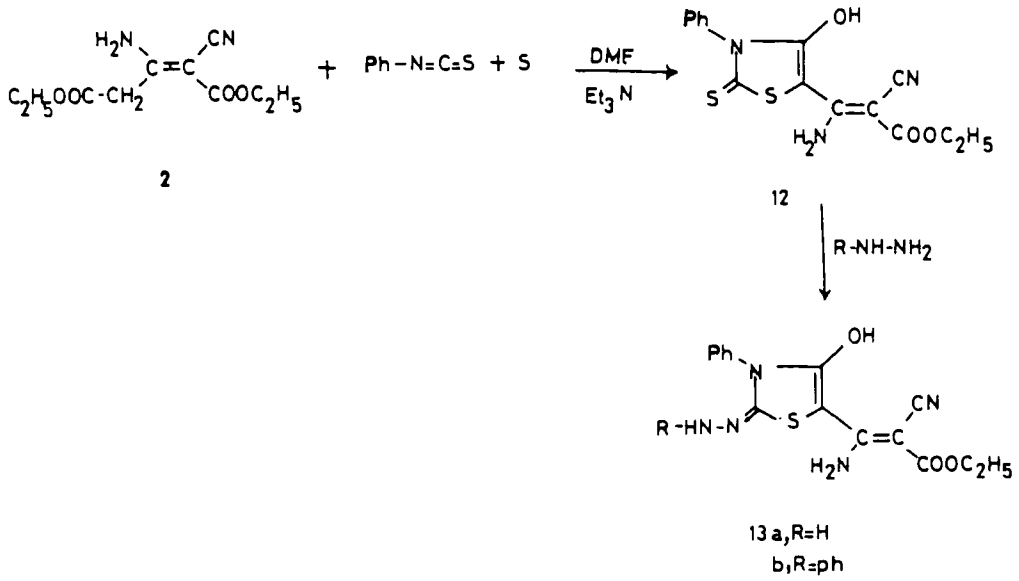
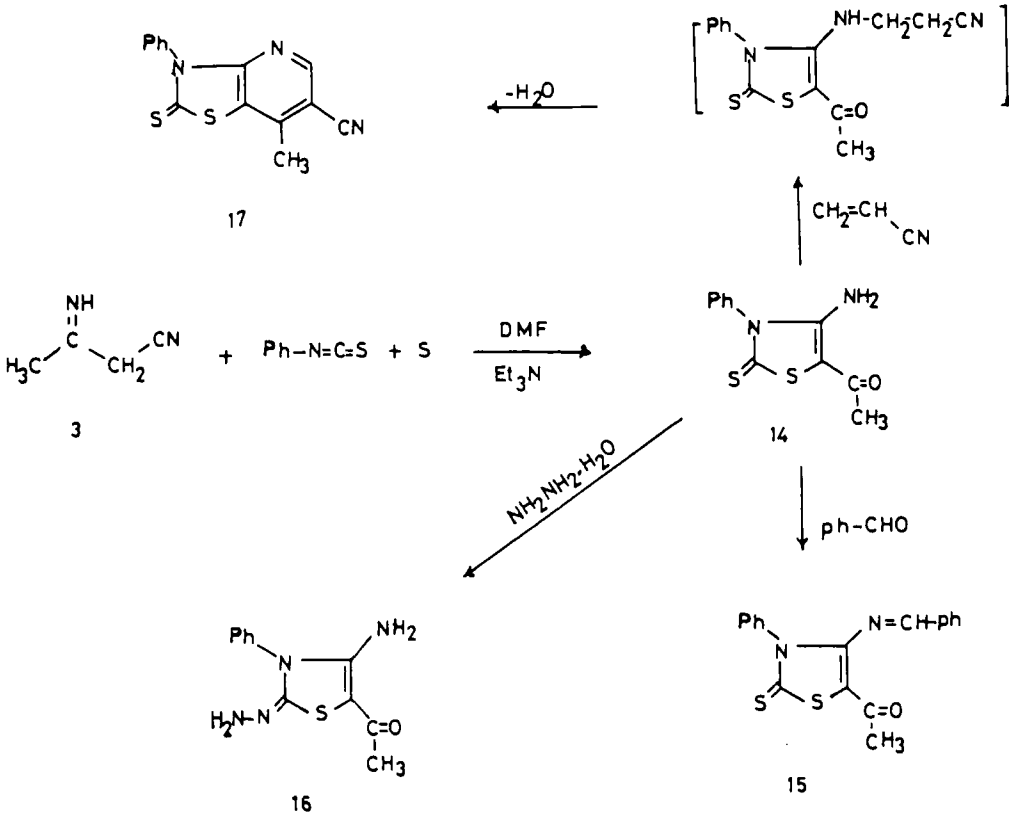


SCHEME II



SCHEME III

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Reaction of **2** with phenyl isothiocyanate and sulfur in refluxing dimethylformamide solution and the presence of triethylamine afforded similarly the thiazole derivative **12**. The structure of **12** was based on the analytical and spectral data. ^{1}H NMR spectrum revealed the presence of a triplet at δ = 1.25 ppm for CH_3 group, a quartet at δ = 4.25 ppm for CH_2), a singlet at δ = 3.35 ppm for the NH_2 group, a multiplet at δ = 7.23–7.50 ppm for C_6H_5 and a broad singlet at δ = 9.08 ppm for the OH group. Reaction of **12** with hydrazine hydrate and phenyl hydrazine gave the hydrazone derivatives **13a**,**b**.

The reaction of **3** with phenyl isothiocyanate and sulfur in refluxing dimethylformamide and the presence of triethylamine afforded the thiazole derivative **14**. Reaction of **14** with benzaldehyde afforded the Schiff's base **15**. Compound **14** reacted with hydrazine hydrate to give the thiazol-2-hydrazone derivative **16**. Structures of compounds **14–16** were confirmed based on analytical and spectral data (cf. Tables I and II).

Compound **14** reacted with acrylonitrile to afford the thiazolo[4,3-b]pyridine derivative **17**. The structure of **17** was established based on the IR spectrum which revealed the presence of one CN group stretching at 2210 cm^{-1} and ^{1}H NMR spectrum which revealed the presence of a singlet at δ = 1.99 ppm for CH_3 and a multiplet at δ = 7.30–7.38 ppm corresponding for C_6H_5 and the pyridine H-2 proton.

BIOLOGICAL ACTIVITY

The diverse biological activities of thiazoles and their fused derivatives promoted our attention to test and study the biological activities of some newly synthesized

TABLE III

In vitro bactericidal and fungicidal activity of some of the newly synthesized compounds

Compd. No.	Staph. albus	Staph. aureus	E. coli
4	+	+	+
7a	+	+	+
7b	+	++	+
8a	+	+	+
8b	+	-ve	++
12	+	+	++
13b	++	+++	+
14	++	++	++
15	++	+	++
16	+	-ve	++

Slight effect = +, Moderate effect = ++, Severe effect = +++

Rating percent control: No effect = 0; slight effect = 10, 20, 30; moderate effect = 40, 50, 60; severe effect = 70, 80, 90; complete effect = 100.

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products. The bactericidal and antifungal activities were studied. The antibacterial effect was determined using Gutter technique, while the antifungal effect was determined turbidimetric.[17,18] Table III shows that most of the tested compounds had high activity.

EXPERIMENTAL

All melting points are uncorrected. IR spectra were recorded (KBr) on a Pye Unicam SP-1000 spectrophotometer. ^{1}H NMR spectra were recorded on a Varian EM-390 MHz spectrometer with DMSO as solvent and TMS as internal reference. Chemical shifts are expressed as δ units (ppm). Analytical data were obtained from the Microanalytical Data Centre at Cairo University, Egypt.

4-Animo-5-(2′-amino-1′,1′-dicyanovinyl)-3-phenyl-thiazolin-2-thione (**4**); 5-(2′-amino-1′-cyano-1′-ethoxycarbonylvinyl)-4-hydroxy-3-phenyl-thiazolin-2-thione (**12**); 5-Acetyl-4-amino-3-phenyl-thiazol-2-thione (**14**).

General procedure: To a solution of each of **1** (0.01 mol), **2**, (0.01 mol) or **3** (0.01 mol) in dimethylformamide (30 ml) containing triethylamine (0.5 ml), phenyl isothiocyanate (0.01 mol) and sulfur element (0.01 mol) were added. The reaction mixture was heated under reflux for 3 h then was left at room temperature overnight. The solid product formed upon dilution with water was collected by filtration.

7-Dicyanomethino-3-phenyl-thiazolo[4,5-d]pyrimidin-2-thione (**7a**); 7-Dicyanomethino-3,5-diphenyl-thiazolo[4,5-d]pyrimidin-2-thione (**7b**); 5-Acetyl-4-benzalimino-3-phenyl-thiazol-2-thione (**15**).

General procedure: A solution of each of **4** (0.01 mol) or **14** (0.01 mol) in dimethylformamide (30 ml) containing triethylamine (0.5 ml), each of formaldehyde or benzaldehyde (0.01 mol) was added. The reaction mixture in each case was heated under reflux for 3 h. The solid product formed upon dilution with water containing few drops of hydrochloric acid was collected by filtration.

7-Dicyanomethino-2-hydrazono-3-phenyl-thiazolo[4,5-d]pyrimidine (**9a**); 7-Dicyanomethino-3-phenyl-2-phenylhydrazono-thiazolo[4,5-d]pyrimidine (**9b**); 7-Dicyanomethino-3,5-diphenyl-2-hydrazono-thiazolo[4,5-d]pyrimidine (**9c**); and 7-Dicyanomethino-3,5-diphenyl-2-phenylhydrazono-thiazolo[4,5-d]pyrimidine (**9d**).

General procedure: METHOD (A). To a solution of **7a** or **7b** (0.01 mol) in dimethylformamide (50 ml) each of hydrazine hydrate or phenyl hydrazine (0.01 mol) was added. The reaction mixture was heated under reflux for 4 h. The solid product formed upon pouring into ice/water containing few drops of hydrochloric acid was collected by filtration.

METHOD (B). To a solution of each of **8a** or **8b** (0.01 mol) in dimethylformamide (50 ml) containing piperidine (0.5 ml) each of formaldehyde (0.01 mol) or benzaldehyde (0.01 mol) was added. The reaction mixture was heated under reflux for 3 h then evaportated in vacuo. The remaining product was triturated with ethanol then collected by filtration.

4-Amino-5-(2′-amino-1′,1′-dicyanovinyl)-2-hydrazono-3-phenyl-thiazoline (**8a**); 4-Amino-5-(2′-amino-1′,1′-dicyanovinyl)-2-phenylhydrazono-3-phenyl-thiazoline (**8b**); 5-(2′-Amino-1′-cyano-1′-ethoxycarbonylvinyl)-4-hydroxy-2-hydrazono-3-phenyl-thiazoline (**13a**); 5-(2′-Amino-1′-cyano-1′-ethoxycarbonylvinyl)-4-hydroxy-2-phenylhydrazono-3-phenyl-thiazoline (**13b**); and 5-Acetyl-4-amino-2-hydrazino-3-phenyl-thiazoline (**16**).

General procedure: To a suspension of each of **4** (0.01 mol) or **12** (0.01 mol) or **14** (0.01 mol) in dimethylformamide (30 ml), hydrazine hydrate (0.01 mol) or phenyl hydrazine (0.01 mol) was added. The reaction mixture was heated under reflux for 3 h then poured into ice/water mixture and the solid product formed was collected by filtration.

6-Cyano-7-dicyanomethino-3-phenyl-thiazolo[4,5-b]pyridin-2-thione (**11**); 6-Cyano-3-phenyl-7-methyl-thiazolo[4,5-b]pyridin-2-thione (**17**).

General procedure: To a solution of each of **4** (0.01 mol) or **14** (0.01 mol) in pyridine (30 ml) containing water (5 ml), acrylonitrile (0.01 mol) was added. The reaction mixture was heated under reflux for 3 h. The solid product formed upon pouring into ice/water mixture containing few drops of hydrochloric acid was collected by filtration.

Procedure of biological tests: The newly synthesized compounds were tested against the specified microorganism as 400 ug/ml (w/v) solution in sterile DMSO. A solution of the tested compound (0.1 mol)

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was poured aseptically in a well of g diameter made by a borer in the seeded agar medium. After pipetting the same volume in wells of all tested microorganisms, plates were incubated after 37°C for 24 h. The activities were expressed as inhibition zones (mm diameter, clear areas) as antibacterial and antifungal effect, were measured to the nearest 0.5 mm. The least concentration which showed inhibitory effect on any specific microorganism was considered as the minimum inhibitory concentration (MIC) which was determined using Streptomycin and Mycostatin as the references.

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