REACTION PRODUCTS OF THIOSEMICARBAZIDE WITH 1, 5-DIARYLPENT-1-YNE-3, 5-DIONES

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SUMMARY: Acetylenic ß-diketones reacted with thiosemicarbazide to give either 6H or 3H, 6H-pyrazolo [1, 5-c] pyrimidinethiones depending on the nature of the substituents as well as the reaction conditions. The reaction of 2-aryl-5-phenyl-6H-pyrazolo [1, 5-c] pyrimidine-7-thiones with nitrous acid and benzenediazonium chloride gave the corresponding disulfide and phenylazo disulfide derivatives, respectively. With certain electrophiles, 3-substituted-7-thiones are formed. Moreover, oxidation with alkaline hydrogen peroxide afforded the respective pyrazolopyrimidinones.

Key Words: Thiosemicarbazide, 5-diarylpenth-1-yne-3, 5-diones.

INTRODUCTION

1, 5-diarylpent-1-yne-3, 5-diones are polyfunctional reagents which were found to give rise to a great variety of five and six-membered heterocyclic rings (1).

The versatility of these substrates led us to suppose that the polyfunctionalized reagents would constitute suitable building blocks for the synthesis of biheterocyclic compounds which have a great variety of useful properties.

In this paper, we would like to report the reactivity of 1, 5-diarylpent-1-yne-3, 5-diones 1 with thiosemicarbazide. Starting materials 1 are easily available from condensation of acetylenic esters with suitable ketones, as described previously (2, 3).

Thus, the reaction of compounds 1a-c with thiosemicarbazide in ethanol at room temperature in the presence of concentrated hydrochloric acid or thiosemicarbazide hydrochloride afforded the respective 2-aryl-5-phenyl-6H-pyrazolo [1, 5-c] pyrimidine-7-thiones 4a-c as only separated product in high yields in a one step procedure. However, under identical conditions 1d, e gave 4d, e alongside with their isomers 6d, e as minor products (Scheme 1).

On the other hand, 2-phenyl-5-aryl-6H-pyrazolo [1, 5-c]-pyrimidine-7-thiones 6b, c were obtained by the reaction of 1b, c with thiosemicarbazide in refluxing ethanol. However, 5-furyl-9-hydroxy-2-phenyl-3H, 6H-pyrazolo[1, 5-c] pyrimidine-7-thione 5 was formed from 1f under the same conditions. The latter 5 could be converted into its thione 6f on heating in xylene (Scheme 1).

Thiosemicarbazide can attack the keto group or the acetylenic linkage of 1. There are indications that these paths are competitive, but in acid medium, the hydrazone 2 is favored, while in neutral medium, an amine-type intermediate 3 is formed as described in the Scheme 1. Both intermediates may cyclize under the reaction conditions giving pyrazolopyrimidines 4 and 6, respectively.

The above reaction provide new routes of synthesis of pyrazolo [1, 5-c] pyrimidine thiones containing nuclear aryl groups of which only few examples are reported (4, 5); however, some of them are known as bactericides (5).

In agreement with the assigned structures, the infrared spectra of the pyrazolopyrimidines 4 and 6 gave characteristic pyrazolo and pyrimidine C=N ring bands at 1569-1665 cm^{-1} and pyrimidine C=C band at 1520-1548 cm^{-1}, a thiocarbonyl and NH absorptions at 1074-1217 and 3122-3459 cm^{-1}, respectively. It is noted that the lack of υSH absorption in their spectra may suggest that in the solid state the thione form predominants.

The ^{1}H nmr spectra of 4 and 6 (Table 1), showed two singlets at δ 6.65-7.10 and 6.93-7.28 for H-3 and H-4 ring protons. Moreover, besides the NH proton in the case of 4d, e and 6d, e a singlet at δ 3.37-4.43 was observed which may suggest some contribution of the thiol form (1:3).

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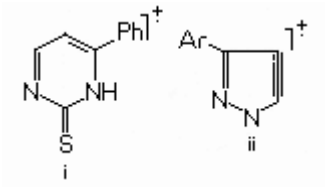
Further support of the structure of the pyrazolopyrimidinethiones was obtained from their mass spectra. These compounds gave their molecular ions as the base or very intense peaks which gave rise to the pyrimidine radical cation i and the pyrazolyne species ii characteristic for the pyrazolopyrimidine system (6).

While the chemistry of the increasingly important pyrazolo [1, 5-c] pyrimidines have been very little explo-



i ii

red, the other pyrazolopyrimidines have received considerable attention (7). In the present work, oxidation of 2-aryl-5-phenyl-6H-pyrazolo [1, 5-c] pyrimidinethiones 4a-e with sodium nitrite in glacial acetic acid gave the respective 7, 7`-bis-(2-aryl-5-phenylprazolo [1, 5-c] pyrimidinyl) disulfides 7a-e similar to that reported for 4, 6-diarylpyrimidine-2 (1H)-thiones (8). On the other hand, treatment of 4a-e with benzenediazonium chloride in the presence of sodium hydroxide gave the 3-phenylazo disulfide derivatives 8a-e. The reaction is assumed to proceed by introduction of phenylazo group as well as oxidation of the thiol under the action of nitrous acid already present in the reaction medium (Scheme 1).

No reports on the electrophilic substitution reactions of pyrazolo [1, 5-c] pyrimidines have been made earlier. However, the reaction of pyrazolo [1, 5-a] pyrimidines with the same regents generally leads to the formation of their 3-or 3, 6-disubstituted derivatives (7, 9). In the present work, bromination and iodination of 4a-e with bromine and iodine monochloride in chloroform afforded the corresponding 3-bromo 9a-e and 3-iodo 10a-e derivatives, respectively. Moreover, nitration of 4a-e with nitric and sulfuric acids in glacial acetic acid led to the formation of the corresponding 3-nitro 11a-e derivatives (Scheme 2). The structure of the above products was confirmed from their spectral data which included infrared, ^{1}H nmr spectra.

It is worthy to note that the detection of the nitropyrazole species at m/e 265 in the mass spectrum of 11b, while no nitropyrimidine fragment appeared supports the introduction of the nitro group at position 3 in the parent pyrazolopyrimidinethiones.

3-Cyano-5, 7-dimethylpyrazolo [1, 5-a] pyrimidine is susceptible to pyrimidine ring opening and subsequent recyclization with rearrangement by alkaline hydrogen peroxide leading to 4-hydroxy-6-methyl-1H-pyrazolo [3, 4-d] pyrimidine (10), while the pyrazolo [1, 5-c] pyrimidinethiones 4a-e under the same reagent gave the respective pyrazolo [1, 5-c] pyrimidin-7-ones 12a-e in high yields (Scheme 2). The presence of a broad OH absorption in the infrared spectra of 12 may suggest that in the solid state a significant contribution of the enol form takes place.

EXPERIMENTAL

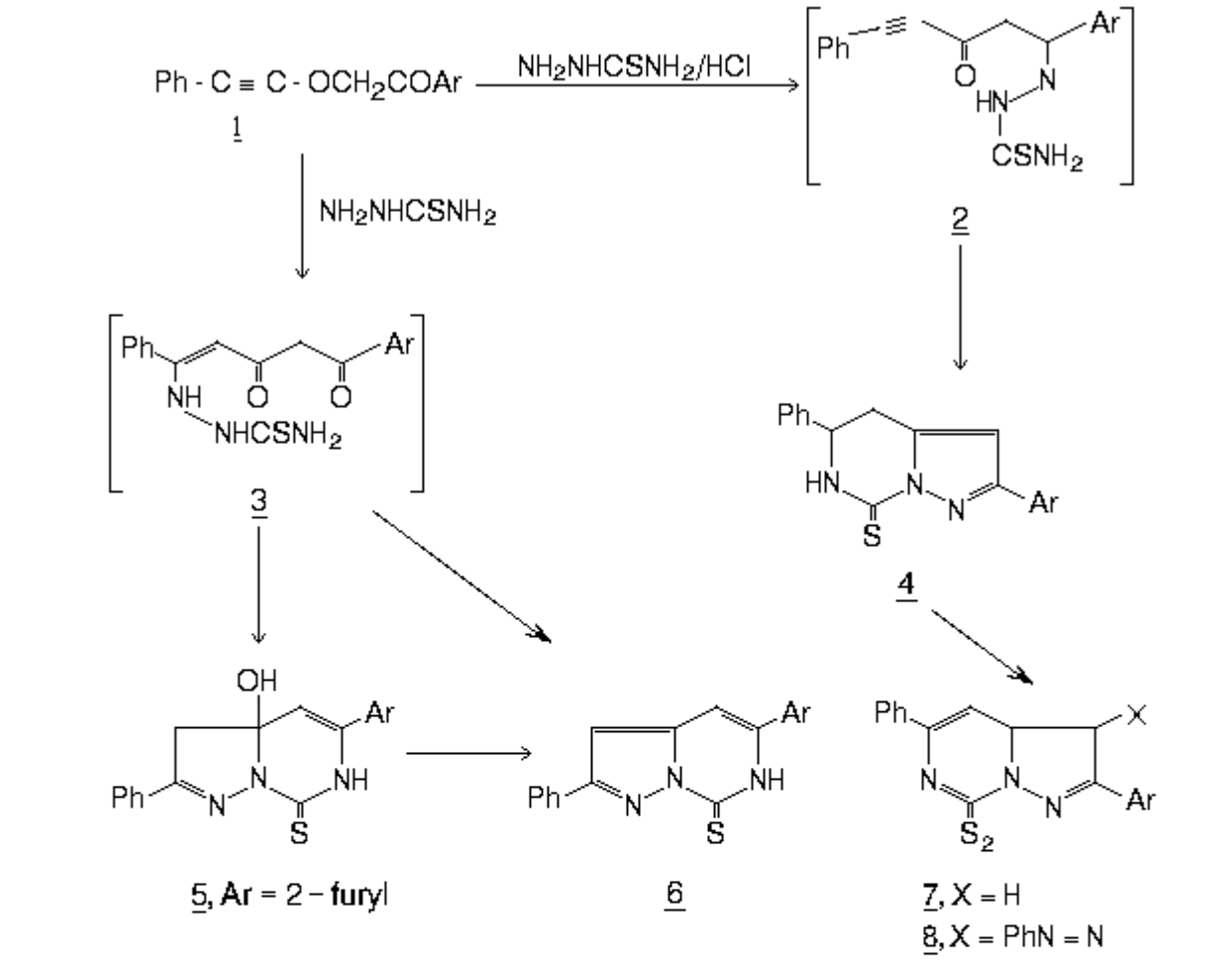
Microanalyses were performed by Microanalysis Unit, Cairo University, Cairo. Infrared spectra were measured with a Unicam SP 1025 spectrophotometer for potassium bromide pellets. The ^{1}H nmr spectra were recorded on a Varian EM-390 nmr spectrometer at 90 MHz with TMS as internal standard. Mass spectra were recorded at 70 eV with an AEL MS-9 spectrometer coupled to a DS-50 Data system using a direct insertion probe for introduction of samples.

2-Aryl-5-phenyl-6H-pyrazolo [1, 5-c] pyrimidine-7-thiones 4 (Table 1)

A solution of 1, 5-diarylpent-1-yne-3, 5-diones 1a-e (0.0018 mole) in ethanol (20 ml) was stirred with thiosemicarbazide (0.0018 mole) in the presence of hydrochloric acid (1 ml) or thiosemicarbazide hydrochloride (0.0022 mole) for 6 hours at room temperature. The pyrazolopyrimidinethione 4a-c which separated was crystallized from methanol as pale yellow needles, whereas 4d, e was separated from its isomer 6d, e by fractional crystallization from chloroform; ms: m/e (relative abundance) 4a: M^+ 303 (100), 302 (99), 187 (10), 141 (14); 4b: M^+ 381 (80), 380 (100), 219 (26), 187 (18), 77 (50); 4c: M^+ 337 (100), 336 (85), 187 (22), 175 (35), 77 (35); 6d: M^+ 317 (89), 316 (70), 210 (30), 141 (16), 77 (100).

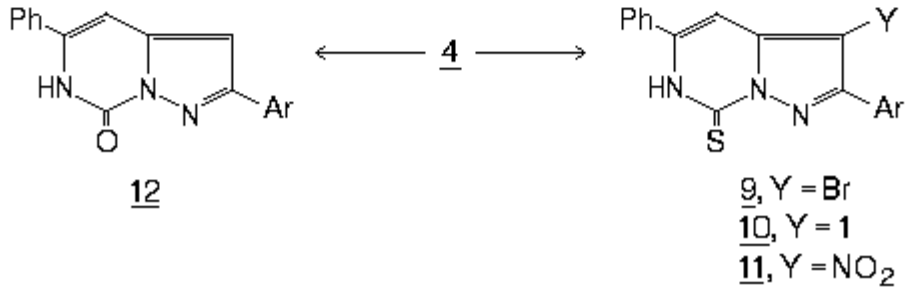
5-Furyl-9-hydroxy-2-phenyl-3H, 6H-pyrazolo [1, 5-c] pyrimidine-7-thione 5 and 5-Aryl-2-phenyl-6H-pyrazolo [1, 5-c] pyrimidine-7-thiones 6 (Table 1)

A solution of 1a-c, f (0.0021 mole) in ethanol (20 ml) was refluxed with thiosemicarbazide (0.0023 mole) for three hours. The thione 5 and 6a-c which separated were crystallized from methanol in yellow needles; ir (υmax, cm^{-1}) 5: 1074 (C=S), 1518 (pyrimidine C=C), 1595-1629 (two C=N), 3423 (OH).



Ar : a = Ph, b = P - Br - C_6H_4, c = p - Cl - C_6H_4, d = p-Me - C_6H_4, e = p-MeO - C_6H_4, f = 2 - fury

Scheme 1



Scheme 2

Table 1: Characterization of Pyrazolopyrimidines Derivates 4-12.

Comp.	M.P. (°C)	Yield (%)	Molecular Formula	Analysis Calcd./Found					^{1}H-NMR, DMSO-d_6, (δ/ppm)*
				C	H	N	S	X	
4a	236-238	65	$C_{18}H_{13}N_3S$	71.3 71.1	4.3 4.2	13.9 14.1	10.6 10.4		7.10-7.23 (s,2H,$H_{3,4}$), 13.20 (s,1H,NH)
4b$^+$	146-148	81	$C_{18}H_{12}BrN_3S$	56.7 56.9	3.1 3.4	11.0 10.7	8.4 8.1	20.7 20.8	6.65-7.20 (s,2H,$H_{3,4}$), 3.39 (s,1H,SH)
4c$^+$	179-180	78	$C_{18}H_{12}ClN_3S$	64.0 64.1	3.6 3.9	12.5 12.3	9.5 9.7	10.4 10.1	6.70-7.28 (s,2H,$H_{3,4}$)
4d$^+$	191-192	71	$C_{19}H_{15}N_3S$	71.9 71.6	4.7 4.4	13.2 13.3	10.1 10.3		6.73-7.07 (s,2H,$H_{3,4}$), 3.37 (s,1H,SH), 2.33 (s,3H,CH_3)
4e$^+$	202-204	68	$C_{19}H_{15}N_3OS$	68.5 68.4	4.5 4.2	12.6 12.7	9.6 9.8		6.73-6.93 (s,2H,$H_{3,4}$), 4.20 (s,1H,SH), 3.73 (s,3H,CH_3)
5	160-163	75	$C_{16}H_{13}N_3O_2S$	61.7 61.2	4.2 3.9	13.5 13.1	10.3 9.9		7.08 (s,1H,H_4), 4.36 (s,2H,CH_2), 8.46 (s,1H,OH)
6b	216-218	80	$C_{18}H_{12}BrN_3S$	56.7 56.4	3.1 2.9	11.0 10.8	8.4 8.1	20.7 20.3	6.88-7.20 (s,2H,$H_{3,4}$), 8.40 (s,1H,NH)
6c	228-231	82	$C_{18}H_{12}ClN_3S$	64.0 64.2	3.6 3.6	12.5 12.3	9.5 9.3	10.4 10.1	7.05-7.18 (s,2H,$H_{3,4}$), 12.65 (s,1H,NH)
6d	269-270	26	$C_{19}H_{15}N_3S$	71.9 71.6	4.7 4.6	13.2 12.9	10.1 10.3		7.07-7.13 (s,2H,$H_{3,4}$), 12.97 (s,1H,NH), 2.33 (s,3H,CH_3)
6e	287-290	23	$C_{19}H_{15}N_3OS$	68.5 68.1	4.5 4.3	12.6 12.6	9.6 9.8		7.03-7.28 (s,2H,$H_{3,4}$), 13.13 (s,1H,NH), 3.90 (s,3H,OCH_3)
6f	208-210	66	$C_{16}H_{11}N_3OS$	65.5 65.1	3.8 3.3	14.3 13.6	10.9 10.7		6.72-7.18 (s,2H,$H_{3,4}$), 4.43 (s,1H,SH)
7a	284-286	76	$C_{36}H_{24}N_6S_2$	71.5 71.3	3.9 3.8	13.9 13.6	10.9 10.8		7.32-7.80 (m,24H,ArH+$H_{3,4}$)
7b	265-267	73	$C_{36}H_{22}Br_2N_6S_2$	56.8 56.6	2.9 2.7	11.0 11.2	8.4 8.2	20.8 20.6	7.28-7.76 (m,22H,ArH+H)
7c	283-285	71	$C_{36}H_{22}C_{12}N_6S_2$	64.2 64.1	3.3 3.2	12.5 12.2	9.5 9.8	10.4 10.2	7.24-7.86 (m,22H,ArH+$H_{3,4}$)
7d	222-224	70	$C_{36}H_{28}N_6S_2$	72.2 72.3	4.4 4.3	13.3 13.1	10.1 10.3		7.32-7.90 (m,22H,ArH+$H_{3,4}$), 2.23 (s,6H,2CH_3)

Table 1: Characterization of Pyrazolopyrimidines Derivates 4-12 (Cont.).

Comp.	M.P. (°C)	Yield (%)	Molecular Formula	Analysis Calcd./Found					^{1}H-NMR, DMSO-d_6, (δ/ppm)*
				C	H	N	S	X	
7e	236-237	77	$C_{38}H_{28}N_6O_2S_2$	68.7 68.5	4.2 4.0	12.7 12.3	9.6 9.6		7.29-7.84 (m,22H,ArH+$H_{3,4}$), 4.05 (s,6H,20CH_3)
8a	110-112	73	$C_{48}H_{32}N_{10}S_2$	70.9 71.1	3.9 3.7	17.2 17.4	7.9 7.8		7.24-7.92 (m,32H,ArH+$H_{3,4}$)
8b	118-120	74	$C_{48}H_{30}Br_2N_{10}S_2$	59.5 59.1	3.1 3.3	14.5 14.4	6.6 6.3	16.3 16.2	7.21-7.88 (m,30H,ArH+$H_{3,4}$)
8c	128-131	76	$C_{48}H_{30}C_{12}N_{10}S_2$	65.4 65.2	3.4 3.4	15.9 15.6	7.3 7.1	7.9 7.8	7.28-7.94 (m,30H,ArH+$H_{3,4}$)
8d	162-163	82	$C_{50}H_{36}N_{10}S_2$	71.4 71.1	4.3 4.1	16.7 16.3	7.6 7.4		7.22-7.96 (m,30H,ArH+$H_{3,4}$), 2.43 (s,6H,2CH_3)
8e	142-144	80	$C_{60}H_{36}N_{10}O_2S_2$	68.8 68.6	4.1 4.3	16.1 16.3	7.3 7.1		7.24-7.96 (m,30H,ArH+$H_{3,4}$), 3.74 (s,6H,20CH_3)
9a	236-238	78	$C_{18}H_{12}BrN_3S$	56.5 56.1	3.1 3.2	10.9 10.7	8.4 8.1	20.9 20.6	7.07 (s,1H,+H_4), 7.18-7.80 (m,10H,ArH)
9b	143-145	77	$C_{18}H_{11}Br_2N_3S$	47.0 46.8	2.4 2.1	9.1 9.3	7.0 7.3	34.4 34.1	6.82 (s,1H,H_4), 7.16-7.82 (m,9H,ArH)
9c	162-165	81	$C_{18}H_{11}BrClN_3S$	52.0 51.9	2.6 2.5	10.1 10.3	7.7 7.5	19.0-8.4 18.8-8.1	7.02 (s,1H,H_4), 7.19-7.86 (m,9H,ArH)
9d	215-217	73	$C_{19}H_{14}BrN_3S$	57.7 57.3	3.5 3.4	10.6 10.3	8.1 8.2	20.0 19.8	7.06 (s,1H,H_4), 2.98 (s,3H,CH_3), 7.14-7.86 (m,9H,ArH)
9e	196-198	75	$C_{19}H_{14}BrN_3OS$	55.5 55.3	3.4 3.2	10.2 10.1	7.8 7.6	19.2 19.3	6.96 (s,1H,H_4), 3.96 (s,3H,OCH_3), 7.18-7.88 (m,9H,ArH)
10a	262-264	78	$C_{18}H_{12}IN_3S$	50.3 50.6	2.8 2.6	9.8 9.7	7.5 7.1	29.6 29.3	7.00 (s,1H,H_4), 7.16-7.89 (m,10H,ArH)
10b	166-168	73	$C_{18}H_{11}BrIN_3S$	42.6 42.3	2.2 2.1	8.3 8.3	6.3 6.6	15.6-25.0 15.2-24.7	6.86 (s,1H,H_4), 7.12-7.82 (m,9H,ArH)
10c	181-182	78	$C_{18}H_{11}ClIN_3S$	46.6 46.3	2.4 2.3	9.1 8.9	6.9 6.8	7.6-27.4 7.3-27.7	6.82 (s,1H,H_4), 7.21-7.89 (m,9H,ArH)
10d	168-170	80	$C_{19}H_{14}IN_3S$	51.5 51.3	3.2 3.3	9.5 9.3	7.2 7.4	28.7 28.3	7.06 (s,1H,H_4), 7.18-7.89 (m,9H,ArH), 2.42 (s,3H,CH_3)

Table 1: Characterization of Pyrazolopyrimidines Derivates 4-12 (Cont.).

Comp.	M.P. (°C)	Yield (%)	Molecular Formula	Analysis Calcd./Found					^{1}H-NMR, DMSO-d_6, (δ/ppm)*
				C	H	N	S	X	
10e	201-203	83	$C_{19}H_{14}IN_3OS$	49.7 49.4	3.1 2.8	9.1 9.3	7.0 7.3	27.6 27.4	7.10 (s,1H,H_4), 7.21-7.88 (m,9H,ArH), 3.52 (s,3H,OCH_3)
11a	273-275	81	$C_{18}H_{12}N_4O_2S$	62.1 62.4	3.4 3.2	16.1 16.3	9.2 9.4		6.96 (s,1H,H_4), 7.14-7.88 (m,10H,ArH)
11b	178-180	71	$C_{18}H_{11}BrN_4O_2S$	50.7 50.4	2.6 2.3	13.1 12.9	7.5 7.6	18.5 18.3	6.64 (s,1H,H_4), 7.21-7.86 (m,9H,ArH)
11c	203-205	73	$C_{18}H_{11}ClIN_4O_2S$	56.5 56.3	2.9 2.7	14.6 14.3	8.4 8.2	9.2 9.1	6.82 (s,1H,H_4), 7.28-7.88 (m,9H,ArH)
11d	286-288	76	$C_{19}H_{14}N_4O_2S$	62.9 62.7	3.9 3.6	15.5 15.2	8.8 8.6		7.00 (s,1H,H_4), 7.23-7.89 (m,9H,ArH), 2.34 (s,3H,CH_3)
11e	231-233	74	$C_{19}H_{14}N_4O_3S$	60.3 60.6	3.7 3.6	14.8 14.4	8.5 8.3		6.88 (s,1H,H_4), 7.22-7.89 (m,9H,ArH), 3.46 (s,3H,OCH_3)
12a	203-206	71	$C_{18}H_{13}N_3O$	75.3 75.3	4.5 4.1	14.6 14.3			7.20-7.34 (s,2H,$H_{3,4}$), 7.26-7.88 (m,10H,ArH)
12b	215-217	73	$C_{18}H_{12}BrN_3O$	59.2 59.1	3.3 3.6	11.5 11.2	21.6 21.3		6.88-7.14 (s,2H,$H_{3,4}$), 7.22-7.86 (m,9H,ArH)
12c	226-228	70	$C_{18}H_{12}ClN_3O$	67.3 67.1	3.7 3.4	13.1 12.8	10.9 10.7		6.82-7.08 (s,2H,$H_{3,4}$), 7.22-7.88 (m,9H,ArH)
12d	328-329	78	$C_{19}H_{15}N_3O$	75.7 75.3	4.9 4.6	13.9 13.7			6.73-6.80 (s,2H,$H_{3,4}$), 7.21-7.82 (m,9H,ArH) 2.30 (s,3H,CH_3)
12e	290-291	81	$C_{19}H_{15}N_3O_2$	71.9 71.8	4.7 4.8	13.2 13.3			6.70-6.88 (s,2H,$H_{3,4}$), 7.24-7.89 (m,9H,ArH) 3.78 (s,3H,CH_3)

* s: singlet; m: multiplet..

All NH, SH and OH signals were exchangeable with deuterium oxide.

+: Spectra were carried out in $CDCl_3$.

5-Furyl-2-phenyl-6H-pyrazolo [1,5-c]pyrimidine-7-thione 6f (Table 1)

A suspension of 5 (0.0009 mole) in xylene (15 ml) was refluxed for six hours. The thione 6f which formed after concentration was crystallized from methanol in orange needles.

7,7`-Bis (2-aryl-5-phenyl-6H-pyrazolo [1, 5-c] pyrimidinyl) Disulfides 7 (Table 1)

A solution of 4a-e (0.0021 mole) in glacial acetic acid (20 ml) was treated portion wise with 25% aqueous solution of sodium nitrite (10 ml). The mixture was heated on a boiling water bath with stirring for 20-30

minutes, whereby a yellow solid started to separate. The reaction mixture was then diluted with water and the precipitated 7a-e was filtered and crystallized from benzene in yellow needles; ir (υmax, cm^{-1}): 1480-1525 (pyrimidine C=C), 1585-1636 (two C=N).

7, 7` -Bis (2-aryl-3-phenylazo-5-phenylpyrazolo [1, 5-c] pyrimidinyl) Disulfides 8 (Table 1)

An aqueous solution of sodium hydroxide (10 ml, 10%) was added to a suspension of 4a-e (0.0018 mole) in ethanol (15 ml). The reaction mixture was gradually treated with a solution of benzenediazonium chloride (prepared from 1 ml of aniline) at 5° with stirring for one hour. The disulfides 8a-e so formed, were collected by filtration and crystallized from ethanol in reddish brown needles; ir (υmax, cm^{-1}): 1485-1525 (pyrimidine C=C), 1580-1655 (two C=N).

2-Aryl-3-halo-5-phenyl-6H-pyrazolo [1, 5-c] pyrimidine-7-thiones 9 and 10 (Table 1)

A solution of bromine (0.0019 mole) or iodine monochloride (0.0022 mole) in chloroform (15 ml) was gradually added to a suspension of 4a-e (0.0018 mole) in chloroform (15 ml) with stirring for 20-40 minutes at room temperature. The precipitated 3-halo thione 9a-e or 10a-e was filtered, washed with methanol, dried and crystallized from benzene or chloroform-methanol in yellow needles; ir (υmax, cm^{-1}): 1082-1228 (C=S), 1485-1525 (pyrimidine C=C), 1585-1620 (two C=N), 3180-3345 (NH).

2-Aryl-3-nitro-5-phenyl-6H-pyrazolo [1, 5-c] pyrimidine-7-thiones 11 (Table 1)

A mixture of nitric (d 1.41; 1 ml) and sulfuric (d 1.84; 1 ml) acids in glacial acetic acid (10 ml) was gradually added to a solution of 4a-e (0.002 mole) in glacial acetic acid (10 ml) with stirring for three hours at room temperature. The reaction mixture was then poured into cold water with stirring and the precipitated 3-nitro derivative 11a-e was filtered, washed with water, dried and crystallized from ethanol in yellow needles; ir (υmax, cm^{-1}): 1025-1218 (C=S), 1330-1352 and 1515-1540 (NO_2), 1495-1525 (pyrimidine C=C), 1575-1649 (two C=N), 3174-3440 (NH). MS: m/e (relative abundance) 11b: M^+ 426 (15), 425 (22), 265 (21), 219 (20), 187 (60), 155 (30), 77 (100).

2-Aryl-5-phenyl-6H-pyrazolo [1, 5-c] pyrimidin-7-ones 12 (Table 1)

A mixture of 4a-e (0.0015 mole), 30% hydrogen peroxide (4 ml) and 10% aqueous sodium hydroxide (18 ml) was heated on a boiling water bath for 2-3 hours. The pH of the resulting solution was adjusted to 6 by addition of concentrated hydrochloric acid. The precipitated ketone 12a-e was washed several times with water, dried and crystallized from ethanol in pale yellow needles; ir (υmax, cm^{-1}): 1502-1525 (pyrimidine C=C), 1582-1636 (two C=N), 1690-1720 (CO), 3385-3450 (OH).

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