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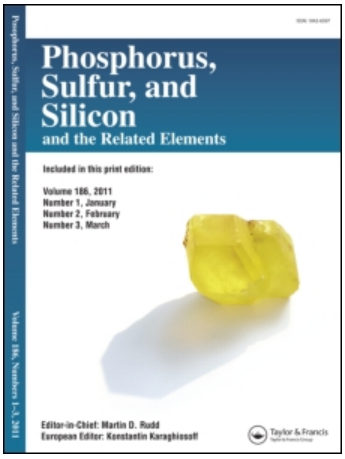

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SYNTHESIS OF DIARYLSULPHIDES AND DIARYLSULPHONES CONTAINING PYRAZOLINE, ISOXAZOLINE, PYRIMIDINE AND CONDENSED PYRIDAZINE MOIETIES

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Arylhydrazones **2a–c** obtained by the reaction of diazotized 4-amino-4′-nitrodiaryl sulphide (**1**) with active methylene compounds i.e. ethyl acetoacetate, acetylacetone and diethyl malonate are condensed with hydrazines, hydroxylamine, urea and thiourea to give the corresponding pyrazolines (**3, 4, 8, 9, 13**), isoxazolines (**5, 10**) and pyrimidines (**6, 7, 11, 12, 14, 15**). Intramolecular cyclization of arylhydrazones **2a, c** with $AlCl_3$ in chlorobenzene yielded the cinnoline derivatives (**16, 17**). Reaction of cinnoline derivative (**16**) with hydrazines afforded the corresponding pyrazolo[4,3-c]cinnoline derivatives (**18, 19**). Oxidation of some of the prepared sulphides with H_2O_2/AcOH at room temperature gave the corresponding sulphones (**20–30**).

Key words: Synthesis; diarylsulphides; diarylsulphones; pyrazoline; pyrimidine; cinnoline.

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INTRODUCTION

Literature reports the synthesis and utility of substituted diaryl sulphides as antileprotic, nematocidal, bactericidal[1,2] and insecticidal activities.[3] Also, many pyrazoles are used as therapeutic agents,[4,5] pyrimidines as antitumor agents[6] and analgesics[7] and isoxazoles possess antituberculosis[8] and antileprous activities.[9] In the present investigation we report convenient routes for the synthesis of diaryl sulphides and sulphones containing pyrazoline, isoxazoline and pyrimidine moieties.

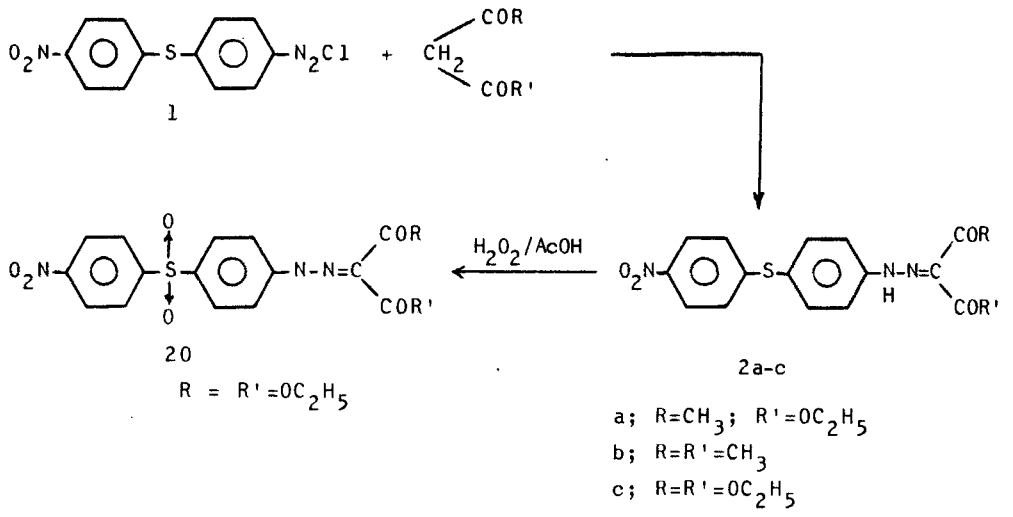
In the first approach the diazonium salt of 4-amino-4′-nitrodiaryl sulphide[10] (**1**) was coupled with active methylene compounds namely, ethyl acetoacetate, acetylacetone and diethyl malonate in aqueous alcoholic sodium acetate solution to give the corresponding arylhydrazones (**2a–c**).

Condensation of arylhydrazone (**2a**) with hydrazine hydrate, phenylhydrazine, hydroxylamine, urea and thiourea gave respectively pyrazolines (**3, 4**) isoxazoline (**5**) and pyrimidine derivatives (**6, 7**).

Similar reactions of arylhydrazone (**2b**) with hydrazines, hydroxylamine, urea and thiourea yielded the corresponding pyrazole (**8, 9**), isoxazole (**10**) and pyrimidine derivatives (**11, 12**).

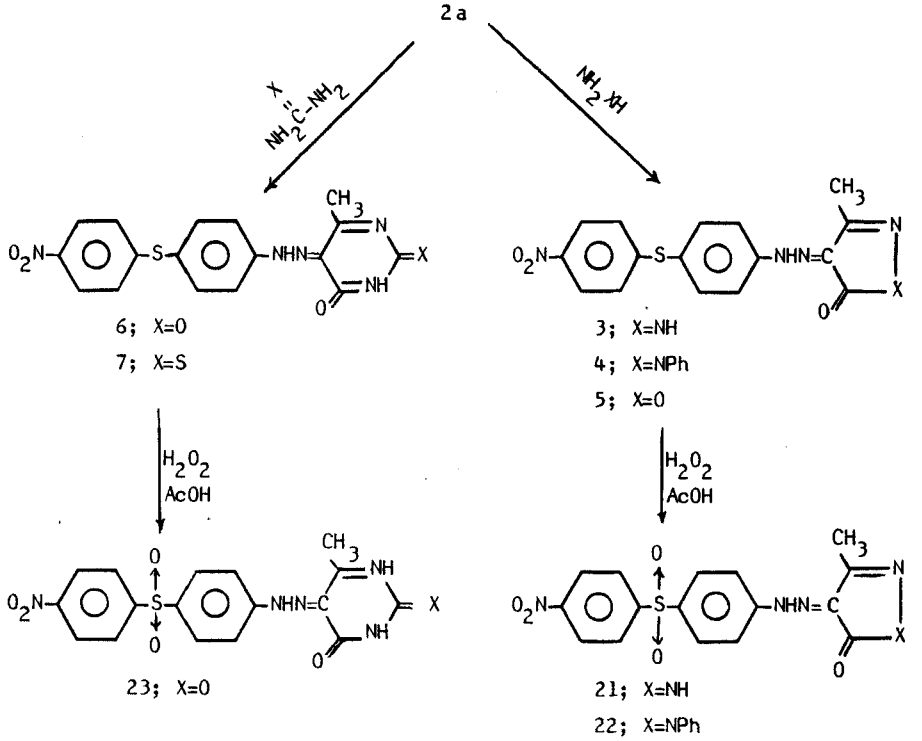
Arylhydrazone (**2c**) also was allowed to react with hydrazine-hydrate, urea and thiourea to give the corresponding pyrazoline (**13**) and pyrimidine derivatives (**14, 15**).

The chemical structure of compounds **2–15** was confirmed on the basis of elemental analyses, IR and H^1NMR (cf. experimental section).



On the other hand, condensed heterocycles to diaryl sulphide was achieved by intramolecular cyclization of arylhydrazones (**2a, c**) with $AlCl_3$ in chlorobenzene to give 6-arylthio-1H-cinnolin-4-ones (**16, 17**).

Pyrazolo[4,3-c]cinnoline derivatives (**18, 19**) were obtained in good yield by the



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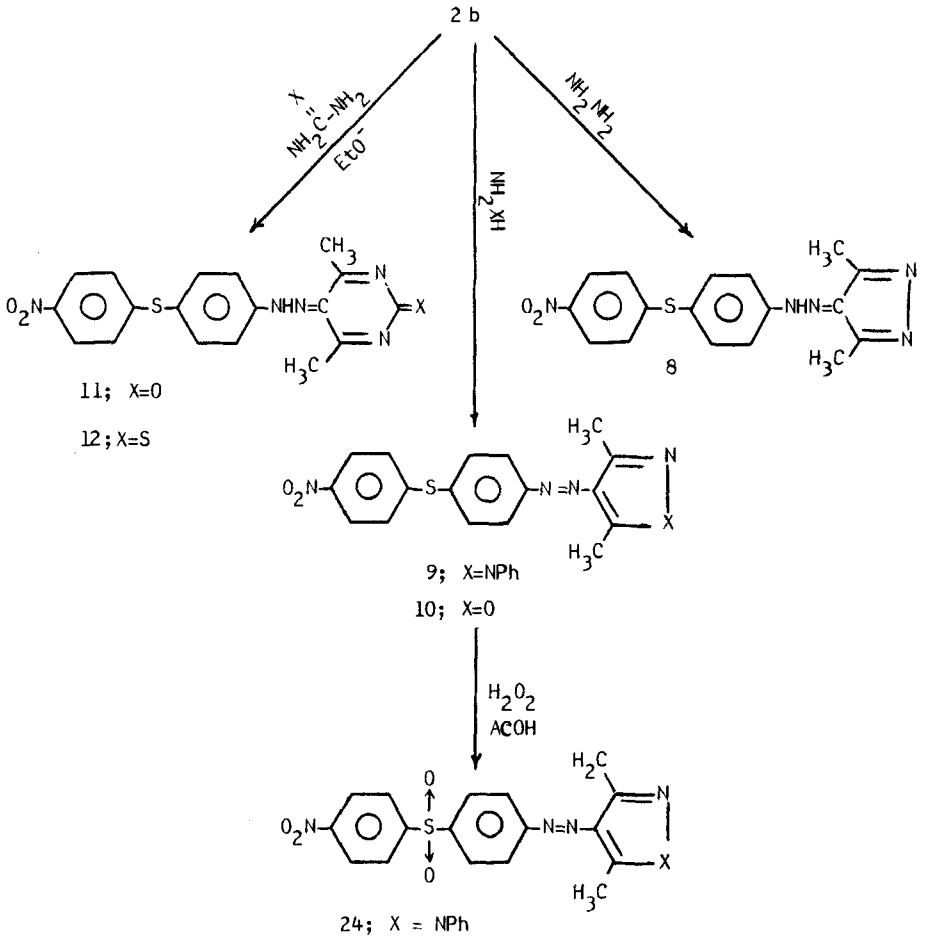

reaction of cinnoline derivative (**16**) with hydrazine hydrate in ethanol and with phenylhydrazine in acetic acid.

Oxidation of sulphides (**2c, 3, 4, 6, 9, 14–16, 18, 19**) by hydrogen peroxide in glacial acetic acid at room temperature for 2–6 days afforded the corresponding sulphones (**20–29**). The chemical structure of sulphones (**20–30**) was elucidated from their elemental analyses and IR spectra.

EXPERIMENTAL

Melting points were determined on Fisher-Johns melting point apparatus and were uncorrected. Elemental analyses were performed on a Perkin-Elmer 240 elemental analyser. IR spectra were recorded on a Pye-Unicam SP 3-100 spectrophotometer using KBr wafer technique. H^1 NMR spectra were recorded on a Varian EM-390 90 MHz NMR spectrometer in suitable deutrated solvent using TMS as internal standard.

Reaction of diazonium salt (**1**) *with active methylene compounds. Formation of ethyl acetoacetate-, acetylacetone-, diethyl malonate-4[(4'-nitrophenyl)thio]phenylhydrazones* (**2a–c**). General procedure: To a well stirred solution of ethyl acetoacetate, acetylacetone and/or diethyl malonate (0.1 mol) in

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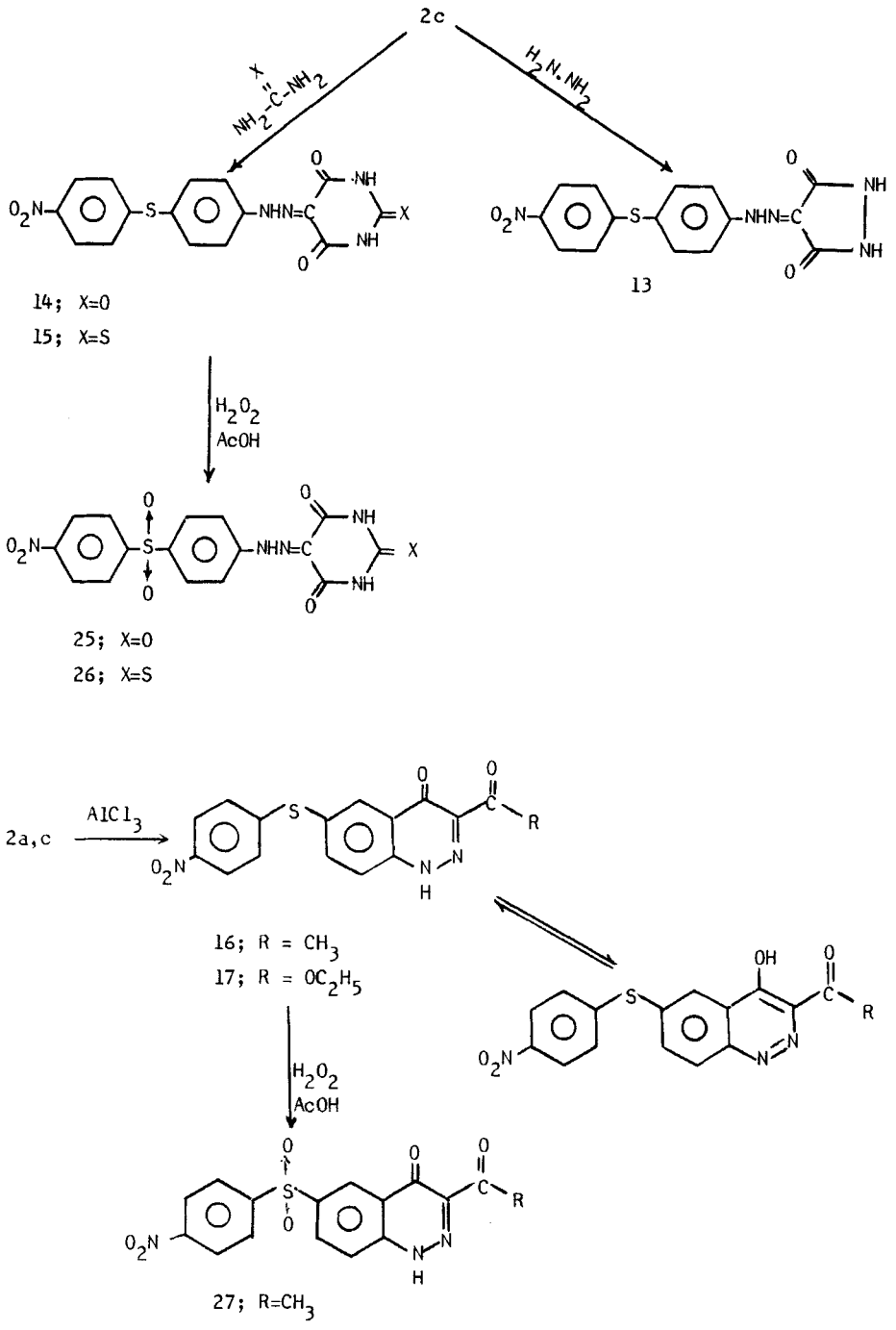

ethanol (200 ml) and water (20 ml) containing pot. acetate (10 gm), the diazonium salt (**1**) (prepared by the usual way) was added gradually with stirring during 30 min. at 0–5°C. The formed product was filtered, washed with water, dried and recrystallised from ethanol. Physical constants and spectral data of compounds **2a–c** were represented in Table I.

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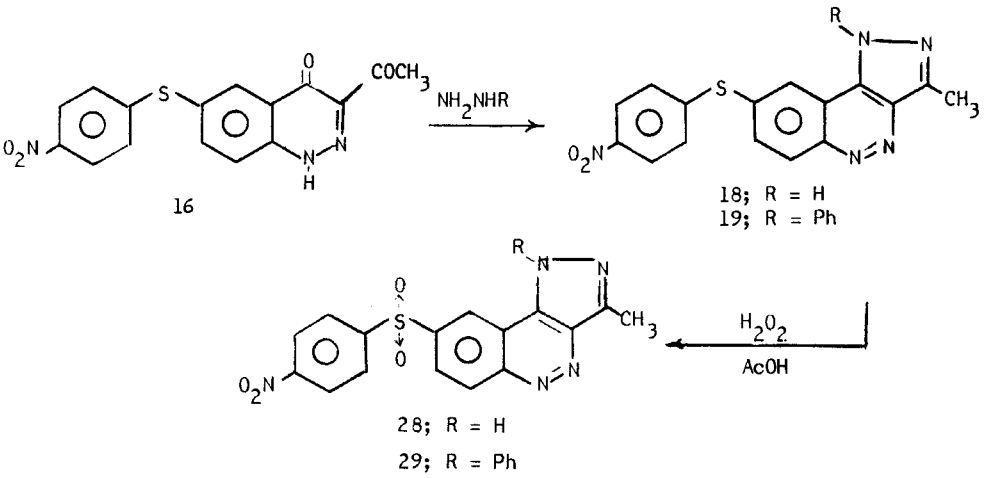

Reaction of hydrazones **2a–c** *with hydrazines. Formation of pyrazoline derivatives* **3, 4, 8, 9** *and* **13**: A mixture of hydrazone **2a–c** (0.005 mol) and hydrazine hydrate (0.02 mol) in ethanol (30 ml) or phenylhydrazine (0.02 mol) in acetic acid (20 ml) was refluxed for 3 hrs. The product obtained on concentration and cooling was collected and recrystallized from suitable solvent. Physical and spectral data are summarized in Table I.

Reaction of **2a, b** *with hydroxylamine. Formation of isoxazoline derivatives* **5** *and* **10**. A mixture of **2a, b** (0.005 mol) and hydroxylamine (0.02 mol) (prepared from NH_2OH—HCl and sodium acetate) in ethanol (30 ml) was refluxed for 5 hrs. Concentration and cooling afforded the desired product.

4-Nitro-4′[(3″-methyl-5″-oxo-isoxazole-4″-yl)hydrazono]diaryl sulphide (**5**) obtained in 76% yield as pale yellow crystals (ethanol), m.p. 136–8°C.

Anal. Calcd. for $C_{16}H_{12}N_4O_4S$: C, 53.93; H, 3.39; N, 15.72; S, 9.00.
Found: C, 54.12; H, 3.50; N, 15.55; S, 9.20.

IR: at 3100–2900 cm^{-1} (NH), 1720 cm^{-1} (C=O) and 1580 cm^{-1} (C=N).

4-Nitro-4′[(3″, 5″-dimethylisoxazol-4″-yl)diazo]diarylsulphide (**10**) obtained in 81% yield as deep yellow crystals (ethanol) m.p. 201–3°C.

Anal. Calcd. for $C_{17}H_{14}N_4O_3S$: C, 57.62; H, 3.98; N, 15.81; S, 9.05.
Found: C, 57.41; H, 3.77; N, 15.60; S, 9.20.

IR: at 1610 cm^{-1} (C=N) and disappearance the characteristic band of carbonyl group at 1660 cm^{-1}. H^1NMR in $CDCl_3$: at δ 8.1-7.1 (m, 8H, Ar-H) and at δ 2.7, 2.4 (2s, 6H, 2 CH_3).

Reaction of **2a–c** *with urea and thiourea. Formation of Pyrimidine derivatives* **6, 7, 11, 12, 14** *and* **15**: A mixture of **2a–c** (0.003 mol) and urea or thiourea (0.003 mol) in absolute ethanol (20 ml) containing sodium ethoxide solution (2 ml, prepared from 0.5 g sodium metal in 10 ml absolute ethanol) was refluxed for 7 hrs. The product formed on cooling and acidification with acetic acid was filtered off, dried and recrystallised from the proper solvent. Results are summarized in Table II.

Cyclization of **2a, c** *with* $AlCl_3$. *Formation of cinnoline derivatives* **16** *and* **17**. A mixture of **2a, c** (0.01 mol) and anhydrous $AlCl_3$ (0.02 mole) in chlorobenzene (30 ml) was refluxed for one hour. The complex formed was decomposed with conc. HCl (30 ml) and diluted with ice-water. The formed product was filtered, washed with water, dried and recrystallised from the proper solvent.

3-Acetyl-6(4′-nitrophenylthio)-4-oxo-1H-cinnoline (**16**) obtained in 78% yield as yellow crystals (acetic acid), m.p. 181–3°C.

Anal. Calcd. for $C_{16}H_{11}N_3O_4S$: C, 56.30; H, 3.25; N, 12.31; S, 9.39.
Found: C, 56.44; H, 3.50; N, 12.11; S, 9.45.

IR: at 3700–3240 cm^{-1} (NH br.) and at 1680 cm^{-1} (C=O). ^{1}H NMR in $CDCl_3$: at δ 14.1 (s, 1H, NH), at δ 8.1-7.1 (m, 7H, Ar-H) and at δ 2.6 (s, 3H, $COCH_3$).

3-Carbethoxy-6(4′-nitrophenylthio)-4-oxo-1H-cinnoline (**17**) obtained in 69% yield as yellow crystals (acetic acid) m.p. 178–80°C.

Anal. Calcd. for $C_{17}H_{13}N_3O_5S$: C, 54.98; H, 3.53; N, 11.32; S, 8.63.
Found: C, 55.15; H, 3.64; N, 10.98; S, 8.70.

IR at 3185–3000 cm^{-1} (NH) and at 1700 cm^{-1} (C=O).

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TABLE I

Physical and spectral data of hydrazones 2 and pyrazole derivatives (**3, 4, 8, 9, 13**)

Compd. No.	m.p. °C (solvent of cryst.)	Yield %	Molecular formula	Elemental analysis calcd./Found % C	H	N	S	Spectral data
2a	164–6 (EtOH)	72	$C_{18}H_{17}N_3O_5S$	55.81 55.60	4.42 4.25	10.85 11.10	8.28 8.12	IR: 3120–2670 cm^{-1} (NH), 1680 and 1660 cm^{-1} (C=O). NMR($CDCl_3$): δ 12.65 (s, 1H, NH), δ 8.10-7.05 (m, 8H, Ar-H), δ 4.45-4.20 (q, 2H, OCH_2-), δ 2.50 (s, 3H, $COCH_3$) and δ 1.45-1.30 (t, 3H, CH_3 ester).
2b	188–90 (Bz-pet.eth. 60–80)	80	$C_{17}H_{15}N_3O_4S$	57.13 56.95	4.23 4.45	11.76 11.85	8.97 9.10	IR: 3080–2800 cm^{-1} (NH) and 1670 cm^{-1} (C=O).
2c	116–8 (EtOH)	68	$C_{19}H_{19}N_3O_6S$	54.67 54.42	4.59 4.75	10.07 10.22	7.68 7.48	IR: 3080–2800 cm^{-1} (NH) and 1710, 1670 cm^{-1} (C=O ester).
3	257–9 (Acetic acid)	83	$C_{16}H_{13}N_5O_3S$	54.08 53.94	3.69 3.33	19.71 19.50	9.02 9.22	IR: 3400–3320 cm^{-1} (NH) and 1660 cm^{-1} (C=O). NMR (DMSO-d_6): δ 11.50 (s, 1H, NH), δ 8.10-7.20 (m, 8H, Ar-H) and δ 2.15 (s, 3H, CH_3).
4	195 (Acetic acid)	69	$C_{22}H_{17}N_5O_3S$	61.24 61.36	3.97 4.12	16.23 16.55	7.43 7.59	IR: 3100–2900 cm^{-1} (NH) and 1670 cm^{-1} (C=O). NMR ($CDCl_3$): δ 11.3 (s, 1H, NH), δ 8.1-7.1 (m, 13H, Ar-H) and δ 2.4 (s, 3H, CH_3).
8	167–169°C (EtOH)	84	$C_{17}H_{15}N_5O_2S$	57.78 57.57	4.28 4.50	19.82 19.02	9.07 9.00	IR: 3500–3300 cm^{-1} (NH) and 1610 cm^{-1} (C=N).
9	134 (EtOH)	63	$C_{23}H_{19}N_5O_2S$	64.32 64.65	4.46 4.50	16.31 16.35	7.46 7.57	NMR ($CDCl_3$): δ 8.1-7.2 (m, 13H, Ar-H) and δ 2.6 (s, 6H, $2CH_3$).
13	235–7 (EtOH)	80	$C_{15}H_{11}N_5O_4S$	50.42 50.47	3.10 3.30	19.60 19.54	8.97 9.12	IR: 3460–3100 cm^{-1} (br NH) and 1660 cm^{-1} (C=O cyclic).

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TABLE II

Physical and spectral data of pyrimidine derivatives (**6, 7, 11, 12, 14, 15**)

Compd. No.	m.p. °C (solvent of cryst.)	Yield %	Molecular formula	Elemental analysis calcd./Found % C	H	N	S	Spectral data
6	187–9 (Acetic acid)	50	$C_{17}H_{13}N_5O_4S$	53.26 53.12	3.42 3.40	18.27 18.18	8.36 8.50	IR: 3140–2900 cm^{-1} (NH) and 1670 cm^{-1} (C=O). NMR ($CDCl_3$): δ 14.05 (s, 1H, NH), δ 8.15-7.20 (m, 9H, Ar-H and NH-pyrimidine) and at δ 2.55 (s, 3H, CH_3).
7	228–30 decomp. (Acetic acid)	52	$C_{17}H_{13}N_5O_3S_2$	51.12 50.92	3.28 3.30	17.53 17.65	16.05 16.00	IR: 3100–2800 cm^{-1} (NH), 1655 cm^{-1} (C=O), 1200 cm^{-1} (C=S).
11	180–2 (Bz.-EtOH)	59	$C_{18}H_{15}N_5O_3S$	56.68 56.58	3.96 4.12	18.36 18.34	8.41 8.55	IR: 3090–2800 cm^{-1} (NH) and 1670 cm^{-1} (C=O).
12	184–6 (Acetic acid)	67	$C_{18}H_{15}N_5O_2S_2$	54.39 54.36	3.80 4.01	17.62 17.60	16.13 16.02	IR: 3550–3100 cm^{-1} (NH) and 1665 cm^{-1} (C=N). NMR ($CDCl_3$): δ 14.50 (s, 1H, NH), δ 8.10-7.05 (m, 8H, Ar-H) and δ 2.30 (s, 6H, 2 CH_3).
14	163 (EtOH)	59	$C_{16}H_{11}N_5O_5S$	48.87 49.10	2.88 3.02	18.17 18.12	8.32 8.50	IR: 3240–2800 cm^{-1} (NH) and 1700 cm^{-1} (C=O).
15	186–8 (Acetic acid)	62	$C_{16}H_{11}N_5O_4S_2$	47.87 47.92	2.76 2.83	17.45 17.56	15.97 16.10	IR: 3170–3000 cm^{-1} (NH), 1695 cm^{-1} (C=O) and 1210 cm^{-1} (C=S).

Synthesis of pyrazolo[4,3-c]cinnoline derivatives (**18** *and* **19**). A mixture of 16 (0.005 mol) and hydrazine hydrate in ethanol or phenylhydrazine (0.02 mol) in acetic acid was refluxed for 3 hrs. The precipitated product was collected and recrystallised from the proper solvent.

3-Methyl-8(4'-nitrophenylthio)-1H-pyrazolo[4,3-c]cinnoline (**18**) was obtained in 78% yield as deep yellow crystals (acetic acid), m.p. 258–60°C.

Anal. Calcd. for $C_{16}H_{11}N_5O_2S$: C, 56.97; H, 3.29; N, 20.76; S, 9.50.
Found: C, 59.10; H, 3.52; N, 20.65; S, 10.12

IR 3400–3300 cm^{-1} (NH) and at 1650 cm^{-1} (C=N).

3-Methyl-1-phenyl-8(4'-nitrophenylthio)-1H-pyrazolo[4,3-c]cinnoline (**19**) obtained in 69% yield as yellow orange crystals (acetic acid), m.p. 200–2°C.

Anal. Calcd. for $C_{22}H_{15}N_5O_2S$: C, 63.91; H, 3.66; N, 16.94; S, 7.75.
Found: C, 63.40; H, 3.95; N, 17.01; S, 7.50.

IR at 1650 cm^{-1} (C=N). ^1H NMR in $CDCl_3$ at δ 8.15-7.20 (m, 12H, Ar-H) and at δ 2.3 (s, 3H, CH_3).

Oxidation of sulphides (**2c, 3, 4, 6, 9, 14–16, 18** *and* **19**) *to their corresponding sulphones* (**20–29**): To the above sulphides (0.005 mol) in glacial acetic acid (30 ml) was added dropwise hydrogen peroxide (30%, 10 ml.). The reaction mixture was kept at room temperature for 2–6 days, whereby, crystalline products were deposited, collected and recrystallised from acetic acid. Results are summarized in Table III.

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TABLE III

Physical and spectral data of sulphones (**20–29**)

Compd. No.	m.p. °C	Yield %	Molecular formula	Elemental analysis calcd./Found % C	H	N	S	Spectral data
20	171–3	57	$C_{19}H_{19}N_3O_8S$	50.78 50.70	4.26 4.25	9.35 9.30	7.13 7.10	IR: 3140 cm^{-1} (NH), 1725, 1690 cm^{-1} (C=O) and 1160, 1360 cm^{-1} (SO_2). NMR ($CDCl_3$): δ 12.55 (s, 1H, NH), δ 8.35-7.30 (m, 8H, Ar-H), δ 4.45-4.20 (q, 2H, OCH_2 ester) and at δ 1.45-1.3 (t, 3H, CH_3 ester).
21	298–300	60	$C_{16}H_{13}N_5O_5S$	49.61 49.63	3.38 3.35	18.08 18.10	8.28 8.15	IR: 3360–2800 cm^{-1} (NH), 1680 cm^{-1} (C=O) and 1350, 1155 (SO_2).
22	227	37	$C_{22}H_{17}N_5O_5S$	57.01 57.12	3.70 3.50	15.11 15.00	6.92 7.10	IR: 3150–3050 cm^{-1} (NH), 1655 cm^{-1} (C=O) and 1340, 1150 cm^{-1} (SO_2). NMR (DMSO): δ 8.30-7.20 (m, 13 H, Ar-H) and 2.65 (s, 3H, CH_3).
23	318–20	39	$C_{17}H_{13}N_5O_6S$	49.16 49.15	3.15 3.20	16.86 16.70	7.72 7.80	IR: 3320–2600 cm^{-1} (NH), 1650 cm^{-1} (C=O) and 1340, 1150 cm^{-1} (SO_2). NMR (DMSO-d_6): δ 14.10 (s, 1H, NH), δ 8.20-7.50 (m, 9H, 8 Ar-H and NH Pyrimidine ring) and δ 2.75 (s, 3H, CH_3).

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24	190	45	$C_{23}H_{19}N_5O_4S$	59.86 60.00	4.15 4.20	15.18 15.25	6.95 7.15	IR: 1345 and 1155 cm^{-1} (SO_2). NMR ($CDCl_3$): δ 8.40-7.50 (m, 13H, Ar-H) and at δ 2.65, 2.55 (2s, 6H, 2 CH_3).
25	210	56	$C_{16}H_{11}N_5O_7S$	46.05 46.13	2.66 2.65	16.78 16.70	7.68 7.55	IR: 3220–2800 cm^{-1} (NH), 1700 cm^{-1} (C=O) and 1345, 1150 cm^{-1} (SO_2). NMR ($CDCl_3$): δ 13.3 (s, 1H, NH) and at δ 8.30-7.40 (m, 10 H, 8 Ar-H and 2 NH-pyrimidine ring).
26	220–2	52	$C_{16}H_{11}N_5O_6S_2$	44.34 44.49	2.56 2.66	16.16 16.20	14.79 14.80	IR: 3200–3030 cm^{-1} (NH), 1710 cm^{-1} (C=O), 1340, 1150 cm^{-1} (SO_2) and 1210 cm^{-1} (C=S).
27	248–50	43	$C_{16}H_{11}N_3O_6S$	51.48 51.52	2.97 3.10	11.26 11.35	8.59 8.50	IR: 3220–3000 cm^{-1} (NH), 1700 cm^{-1} (C=O) and 1350, 1150 cm^{-1} (SO_2).
28	297	42	$C_{16}H_{11}N_5O_4S$	52.03 52.17	3.00 3.12	18.96 19.10	8.68 8.70	IR: 3280–3000 cm^{-1} (NH), 1680 cm^{-1} (C=N) and 1350, 1150 cm^{-1} (SO_2).
29	326	39	$C_{22}H_{15}N_5O_4S$	59.32 59.43	3.39 3.59	15.72 15.55	7.20 7.20	IR: 1650 cm^{-1} (C=N) and 1350, 1150 cm^{-1} (SO_2). NMR (DMSO): δ 8.35-7.3 (m, 12 H, Ar-H) and at δ 2.35 (s, 3H, CH_3).

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