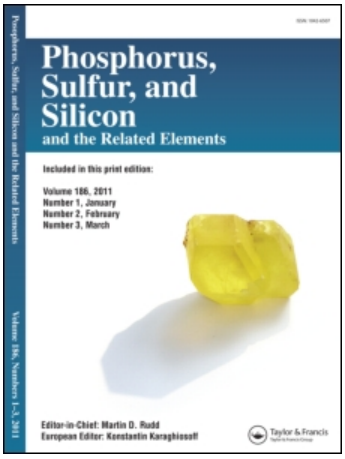

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REACTIONS WITH HYDRAZONOYL HALIDES XIII[1]: SYNTHESIS OF SOME NEW THIADIAZOLINE, ARYLAZOTHIAZOLE AND PYRAZOLE DERIVATIVES

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Hydrazonoyl halide **4** reacts with sodium thiophenolate and sodium benzenesulfinate to give the corresponding hydrazones **5** and **6**, respectively. Also, reacts with thiourea, potassium thiocyanate and β-ketosulfones, to give arylazothiazoles **7**, thiadiazolines **9**, and pyrazols **13**, respectively. Compounds **19** reacts with substituted thioanilide to afford thiadiazolines **24**. The structures of the newly synthesized compounds were assigned and confirmed on the basis of their elemental analyses, spectral data and alternate synthesis whenever possible.

Keywords: Hydrazonoyl halides; thiadiazoline; arylazothiozole; pyrazole; NMR spectra

INTRODUCTION

α-Ketohydrazonoyl halides have been largely employed as an exceedingly useful tool for the synthesis of heterocyclic compounds. The reactions take place through neucleophilic substitution or 1,3-dipolar cycloaddition reactions[2–5]. The results of the reaction of 2-bromo-1-naphthylglyoxal-2-arylhydrazones **4** with sodium thiophenolate, sodium benzenesulfinate, thiourea, potassium thiocyanate and β-ketosulfones are reported. Hydrazonoyl chlorides **19** reacted with thioanilide **17** and β-ketosulfone **3**a to give 2,3-dihydrothiadiazoles **21** and pyrazoles **24**, respectively. The reactions permitted synthesis of several new heterocyclic

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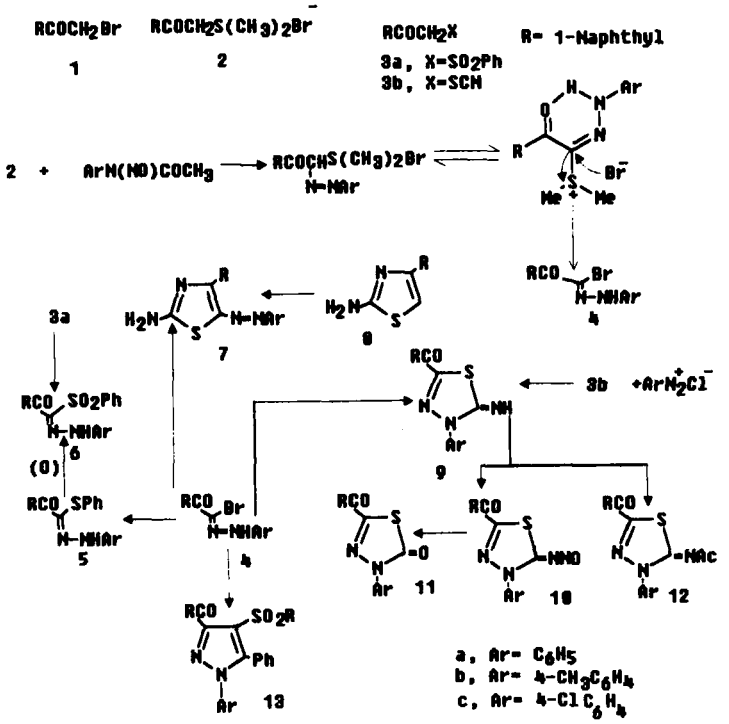
derivatives which have been reported to exhibit antiprolozoal[6], antiviral[7], bactericidal[8], and fungicidal[9] properties.

RESULTS AND DISCUSSION

Treatment of hydrazonoyl bromide **4** with sodium thiophenolate and sodium benzenesulfinate in ethanol gave a product with analytical and spectral data in accord with its formulation as 2-sulfoxynaphthylglyoxal-2-arylhydrazones (5) and 2-benzenesulfinylnaphthylglyoxal-2-arylhydrazones (6), respectively. Compound **5** was easily oxidised by hydrogen peroxide in acetic acid to give hydrazones **6**. Compound **6** was prepared by coupling arendiazonium chlorides with β-ketosulfone **3a** in ethanolic sodium acetate solution (*cf* Scheme 1).



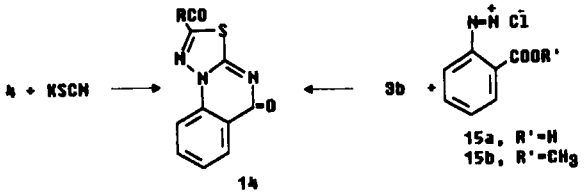
SCHEME 1

Treatment of **4** with excess thiourea in boiling ethanol gave a product which was identical with 2-amino-5-arylazo-1-aryl-3-(1)naphthylthiazole (7). The structure of thiazole **7** was confirmed by spectral data and preparation by another

route [coupling of arendiazonium chloride with 2-amino-4-(1′)naphthylthiazole (**8**) in ethanolic sodium acetate solution]. The ^{1}H NMR spectra of 7 showed signal (δ ppm) 5.8(s, br., 2H,NH_2) which disappeared upon shaking with D_2O and a new singlet appeared at 4.5. The UV spectra of **7** exhibited two intense maxima (log $\varepsilon > 4$) in the 470–420 and 280–260 nm regions. When **4** was treated with potassium thiocyanate in ethanol at room temperature gave only one product (TLC). The IR(cm^{-1}) spectra of the products showed no absorption band at 2150–2200 cm^{-1} due to the SCN group. The ^{1}H NMR spectrum of **9**b(δ ppm) showed signals at 2.3(s,3H,$CH_3C_6H_4$-p) and 7.2–8.4(m, 12H,ArH's and NH). ^{13}C NMR spectrum of **9**a revealed signals at δ 182.50, 158.91, 158.32, 158.0, 144.34, 133.32, 133.30, 128.35, 128.10, 125.15 and 124.91 ppm. The reaction was taken through the intermediate hydrazone which readly cyclized to give the final thiadiazoline **9**. The structure of **9** was further confirmed by independent synthesis. Treatment of **3**b with arenediazonium chlorides in ethanolic sodium acetate solution at 0°C yielded products identical in all respect (m.p., mixed m.p. and spectra) with samples prepared before (*cf.* scheme 1). Treatment of **9** with sodium nitrite in acetic acid at room temperature gave the nitroso derivatives **10** in good yield. The structures of the nitrosothiadiazolines were elucidated by analytical data, spectral studies and chemical reactions. The UV of these products revealed two common maxima in the region of 510–470 nm (log $\varepsilon < 2$) and 360–340 (log $\varepsilon < 4$). These are assigned to the n-π* and π-π* transition of the nitroso groups. IR of compounds **10** showed no NH band but revealed a band at 1530 cm^{-1} due to the (N-N=O) group[10]. Compound **10** decomposed to thiadiazolinones **11** by boiling in xylene solution. The IR (cm^{-1}) spectra of **11** showed two carbonyl absorption bands near 1660 and 1700. Acylation of **9** with acetic anhydride gave the corresponding N-Acetyl derivatives **12**. The structure of the acetylation products was established on the basis of elemental analyses and spectral data. The ^{1}H NMR spectrum of **12**a showed signals at (δ ppm) 2.3(s,3H,CH_3CON=) and 7.2–7.8(m,12H,ArH's). Treatment of **4**c with **3**a in ethanolic sodium ethoxide solution afford only one product which was identical to 4-benzenesulfonyl-1-p-chlorophenyl-5-naphthyl-3-naphthoylpyrazole **13**. The IR (cm^{-1}) showed a band at 1660 cm^{-1} (CO) and its ^{1}H NMR showed a signal at δ 7.1–7.3 (m, ArH's) (*cf* Scheme 1).

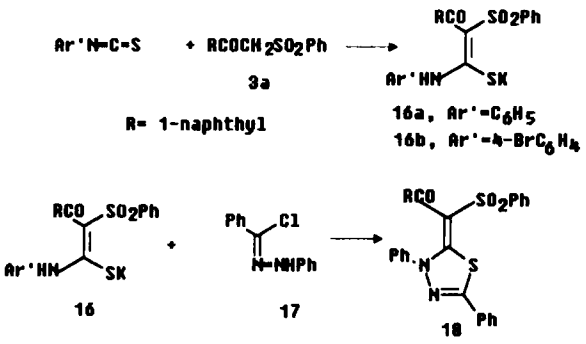
Treatment of hydrazonoyl bromide **4**d with KSCN in ethanol at room temperature gave thiadiazolo[3,2-a]quinazoline **14**. The structure of **14** was established on the basis of elemental analyses and spectral data. The IR (cm^{-1}) spectrum of **14** showed the absence of bands due to SCN, OH and NH groups and the presence of two carbonyl absorption bands at 1685 and 1650. To account for the formation of **14**, it is suggested that the reaction of **4**d with KSCN leads to the formation of hydrazone. The latter undergoes spontaneous cycloaddition[11]

to give the imino-1,3,4-thiadiazoline which completes the reaction by loss of water to give the final product **14**. Unequivocal support of the structure of **14** was achieved by its independent synthesis through another route. Thus, **3b** reacted with the appropriate diazotized aromatic amines **15a,b** in ethanolic sodium acetate solution at 0°C yielded products identical with **14** (*cf.* Scheme 2).



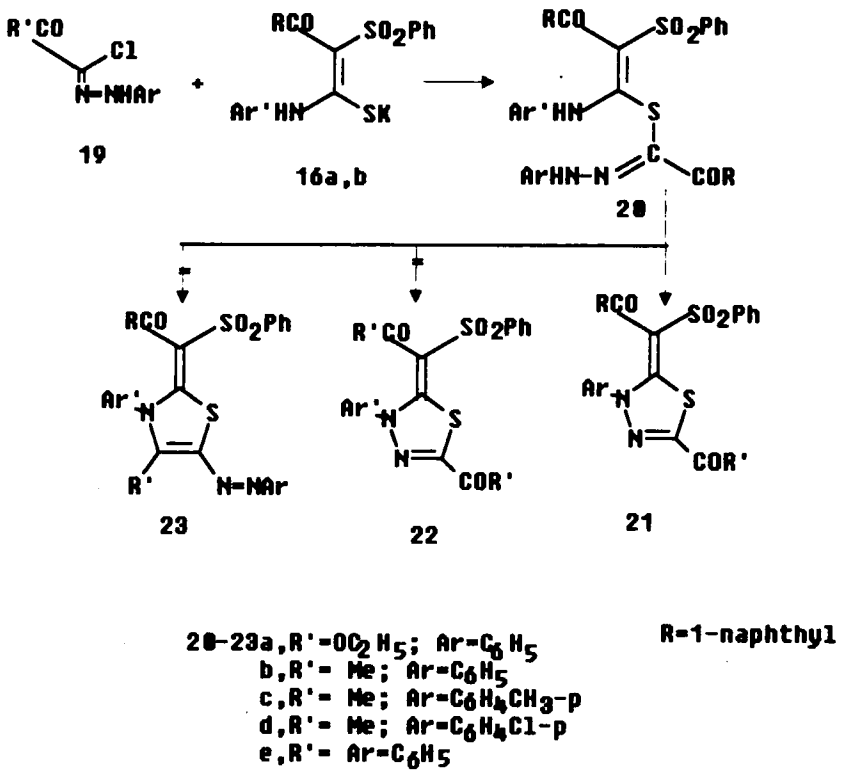
SCHEME 2

Treatment of the intermediate **16a** (prepared by the reaction of β-ketosulfone **3a** with aryl isothicyanate in dimethylformamide in the presence of potassium hydroxide (*cf.* Scheme 3) with hydrazonoyl chloride **17** gave a single product. The structure of the product was inferred from spectral data, the IR (cm^{-1}) spectrum showed a band near 1690 corresponding to the CO group. Its ^1H NMR spectrum contains a multiplet due to ArH's. ^{13}C NMR spectrum of **18** revealed signals at δ 168.40, 167.01, 162.29, 145.44, 143.51, 137.89, 135.89, 133.96, 132.46, 131.44, 131.33, 130.89, 130.69, 130.45, 130.35, 130.23, 129.55, 129.19, 129.01, 128.74, 127.84, 127.80, 125.61, 125.15, 102.45 and 105.50 ppm. The structure was confirmed by treatment of **16b** with **17** which produces a product identical in all respects (mp., mixed mp, IR, ^1H NMR) with the sample prepared before. This indicates that the reaction takes place through the formation of the S-alkylation intermediate which cyclized to give 2,3-dihydrothiadiazole **18** by loss of substituted anilines (*cf.* Scheme 3). Also the reaction of α-keto hydrazonoyl halides **19** with **16a** and **16b** was studied. Treatment of **19a** with **16a** gave only one product (TLC). IR(cm^{-1}) spectrum showed 1755 and 1720 (2 CO) groups. Its ^1H NMR spectrum revealed signals at δ 1.44(t,3H,CH_2CH_3), 4.47(q,2H, CH_2CH_3) and 7.11–7.85(m, 17H,ArH's) ppm. ^{13}C NMR spectrum of **21a** showed signals at δ 168.42, 167.75, 167.01, 162.29, 152.20, 145.42, 143.60, 137.80, 135.89, 134.10, 133.96, 132.46, 131.44, 131.33, 130.98, 130.69, 130.50, 130.30, 130.20, 129.19, 129.01, 128.74, 127.84, 127.80, 125.61, 125.15, 61.63 and 14.15 ppm. The structure of **21a** was further confirmed by the reaction of **19a** with **16b** which gives a product identical in all respects (mp., mixed mp., spectra) with **21a** (*cf.* scheme 4). The reaction of hydrazonoyl chlorides **19** with

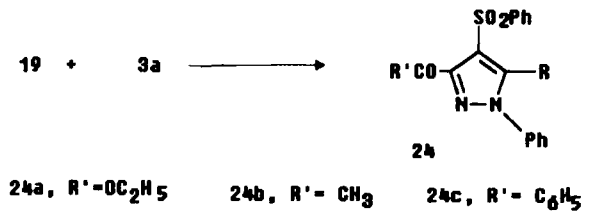


SCHEME 3

3a in the presence of sodium ethoxides afford substituted pyrazoles **24**. The structure of **24** was inferred from its spectral data (*cf* Scheme 5 and Experimental).



SCHEME 4



SCHEME 5

EXPERIMENTAL

All melting points were determined on a Gallenkamp and Mettler FP 61 melting point apparatus and are uncorrected. IR spectra in KBr disc on a Perkin-Elmer model 883 spectrophotometer. ^{1}H NMR spectra in ($CDCl_3$) and $(CD_3)_2SO$ on Joel-100 Mz FT spectrometer. UV spectra (EtOH) on Perkin-Elmer Lambda 4 spectrophotometer. Elemental analyses were performed by Microanalytical Center, Faculty of Science, Cairo University. 2-Bromoacetyl-(1)naphthaline[12], sulfonium bromide **2**[12] and 2-amino-4-(1′)naphthylthiazole (8)[13] were prepared according to previosly described methods.

2-Bromo-1′-napthylglyoxal-2-arylhydrazones (4a–d)

A mixture of sulfonium bromide **2** (0.1 mol) and the appropriate N-nitroso-substituted acetanilide[14] (0.12 mol) was stirred in ethanol (100 ml) for 1h. at room temperature. The solid was collected, washed with water and then recrystallized from ethanol to give hydrazonoyl bromides **4a–d** in 65–70% yield, respectively (*cf.* Table I).

Synthesis of 2-benzenesulfonyl-2(1′-naphthyl)ethan-2-one (3a), 2-thiocyanatoacetyl-1′-naphthaline (3b)

To a suspention of 2-bromoacetyl-1′-naphthaline (1) (12.5g, 0.05 mol) in ethanol (50 ml) a solution of the appropriate $PhSO_2Na$ or KSCN (0.06 mol) in water (10 ml) was added and the reaction mixture was refluxed for 30 min., cooled and diluted with water (50 ml). The solid was collected and recrystallized from ethanol to give **3a,b** in 68,73% yield, respectively. (*cf.* Table I).

TABLE I Characterization data of the newly synthesised products

Compd. no.	*M.p* °C	*Mol. Formula* *Mol. Wt.*	*% Analysis Calcd/Found* C	H	N	S
3a	145–146	$C_{18}H_{14}SO_3$	69.66	4.54	—	10.32
		(310.35)	69.50	4.40	—	10.20
3b	90–91	$C_{13}H_9NSO$	68.70	3.99	6.16	14.10
		(227.26)	68.60	4.10	6.20	14.20
4a	103–105	$C_{18}H_{13}N_2BrO$	61.20	3.70	7.93	
		(353.23)	61.10	3.60	7.90	
4b	153–154	$C_{19}H_{15}N_2BrO$	62.13	4.11	7.62	
		(367.25)	62.30	4.00	7.70	
4c	177–179	$C_{18}H_{12}BrClN_2O$	55.76	3.12	7.22	
		(387.67)	55.60	3.20	7.30	
4d	180–181	$C_{19}H_{13}N_2BrO_3$	57.44	3.29	7.05	
		(397.24)	57.30	3.10	6.90	
5c	162–164	$C_{24}H_{17}ClN_2SO$	69.14	4.11	6.71	7.68
		(416.91)	69.20	4.20	6.60	7.80
6a	160–161	$C_{24}H_{18}N_2SO_3$	69.55	4.37	6.75	7.73
		(414.46)	69.60	4.20	6.50	7.60
6b	173–175	$C_{25}H_{20}N_2SO_3$	70.07	4.70	6.53	7.47
		(428.49)	70.00	4.60	6.60	7.50
6c	159–161	$C_{24}H_{17}ClN_2SO_3$	64.21	3.81	6.24	7.13
		(448.91)	64.10	3.70	6.40	7.30
7a	256–258	$C_{19}H_{14}N_4S$	69.07	4.26	16.95	9.69
		(330.39)	68.80	4.40	16.90	9.70
7b	218–219	$C_{20}H_{16}N_4S$	69.74	4.68	12.26	9.30
		(344.43)	69.80	4.50	16.10	9.20
7c	257–258	$C_{18}H_{13}ClN_4S$	62.54	3.59	15.35	8.76
		(364.85)	62.40	3.40	15.40	8.70
9a	133–134	$C_{19}H_{13}N_3SO$	68.86	3.95	12.86	9.76
		(331.38)	68.60	4.10	12.70	9.60
9b	111–112	$C_{20}H_{15}N_3SO$	64.16	4.37	12.16	9.27
		(345.40)	64.00	4.40	12.10	9.40
9c	107–108	$C_{19}H_{12}ClN_3SO$	62.37	3.30	11.48	8.76
		(365.83)	62.40	3.10	11.30	8.50
10a	118–120	$C_{19}H_{12}N_4SO_2$	63.32	3.35	15.54	8.89
		(360.37)	63.20	3.50	15.40	8.90
10b	118–119	$'C_{20}H_{14}N_4SO_2$	64.16	3.76	14.96	8.55
		(374.40)	64.20	3.60	14.80	8.70
10c	128–129	$C_{19}H_{11}ClN_4SO_2$	5.79	2.80	14.19	8.11
		(394.83)	57.90	2.70	14.10	8.30
11c	126–128	$C_{19}H_{11}ClN_2SO_2$	62.21	3.02	7.63	8.73
		(399.82)	62.10	3.10	7.80	8.60
12a	191–192	$C_{21}H_{15}N_3SO_2$	67.54	4.04	11.25	8.58
		(373.41)	67.54	3.90	11.10	8.40
12c	170–171	$C_{21}H_{14}ClN_3SO_2$	61.83	3.45	10.30	7.86
		(387.44)	61.70	3.30	10.20	7.80
13c	179–180	$C_{36}H_{23}ClN_2SO_3$	72.17	3.89	4.67	5.34
		(599.09)	72.20	3.90	4.60	5.40
14	237–239	$C_{20}H_{11}N_3SO_2$	67.21	3.10	11.75	8.97
		(357.38)	67.10	3.20	11.90	8.80

TABLE I Continued

Compd. no.	M.p °C	Mol. Formula Mol. Wt.	% Analysis Calcd/Found C	H	N	S
18	234–236	$C_{28}H_{22}N_2S_2O_3$	67.45	4.44	5.61	12.85
		(498.59)	67.50	4.50	5.40	12.70
21a	184–185	$C_{29}H_{22}N_2S_2O_5$	64.19	4.08	5.16	11.81
		(542.60)	64.00	3.90	5.30	12.00
21b	225–227	$C_{28}H_{20}N_2S_2O_4$	65.61	3.93	5.46	12.50
		(512.57)	65.80	4.10	5.20	12.60
21c	232–234	$C_{29}H_{22}N_2S_2O_4$	66.14	4.21	5.31	12.17
		(526.60)	66.00	4.30	5.10	12.20
21d	222–224	$C_{28}H_{19}ClN_2S_2O_4$	61.47	3.50	5.12	11.71
		(547.02)	61.20	3.51	5.00	11.80
21e	215–216	$C_{32}H_{22}N_2S_2O_4$	68.31	3.94	4.97	11.39
		(562.63)	68.10	4.10	5.10	11.50
24a	145–146	$C_{28}H_{22}N_2S_2O_4$	69.69	4.59	5.80	
		(482.54)	69.60	3.80	5.90	
24b	140–142	$C_{27}H_{20}N_2SO_3$	71.66	4.45	6.19	
		(452.51)	71.50	4.50	5.30	
24c	144–146	$C_{32}H_{22}N_2SO_3$	74.69	4.30	5.44	
		(514.58)	74.50	4.40	5.60	

2-Sulfoxy-1′-naphthylglyoxal-2-arylhydrazones (5)

Equimolecular quantities of **4** and NaSPh (0.005 mol) in ethanol (25 ml) were stirred for 2h. and left overnight at room temperature. The product was collected, washed with water and crystallized from ethanol to give **5** in 78% yield (*cf.* Table I).

2-benzenesulfonyl-1′-naphthylglyoxal-2-arylhydrazones (6)

Method (A):- Equimolecular amount of the appropriate **4** and sodium benzenesulfinate (0.82g, 0.005 mol) in EtOH (20 ml) were refluxed for 2h. The reaction mixture was cooled, the solid collected, and then recrystallized from ethanol to give **6** in good yield (*cf.* Table I).

Method (B):- A solution of the appropriate diazotized primary aromatic amine (0.005 mol) was added dropwise to a cold solution of the appropriate **3**a (0.005 mol) and sodium acetate trihydrate (0.65g, 0.005 mol) while stirring. After the addition was complete (15 min), the reaction mixture was stirred for 3h. at 0°C. The solid was collected, washed with water and crystallized from ethanol to give **6**a–c (*cf.* Table I).

Method (C):- To a solution of **5** (2 mmol) in acetic acid (15 ml), a hydrogen peroxide solution (2 ml., 33%) was added, the mixture was left for two days at room temperature, then poured on water. Recrystallization of the product from ethanol gave the corresponding **6** (yield 73%). Mixed mp with samples prepared as above showed no depression.

Synthesis of 1-Aryl-2-imino-3-(1′)napthoyl-1,3,4-thiadiazolines 9, Thiadiazolo-[2,3-a]quinazoline 15

Method (A):- To a suspension of the appropriate hydrazonoyl bromide **4**a–d (0.001 mol) in ethanol (25 ml), a solution of KSCN (0.97g, 0.001 mol) in water (10 ml) was added while stirring at room temperature. The stirring was continued for 4h. and then diluted with water (50 ml). The solid was collected and recrystallized from ethanol or acetic acid to give the corresponding **9**a–c and **15**, (*cf.* Table I).

Method (B):- A solution of the appropriate diazotized primary aromatic amines (0.001 mol) was added dropwise to a cold solution of the appropriate **3**b (0.001 mol) in ethanol (50 ml) containing sodium acetate (1.3g, 0.001 mol) while stirring. After complete addition, the stirring was continued for 3h. at 0°C. The solid was collected, washed with water and recrystallized from ethanol or acetic acid. The product was found to be identical in all respects (mp., mixed mp. and spectra) with that obtained in Method A.

Nitrosation of 9: A solution of the appropriate **9**a–c (1g) in acetic acid (30 ml) was treated with a saturated sodium nitrite solution with stirring at 0–5°C. The reddish precipitate was collected and recrystallized from acetone to give **10**a–c (cf. Table I).

Themal Decomposition of 10c

A solution of the appropriate **9** (1g) in xylene was refluxed for 15 min. The solvent was evaporated under vacuum, then treated with pet. ether 40/60°C. The solid was collected and recrystallized from ethanol to yield **11**c in 80% yield (*cf.* Table I).

Acetylation of 9

The appropriate **9**a,c (0.5g) was stirred in acetic anhydride (10 ml) for 10 min at 70°C and left at room temperature for 2h. The solid was collected and recrys-

tallized from acetic acid. The N-acetyl derivatives **12**a,c was obtained in 77 and 85% yield (*cf.* Table I).

Synthesis of 2-Amino-5-arylazo-4-(1′)-napthylthiazoles 7

Method (A):- A solution of the appropriate hydrazonoyl bromide **4**a–c (0.005 mol) and thiourea (0.76g, 0.01 mol) in ethanol (20 ml) was refluxed for 3h., then, poured onto ice (100g) and two drops of ammonium hydroxide. The solid so formed was collected, washed with water and recrystallized from ethanol or dilute dimethylformamide to give **7**a–c, respectively. (*cf.* Table I).

Method (B):- A diazotized primary aromatic amine (0.001 mol) was added at 0–5°C to a cold solution of 2-amino-4-(1′)naphthylthiazole **8** (0.001 mol) in ethanol (50 ml) containing (1.3g) sodium acetate while stirring. The solid was collected, washed with water and recrystallized from ethanol or dilute dimethylformamide. The products were identical in all respects (mp., mixed mp. and spectra) with **7**a–c from method A.

Synthesis of 4-benzenesulfinyl-1-p-chlorophenyl-3-subistituted pyrazoles 13 and 24a–c

A solution of the appropriate hydrazonoyl halide **4**c and **19**a,b,d (0.005 mol) was added to a solution of ethanol (25 ml) containing β-ketosulfone **3**a (1.55g, 0.005 mol) and sodium metal (0.11g-atom, 0.005 mol) while stirring at room temperature for 2h., the reaction mixture was left overnight. The solid was collected, washed with water and recrystallized from ethanol to give **13**c and **24**a–c, respectively in 65% yield (*cf.* Table I).

Synthesis of 2,3-dihydro-1,3,4-thiadiazoles 18 and 21a–c

To a stirred suspention of potassium hydroxide(0.28g, 5 mmol) in dimethylformamide (20 ml), β-ketosulfone **3**a (1.5g, 5 mmol) and aryl isothiocyanate (5 mmol) were added. The reaction mixture was stirred for 3h at room temperature then the approperate hydrazonyl halides **17** and **19**a–e (5 mmol) were added and the reaction mixture stirred for 12h. The reaction mixture was diluted with water (10 ml) and the solid was collected and crystallized from acetic acid to give **18** and **21** a–e, respectively (*cf* Table I).

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