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Abdou O. Abdelhamid [a], Zeineb H. Ismail [b], Marwa S. El Gendy [b] & Moustafa M. Ghorab [c]

[a] Department of Chemistry, Faculty of Science, Cairo University, Giza, Egypt

[b] Department of Chemistry, Faculty of Science (Girls Branch), Al-Azhar University, Cairo, Egypt

[c] Department of Drug Radiation Research, National Center of Radiation Research and Technology, Naser City, Cairo, Egypt

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Reactions with Hydrazonoyl Halides 53:[1] Synthesis and Antimicrobial Activity of Triazolino[4,3-*a*]pyrimidines and 5-Arylazothiazoles

Abdou O. Abdelhamid
Department of Chemistry, Faculty of Science, Cairo University, Giza, Egypt

Zeineb H. Ismail
Marwa S. El Gendy
Department of Chemistry, Faculty of Science (Girls Branch), Al-Azhar University, Cairo, Egypt

Moustafa M. Ghorab
Department of Drug Radiation Research, National Center of Radiation Research and Technology, Naser City, Cairo, Egypt

6-(2-Naphtyl)-1-phenyl-4-3,5-disubstituted 4,3a-triazolino[4,3-a]pyrimidines, [2-(1-(2-naphthyl)-5-substitued (1-pyrazolin-3-yl)-4-phenyl(thiazol-5-yl)phenyldiazine and 1-(2-aza-2-{[4-phenyldiazenyl)-(1,3-thiazol-2-yl)]amino}vinyl)-naphthalene-2-ol were synthesized via *reactions of hydrazonoyl halides with 4-(2-naphthyl)-6-substituted 3,4-dihydropyrimidine-2-thione, Amino(3-(2-naphthyl)-5-substituted pyrazolin-2-ylmethane-1-thione, and 2-hydroxynaphthalenecarbaldehyde-thiosemicarbazone. All structures of the newly synthesized compounds were elucidated by elemental analysis, spectral data, and alternative synthesis methods whenever possible. Some of the new compounds were tested towards bacteria. In general, all tested compounds were capable of highly inhibiting the growth of gram positive of bacteria and gram negative.*

Keywords 2,3-Dihydro-1,3,4-thiadiazoles; arylazothiazoles; hydrazonoyl halides; pyrazolines; triazolino[4,3-*a*]pyrimidines

INTRODUCTION

1,2,4-Triazolo[4,3-*a*]pyrimidines have been found to exhibit antiviral, antifungal, antimicrobial, herbicidal, plant regulator, antitumor,

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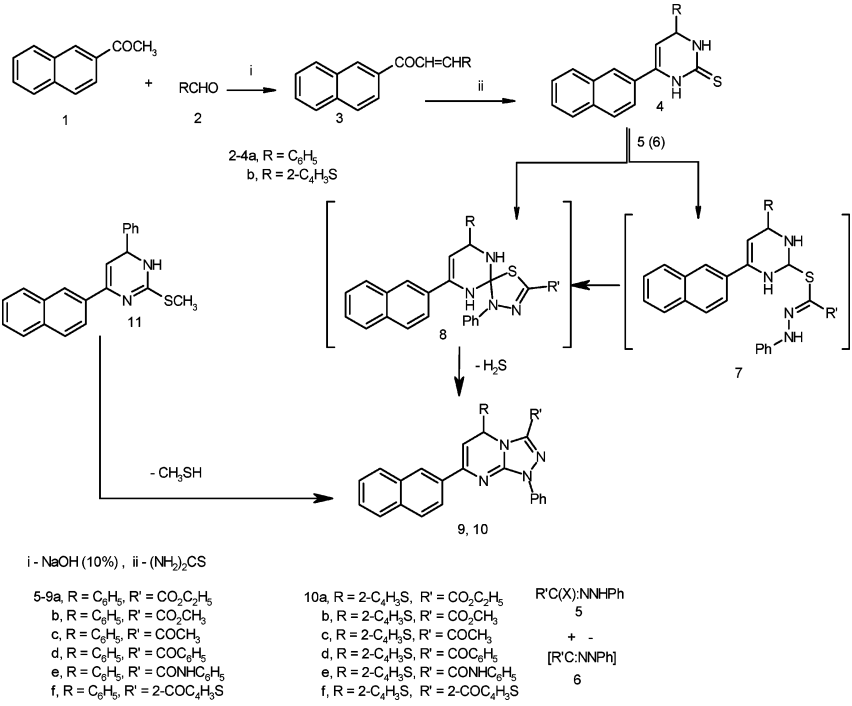
Address correspondence to Abdou O. Abdelhamid, Department of Chemistry, Faculty of Science, Cairo University, Giza 12316, Egypt. E-mail: abdou_abdelhamid@yahoo.com

antihypertensive, cardiovascular, and anxiolytic activities.[2] Also, many interesting papers, which have appeared lately, report on functionalization of thioamides and their use in organic synthesis, including regio- and stereoselective heterocyclization reactions. In particular, this concerns the thioamides, which have another reactive center in the molecule and therefore may serve as convenient building blocks.[3] We report herein the reactivity of hydrazonoyl halides towards 4-(2-naphthyl)-6-substituted 3,4-dihydropyrimidine-2-thione, Amino(3-(2-naphthyl)-5-substituted pyrazolin-2-ylmethane-1-thione and 2-hydroxynaphthalenecarbaldehydethiosemicarbazone.

RESULTS AND DISCUSSION

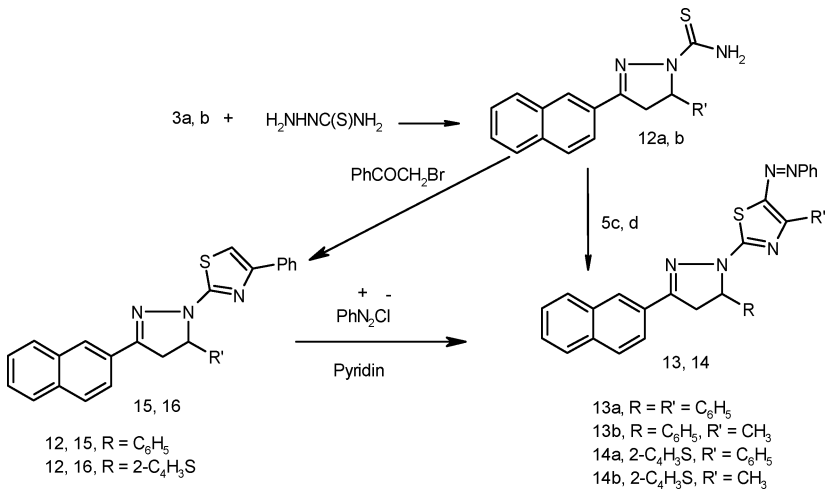
1-(2-Naphthyl)-3-phenylprop-2-en-1-one (**3a**) was reacted with thiourea in ethanolic potassium hydroxide gave 4-(2-naphthyl)-6-phenyl-3,4-dihydropyrimidine-2-thione (**4a**). Structure **4a** was elucidated by elemental analysis, spectra data, and chemical transformation. ^{1}H NMR spectrum of **4a** showed signals at $\delta = 4.59$ (s, 1H, pyrimidine), 7.06–8.88 (m, 14H, Aromatic protons and 2NH). Its MS spectrum showed peaks at m/z = 316 [M^+]. Thus, *C*-ethoxyycarbonyl-N-phenylhydrazonoyl chloride was reacted with **4a** in chloroform in presence of triethylamine afforded ethyl 6-(2-naphtyl)-1,4-diphenyl-4,3a-triazolino[4,3-*a*]pyrimidine-3-carboxylate (**9a**). Structure **9a** was confirmed by elemental analysis, spectral data and alternative synthesis (Scheme 1). ^{1}H NMR spectrum of **9a** showed signals at $\delta = 1.44$ (t, 3H, $J = 7$Hz, CH_3CH_2), 4.46 (q, 2H, $J = 7$H, CH_2CH_3), 4.95 (s, 1H, pyrimidine H-4), and 7.00–8.16 (m, 18H, aromatic protons). Thus, 2-methylthio-6-(2-naphthyl)-4-phenyl-3,4-dihydropyrimidine (**11a**), which prepared via methylation of **4a** with iodomethane in presence of sodium methoxide, in boiling sodium ethoxide under reflux gave identical product (mp., mixed mp., and spectra) with **9a**. Analogously, **4a** and **4b** were reacted with the appropriate hydrazonoyl halides **5a–f** in boiling chloroform under reflux afforded triazolino[4,3-*a*]pyrimidines **9b–f** and **10a–f**, respectively.

In the light of the foregoing results, the mechanism outlined in Scheme 1 seems to be the most plausible pathway for the formation of **9** from the reaction of the hydrazonoyl halides **5** with the dihydropyrimidine-2-thione **4**. The reaction involves initial formation of thiohydrazonates **7**, which undergoes intermolecular cyclization as soon as it is formed to give the spiro intermediate **8**. Ring chain tautomerism of spiro intermediate leads to the end products **9** via elimination of hydrogen sulfide (Scheme 1).



SCHEME 1

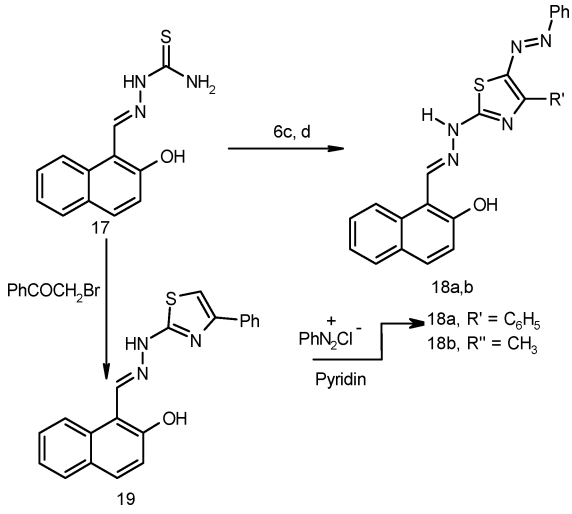
Treatment of thiosemicarbazide with 1-(2-naphthyl)-3-phenylprop-2-en-1-one (**3a**) to give amino(3-(2-naphthyl)-5-phenylpyrazolin-2-ylmethane-1-thione (**12a**). Structure **12a** was elucidated by elemental analysis, spectral data and chemical transformation (Scheme 2). ^{1}HNMR spectrum of **12a** showed signals at $\delta = 3.25$ (dd, 1H, J = 18.1, 5.8 Hz, $CH_{2(pyraz)}$, 3.82 ((dd, 1H, J = 18.1, 12.2 Hz, $CH_{2(pyraz)}$, 5.54 ((dd, 1H, J = 12.2, 5.8 Hz, $CH_{2(pyraz)}$, 6.82–8.10 (m, 14H, aromatic protons and NH_2). **12a** was reacted with *C*-benzoyl-*N*-phenylhydrazonoyl bromide **5d**, in boiling ethanolic triethylamine under reflux to afford [2-(1-(2-naphthyl)-4-phenyl-(1-pyrazolin-3-yl)-4-phenyl(thiazol-5-yl)phenyldiazine (**13a**). Structure **13a** was confirmed by elemental analysis, spectral data and alternative synthesis. Thus, treatment of benzenediazonium chloride with 2-(1-(2-naphthyl)-4-phenyl-(1-pyrazolin-3-yl))-4-phenylthiazole (**15**), which prepared via reaction of ω-bromoacetophenone with **12a** in ethanol, in pyridine gave product identical in all respects (mp. mixed mp. and spectra) with **13a**.



SCHEME 2

Similarly, treatment of the appropriate hydrazonoyl halides **5c**,**d** with the appropriate **12a**,**b** afforded phenylazothiazoles **13b**, **14a** and **14b**.

However, 2-hydroxynaphthalenecarbaldehydethiosemicarbazone (**17**) was reacted with the appropriate hydrazonoyl halides to give 5-aryazothiazole derivatives (Scheme 3). Compound **17** was



SCHEME 3

reacted with hydrazonoyl bromide **6d** in boiling ethanol containing triethylamine under reflux to give 1-(2-aza-2-{[4-phenyldiazenyl)-(1,3-thiazol-2-yl)]amino}vinyl)naphthalene-2-ol (**18**). Structure **18** was elucidated by elemental analysis, spectral data and alternative synthesis. Thus, treatment of benzenediazonium chloride with 3-(2-aza-2-[(4-phenylthiazol-2-yl)amino]vinyl)-naphthalin-2-ol (**19**) in pyridine afforded product identical in all respects (mp., mixed mp., and spectra) with **18a**.

Biological Activity

The tested microorganisms were gram +ve bacteria [*Staphylococcus aureus(ATCC25923)* and *Streptococcus pyrogenes (ATCC19615)*] and gram –ve bacteria (*Pseudomonas syrinage PV phasealicola*). In addition, some fungal pathogens (*Aspergillus niger* and *Fusarium oxysporum*) were also tested. Sensitivity of the selected microorganisms to some synthesized compounds was determined in vitro at two concentrations (100, 400 (mg/mL) in $CHCl_3$. The tests were carried out using the filter paper and hole plate method.[4]

Studies on the biological activity of compounds **3a**, **3b**, **9a**, **10b**, **10d**, and **11** led to the fact that these compounds have moderate biological activity against the tested bacteria, and only weak activity against fungi. Also, it can be observed (Table I) that compounds **12a**, **14a**, **16**, and **18a**, have only a weak effect on bacteria. Compounds **9e**, **12a**, **14a**, and **17** showed weak antifungal activity but compounds **15**, **16**, **18a**, and **19** showed moderate antifungal activity.

EXPERIMENTAL

All melting points were determined on an electrothermal apparatus and are uncorrected. IR spectra were recorded (KBr discs) on a Shimadzu FT-IR 8201 PC spectrophotometer. ^{1}H NMR spectra were recorded in $CDCl_3$and $(CD_3)_2SO$ solutions on a Varian Gemini 300 MHz spectrometer and chemical shifts are expressed in δ units using TMS as an internal reference. Mass spectra were recorded on a GC-MS QP 1000 EX Shimadzu. Elemental analyses were carried out at the Microanalytical Canter of the Cairo University. Hydrazonoyl halides,[5–8] **3a**,[9] and **3b**[10] were prepared as previously reported.

Synthesis of 4-(2-Naphthyl)-6-substituted 3,4-Dihydropyrimidine-2-thione 4a,b

A mixture of the appropriate chalcones **3a**,**b** (10 mmol), thiourea (0.76 g, 10 mmol) and potassium hydroxide (10%, 5 mL) in ethanol

TABLE I Response of Various Microorganisms to Some Synthesized Compounds in vitro Culture

Comp. no.	S.a.	S.p.	P.s.	A.n.	F.o.
3a	M	M	W	W	W
3b	M	M	W	W	W
9a	—	M	—	W	W
9e	—	W	—	W	—
10b	W	W	M	—	W
10d	W	W	M	W	W
11	—	W	M	W	W
12a	W	W	—	—	M
13a	W	W	M	—	W
13b	W	W	M	—	W
14a	—	W	W	—	W
15	—	W	M	—	M
16	W	W	W	—	M
17	W	W	W	—	W
18a	W	W	—	W	M
19	W	W	—	W	M
Control	S	S	S	S	S

Diameter of the zone inhibition: W: low activity (3–5 mm) (+); M: moderate (6–15 mm) (++); S: strong activity (>15 mm)(+ + +). The antibiotic which used as control was (Chlorumphinecol).

(20 mL) was refluxed 6 h. The resulting solid was collected and recrystallized from ethanol to give pyrazolines **4a** and**4b**, respectively (Tables II and III).

6-(2-Naphtyl)-1-phenyl-4-3,5-disubstituted 4,3a-triazolino [4,3-α]pyrimidines 9a–f and 10a–c

Method A. A mixture of the appropriate hydrazonoyl halides **5a–f** (5 mmol), the appropriate pyrimidine-2-thione **4a**,**b** (5 mmol) and triethylamine (0.5 g, 0.7 m mL, 5 mmol) in chloroform was boiled under reflux 10 h. Chloroform was removed under reduced pressure then triturated with petroleum ether 40–60°C. The resulting solid was collected and recrystallized from ethanol to give **9a–f** and **10a–c**, respectively (Tables II and III).

Method B. A mixture of the appropriate hydrazonoyl halides **5a–f** (5 mmol), the appropriate **11a,b** (5 mmol) and triethylamine (0.5 g, 0.7 m mL, 5 mmol) in ethanol was boiled under reflux 3 h. The solid was collected and recrystallized from ethanol gave identical product with corresponding from Method A.

TABLE II Spectral Data of Some Newly Synthesized Compounds

Comp. no	Spectral data
4a	^{1}H NMR: 4.59 (s, 1H, pyrimidine), 7.06–8.88 (m, 15H, Aromatic protons and 2NH).
	IR: 3442(NH) and 1544 (C═S).
	MS: 316 (72.7%), 313 (100%), 256(28.0%), 239(36.4%), 189(24.2%), 127 (22.0%), 77 (28.8%).
9a	^{1}H NMR: 1.44 (t, 3H, J = 7Hz, CH_3CH_2), 4.46 (q, 2H, J = 7H, CH_2CH_3), 4.59 (s, 1H, pyrimidine H-4), and 7.00–8.16 (m, 18H, aromatic protons).
	IR: 3050 (CH), 1736(C═O ester), and 1576 (C═C).
9b	^{1}H NMR: 3.79 (s, 3H, O$\underline{CH_3}$); 4.86 (s, 1H, pyrimidine H-4) and 7.01–7.58 (m, 18H, ArH's).
	IR: 1704 (C═O ester) and 1544 (C═N).
10a	^{1}H NMR: 1.38 (t, 3H, $CH_2\underline{CH_3}$), 5.34 (q, 2H, $\underline{CH_2}$, CH_3), 4.61 (s, 1H, pyrimidine H-4), and 7.05–8.57 (m, 16H, ArH's).
10c	^{1}H NMR: 2.55 (s, 3H, $\underline{CH_3}$), 4.82(s, 1H, pyrimidine H-4), and 7.10–8.51 (m, 16 H, ArH's).
11a	^{1}H NMR: 2.52 (s, 3H, S$\underline{CH_3}$), 4.86 (s, 1H, pyrimidine H–4), 7.24–8.23 (m, 13H, ArH's), and 8.65 (s, 1H, $\underline{NH}$).
12a	^{1}H NMR: 3.25 (dd, 1H, J = 18.1, 5.8 Hz, $CH_{2(pyraz)}$, 3.82 ((dd, 1H, J = 18.1, 12.2 Hz, $CH_{2(pyraz)}$), 5.54 ((dd, 1H, J = 12.2, 5.8 Hz, $CH_{2(pyraz)}$), 6.82–8.10 (m, 14H, aromatic protons and NH_2).
	IR: 3272,3220 (NH_2) and 1582 (C═S).
12b	IR: 3420,3252 (NH_2) and 1584 (C═S).
13b	^{1}H NMR: 2.44 (s, 3H, $\underline{CH_3}$), 3.25 (dd, 1H, J = 18.1, 5.8 Hz, $CH_{(pyraz)}$), 3.82 ((dd, 1H, J = 18.1, 12.2 Hz, $CH_{2(pyraz)}$), 5.54 ((dd, 1H, J = 12.2, 5.8 Hz, $CH_{2(pyraz)}$), 6.82–8.10 (m, 17H, aromatic protons).
	IR: 3028, 2930 (CH), and 1534 (C═C).
	MS: 473(M+, 39.3%), 272 (32.5%), 247(71.2%), 189(87.1%), 123(50.9%), 123(50.3%), 93(50.3%), 92 (76.7%), 77 (100.0%).
14a	IR: 30420 (CH) and 1590 (C═C).
	MS: 541 (54.7%), 153 (41.9%), 77(100.0%), 51(45.3%).
15	IR: 2922(CH) and 1552(C═C).
	MS: 431 (100%), 430 (95.2%), 174 (59.3%), 152 (81.5%), 126 (47.1%), 103 (47.2%), 77(52.8%).
17	IR: 3444(OH), (NH), 3246, 3164(NH_2), and 1608 (C═S).
18b	^{1}H NMR: 2.55 (s, 3H, CH_3), 7.20–7.98(m, 11H, ArH's), 8.50–8.54 (d, 1H, CH), 9.60(s, 1H, NH), and 10.80 (s, 1H, OH).
	IR: 3422(OH), 321(NH) and 1598(C═C).
19	MS: 345 (59.2%), 328 (100.0%), 176(82.2%), 134 (82.1%), 115(36.1%).

2-Methylthio-6-(2-naphthyl)-4-substituted 3,4-Dihydropyrimidine 11a and 11b

A solution of the appropriate **11a**,**b** (5 mmol), sodium methoxide (0.7 g, 5 mmol) in ethanol (20 mL) was stirred at room temperature, and then iodeomethane (072 g, 5 mmol) was added. The reaction mixture was stirred for 2 h at room temperature and the solid which formed

TABLE III Characterization Data of the Newly Synthesized Compounds

Comp. no.	M.p.°C (Solvent)	Color yield %	Mol. Formula (Mol. Wt.)	Calcd. / Found % C	H	N	S
4a	128–30	Yellow	$C_{20}H_{16}N_2S$	75.91	5.09	8.85	10.13
	(EtOH)	60	(316.42)	75.70	4.90	8.64	10.20
4b	178–80	Yellow	$C_{18}H_{14}N_2S_2$	67.04	4.37	8.68	19.88
	(EtOH)	60	(322.44)	6715	4.20	8.86	20.00
9a	142–44	Yellow	$C_{30}H_{24}N_4O_2$	76.25	5.11	11.85	—
	(EtOH)	65	(472.52)	76.37	5.00	11.95	
9b	249–50	White	$C_{29}H_{22}N_4O_2$	75.96	4.83	12.21	—
	AcOH)	60	(458.50)	76.10	4.95	12.40	
9c	180–82	Black	$C_{29}H_{22}N_4O$	78.71	5.01	12.66	—
	(EtOH)	50	(442.51)	78.50	4.95	12.54	
9d	125–27	Orange	$C_{34}H_{24}N_4O$	80.93	4.79	11.10	—
	(EtOH)	70	(504.58)	80.75	4.97	11.25	
9e	215–17	Yellow	$C_{34}H_{25}N_5O$	78.59	4.84	13.47	—
	(EtOH)	70	(519.59)	78.65	4.95	13.62	
9f	208–10	Red	$C_{32}H_{22}N_4OS$	62.78	4.34	10.97	6.70
	(EtOH)	60	(510.60)	62.87	4.43	1079	6.62
10a	142–45	Orange	$C_{28}H_{22}N_4O_2S$	70.27	4.63	11.70	6.69
	(EtOH)	60	(478.55)	70.42	4.58	11.85	7.12
10b	136–38	Yellow	$C_{27}H_{20}N_4O_2S$	69.98	4.33	12.06	6.90
	(EtOH)	50	(464.52)	69.78	4.12	12.25	7.10
10c	138–40	Brown	$C_{27}H_{20}N_4OS$	72.30	4.49	12.42	7.14
	(EtOH)	50	(448.53)	72.50	5.12	12.57	7.24
10d	204–206	Orange	$C_{32}H_{22}N_4OS$	75.27	4.34	10.97	6.27
	(EtOH)	50	(510.60)	75.45	4.43	11.22	6.35
10e	158–60	Yellow	$C_{32}H_{23}N_5OS$	73.12	4.41	13.32	6.09
	(EtOH)	50	(525.61)	73.25	4.24	13.52	6.25
10f	215–17	Red	$C_{30}H_{20}N_4OS_2$	69.74	3.30	10.84	12.42
	(EtOH)	50	(516.65)	69.65	3.25	10.96	12.53
11a	114–16	White	$C_{21}H_{18}N_2S$	76.33	5.49	8.48	9.70
	(EtOH)	64	330.46	76.52	5.74	8.65	9.59
11b	126–28	White	$C_{19}H_{16}N_2S_2$	76.82	4.79	8.32	19.05
	(EtOH)	50	336.47	76.50	5.64	8.20	19.00
12a	170–72	Yellow	$C_{20}H_{17}N_3S$	72.47	5.16	12.98	9.67
	(EtOH)	60	(331.43)	72.65	5.26	13.12	9.52
12b	100–102	Yellow	$C_{18}H_{15}N_3S_2$	64.06	4.48	12.45	19.00
	(EtOH)	60	(337.45)	64.26	4.47	12.35	18.85
13a	108–10	Yellow	$C_{34}H_{25}N_5S$	76.23	4.70	13.07	5.98
	(EtOH)	70	(535.66)	76.32	4.62	13.24	5.85
13b	150–53	Yellow	$C_{29}H_{23}N_5S$	73.54	4.89	14.78	6.76
	(EtOH)	50	(373.59)	73.65	4.98	14.87	6.85
14a	167–70	Red	$C_{32}H_{23}N_5S_2$	80.13	4.83	14.60	13.36
	(EtOH)	70	(541.61)	80.22	4.68	14.80	13.57
14b	136–39	Brown	$C_{27}H_{21}N_5S_2$	67.61	4.41	14.60	13.36
	(EtOH)	60	(479.61)	67.47	4.62	14.49	13.58
15	240–42	Yellow	$C_{28}H_{21}N_3S$	77.92	4.90	9.73	7.42
	(AcOH)	80	(431.55)	77.85	5.12	9.56	7.49

(Continued on next page)

TABLE III Characterization Data of the Newly Synthesized Compounds (*Continued*)

Comp. no.	M.p.°C (Solvent)	Color yield %	Mol. Formula (Mol. Wt.)	Calcd. / Found %			
				C	H	N	S
16	220–22	Yellow	$C_{26}H_{19}N_3S_2$	71.36	4.37	9.60	14.65
	(AcOH)	80	(437.57)	71.63	4.57	9.42	14.56
17	271–73	Yellow	$C_{12}H_{11}N_3OS$	58.76	4.51	17.13	13.07
	(AcOH)	80	(245.28)	58.67	4.35	17.15	13.25
18a	245–47	Brown	$C_{26}H_{19}N_5OS$	69.47	4.26	15.58	7.13
	(EtOH)	60	(449.54)	69.54	4.58	15.28	7.15
18b	216–18	Red	$C_{21}H_{17}N_5OS$	65.10	4.42	18.07	8.28
	(AcOH)	60	(387.47)	64.85	4.36	18.24	8.45
19	220–22	Yellow	$C_{20}H_{15}N_3OS$	69.54	4.37	12.16	9.28
	(AcOH)	70	(345.40)	69.32	4.58	12.34	9.35

was collected and recrystallized from ethanol to afford **11a** and **11b**, respectively (Tables II and III).

Amino(3-(2-naphthyl)-5-substituted Pyrazolin-2-ylmethane-1-thione (12a)

A mixture of the appropriate chalcones **3a**,**b** (10 mmol), thiosemicarbazide (0.91 g, 10 mmol) in acetic acid (20 mL) was boiled under reflux for 2 h. The resulting solid was collected and recrystallized from ethanol to give pyrazolines **12a** and**12b**, respectively (Tables II and III).

[2-(1-(2-Naphthyl)-5-substitued (1-pyrazolin-3-yl)-4-phenyl(thiazol-5-yl)phenyldiazine 13a,b and 14a,b

A mixture of the appropriate **12a**,**b** (5 mmol), the appropriate hydrazonoyl halides (5 mmol) and triethylamine (0.75 mL, 5 mmol) in ethanol (20 mL) was heated under reflux 4 h. The resulting solid was collected and recrystallized from ethanol to give **13a**,**b** and**14a**,**b**, respectively (Tables II and III).

2-(1-(2-Naphthyl)-5-substitued (1-pyrazolin-3-yl))-4-phenylthiazole 15 and 16

A mixture of the appropriate **12a**,**b** (5 mmol) and phenacylbromide (1 g, 5 mmol) in ethanol (20 mL) was heated under reflux 2 h then poured onto ice cold water (50 mL) containing ammonium hydroxide (2 drops). The resulting solid was collected and recrystallized from ethanol to give **15** and**16**, respectively (Tables II and III).

2-Hydroxynaphthalenecarbaldehydethiosemicarbazone (17)

A mixture of 2-hydroxynaphtalenecarbadehyde (1.8 g, 10 mmol), thiosemicarbazide (0.91 g, 10 mmol) ethanol (20 mL) and acetic acid

(3 drops) was stirred at room temperature 30 min. The resulting solid was collected and recrystallized from ethanol to give thiosemicarbazone **17** (Tables II and III).

1-(2-Aza-2-{[4-phenyldiazenyl)-(1,3-thiazol-2-yl)]amino}vinyl) naphthalene-2-ol 18a,b

A mixture of **17** (1.22 g, 5 mmol), the appropriate hydrazonoyl halides **5c**,**d** (5 mmol) and triethylamine (0.75 mL, 5 mmol) in ethanol (20 mL) was heated under reflux 4 h. The resulting solid was collected and recrystallized from ethanol to give **18a**,**b**, respectively (Tables II and III).

3-(2-Aza-2-[(4-phenylthiazol-2-yl)amino]vinyl)naphthalin-2-ol (19)

A mixture of the appropriate **17** (1.22 g, 5 mmol) and phenacylbromide (1 g, 5 mmol) in ethanol (20 mL) was heated under reflux 3 h then poured onto ice cold water (50 mL) containing ammonium hydroxide (2 drops). The resulting solid was collected and recrystallized from ethanol to give **19** (Tables II and III).

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