Quinazoline Derivatives from 2-Phenyl-4-quinazolinylhydrazine

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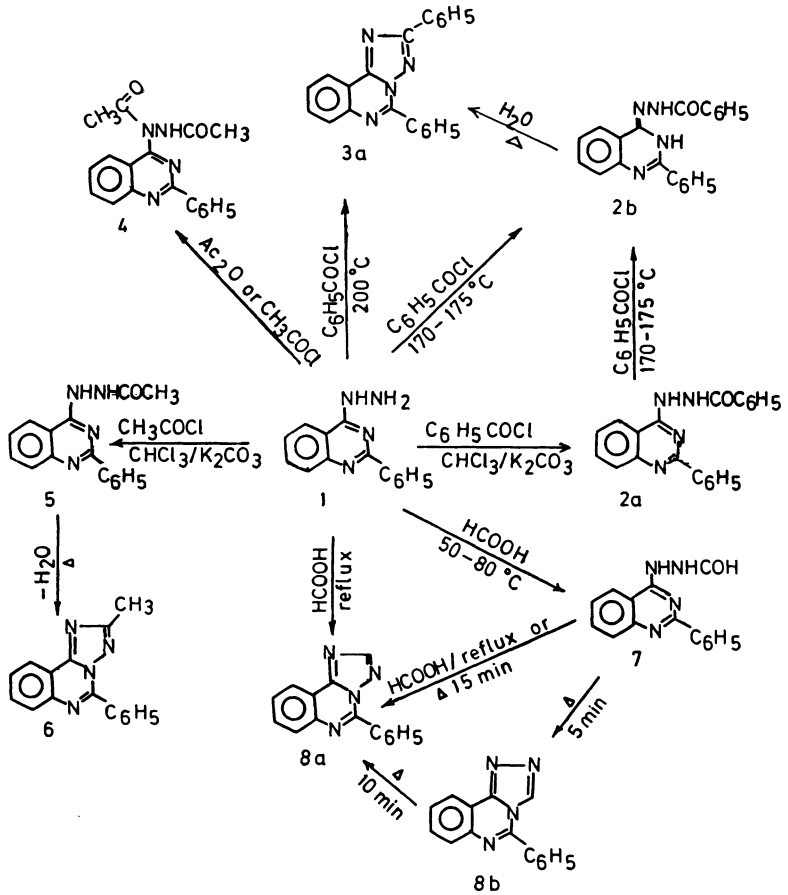
Formic acid and acetyl and benzoyl chloride were treated with 2-phenyl-4-quinazolinylhydrazine (**1**) under mild conditions to afford the corresponding hydrazides which were converted into 5-phenyl[1,2,4]triazolo[1,5-*c*]quinazoline, 2-methyl-5-phenyl[1,2,4]triazolo[1,5-*c*]quinazoline, and 2,5-diphenyl[1,2,4]triazolo[1,5-*c*]quinazoline (**3a**) respectively by heating over their melting points. Reaction of **1** with aldehydes produced the corresponding hydrazones which give the corresponding [1,2,4]triazolo[1,5-*c*]quinazolines (**3a**—**i**) on pyrolysis. Carbon disulfide underwent ring closure with **1** to 5-phenyl-1,2,4-triazolo[4,3-*c*]quinazoline-3-thiol which was readily converted into the corresponding alkylthio compounds by treatment with alkyl halides. Further, 4-(4-arylmethylene-5-oxo-2-phenyl-2-imidazolinylamino)-2-phenylquinazolines were obtained *via* the condensation of 4-arylmethylene-2-phenyl-2-oxazolin-5-ones with **1**.

Literature survey has revealed that some heterocyclic compounds such as 2-hydrazinoquinoxaline which has hydrazino group ortho to ring nitrogen can be cyclized to the corresponding[1,2,4]triazolo[4,3-*a*]quinoxalines[1] *via* the reaction with organic acids, acid chlorides and/or acid anhydrides and the acylated intermediate was not obtained. In some cases, however, only the acylated intermediates were obtained, depending on the nature of heterocyclic compound.[2] The isomerization in ring-fused 1,2,4-triazoles induced by acid or heat has been reported for several of these ring systems.[3,4] A particularly facile isomerization was observed with[1,2,4]triazolo[4,3-*c*]quinazoline system to its [1,2,4]triazolo[1,5-*c*]quinazoline isomer.[5]

It was of interest to mention that the products obtained from the reaction of 2-phenyl-4-quinazolinylhydrazine (**1**) with acyl chlorides, formic acid, or aldehydes depend mainly on the reaction conditions applied.

The reaction of **1** with benzoyl chloride in chloroform containing anhydrous potassium carbonate yielded 2-benzoyl-1-(2-phenyl-4-quinazolinyl)hydrazine (**2a**), while heating a mixture of **1** and benzoyl chloride at 170—175 °C for 1 h, the isomer **2b** was obtained as a major product. Heating of **2a** in benzoyl chloride at 170 °C for 1 h gave the isomer **2b**. Further, heating of both **2a** and **2b** above their melting points for 15 min gave 2,5-diphenyl[1,2,4]triazolo[1,5-*c*]quinazoline (**3a**), which was obtained directly by refluxing a mixture of **1** and benzoyl chloride for 2 h, as shown in



Scheme 1.

Scheme 1.

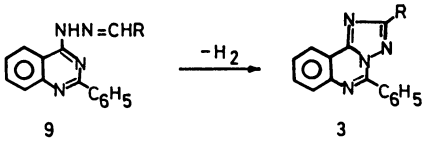
The structures of **2a**, **2b**, and **3a** were established on the basis of their IR, UV, and Mass spectral analysis. Refluxing of **1** with acetic anhydride or acetyl chloride for 2 h gives 1,2-diacetyl-1-(2-phenyl-4-quinazolinyl)hydrazine (**4**). 2-Acetyl-1-(2-phenyl-4-quinazolinyl)hydrazine (**5**) was obtained by the reaction of **1** with acetyl chloride in chloroform containing anhydrous potassium carbonate, which was cyclized to 2-methyl-5-phenyl[1,2,4]triazolo[1,5-*c*]quinazoline (**6**), by heating above its melting point for 15 min (Scheme 1).

It was reported by Potts[5] that 5-phenyl[1,2,4]-triazolo[1,5-*c*]quinazoline (**8a**) was obtained by direct cyclization of **1** with formic acid, while its [4,3-*c*]-quinazoline isomer (**8b**) was achieved by the reaction of **1** with triethyl orthoformate.

In our investigation both isomers **8a** and **8b** were obtained through the formation of *N*-formyl intermediate **7** which was obtained by conducting the reaction of **1** with formic acid at 55—80 °C for 45 min. Heating of **7** over its melting point for 5 min gave **8b**, which was readily isomerized to **8a** by further heating above its melting point for 10 min. On the other hand, **8a** was obtained directly by continuous heating of **7** for 15 min or by refluxing of **7** with formic acid for 1 h.

The UV spectra of **3a**, **6**, and **8a** were characterized by the location of the principal absorption band at shorter wavelength relative to that of **2a**, **5**, and **7** respectively. The observed blue shift (52—58 nm) may be due to the shortening of conjugation as a result of their cyclization to 1,2,4-triazolo ring system.

Reaction of **1** with the appropriate aldehydes in ethanolic solution containing piperidine yielded the corresponding hydrazones (**9a**—**i**). While carrying the same reaction in the absence of solvent at 210—220 °C for 2 h, cyclization took place and the [1,2,4]-triazolo[1,5-*c*]quinazolines (**3a**—**i**) were obtained.



9 3

R	R
a: C_6H_5	f: $C_6H_4CH_3$-p
b: C_6H_4Br-p	g: $C_6H_4NO_2$-p
c: C_6H_4Cl-p	h: $C_6H_4NO_2$-m
d: C_6H_4Cl-m	i: 2-thienyl
e: $C_6H_4OCH_3$-p	

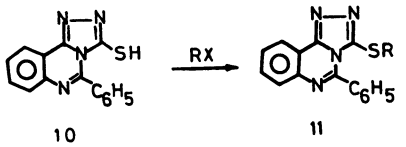
On the other hand, heating of **9a** above its melting point for 1 h, it was cyclized to **3a**. The IR spectra of **9a**—**i** revealed an absorption band at 3370—3350 cm^{-1} attributed to NH group, which disappeared in compounds **3a**—**i**. The UV spectra of **9** were characterized by the presence of a principal absorption band at 362—382 nm, while the UV spectra of **3** exhibited a principal absorption band at 300—310 nm. The observed blue shift (62—72 nm), probably due to the shortening of conjugation as a result of cyclization.

The NMR spectrum of **3e** (in DMSO) showed a singlet at δ=3.8 (3H, OCH_3), a multiplet at 6.8—7.8 (9H, aromatic protons) and another multiplet at 8.4—8.6 (4H, of quinazoline ring); while the NMR spectrum of **3f** showed a singlet at δ=2.6 (3H, CH_3), a multiplet at 7.2—7.8 (9H, aromatic protons) and at 8.2—8.5 (4H, of quinazoline ring).

In the mass spectra of these fused ring systems molecular ions were obtained for all compounds studied.

Further, it was reported that carbon disulfide reacted with 2-pyridylhydrazines[2,6] or 2-thiazolylhydrazines[7] to produce 1,2,4-triazolo[4,3-*a*]pyridine-3-thiols and thiazolo[2,3-*c*]-1,2,4-triazole-3-thiols respectively.

Similarly, reaction of **1** with carbon disulfide provided a convenient synthesis of 5-phenyl-1,2,4-triazolo[4,3-*c*]-quinazoline-3-thiol (**10**), which was readily converted to the corresponding 3-alkylthio compounds (**11a**—**e**) with alkyl halides in basic medium.

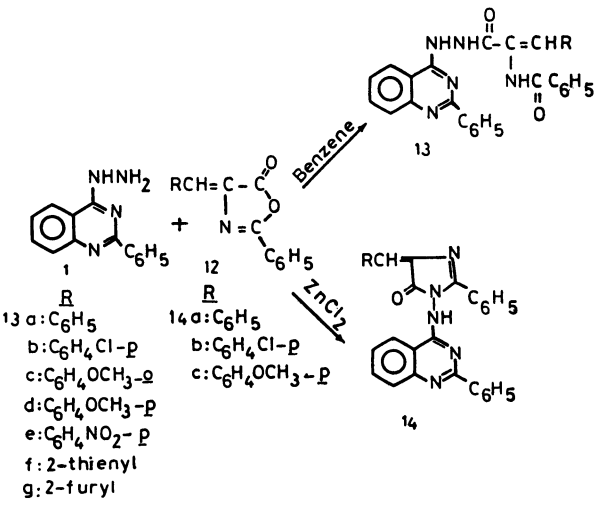


10 11

R
11 a: CH_3
b: C_2H_5
c: $CH_2(CH_2)_2CH_3$
d: $CH_2C_6H_5$
e: $CH_2COOC_2H_5$

The NMR spectrum of **11b** (in $CDCl_3$) showed a triplet at δ=1.3 (J=6 Hz, 3H, CH_3), a quartet at 3.3 (J=6 Hz, 2H, CH_2), and a multiplet at 7.5—8.3 (9H, aromatic protons), while the NMR spectrum of **11d** (in $CDCl_3$) showed signals at δ=4.7 (s, 2H, CH_2) and at 7.4—8.8 (m, 14 H, aromatic protons).

Furthermore, the hitherto unreported reaction of **1** with 4-arylmethylene-2-phenyl-2-oxazolin-5-ones (**12**)[8] in benzene or xylene afforded the corresponding 1-(2-phenyl-4-quinazolinyl)-2-(2-benzamido-3-arylacryloyl)-hydrazine (**13**). The IR spectra of **13** showed two bands characteristic of the stretching vibration of two carbonyl groups at 1680—1670 cm^{-1} and at 1650—1640 cm^{-1} in addition to the NH band appeared at 3320—3290 cm^{-1}.



1 12 13 14

R	R
13 a: C_6H_5	14 a: C_6H_5
b: C_6H_4Cl-p	b: C_6H_4Cl-p
c: $C_6H_4OCH_3$-o	c: $C_6H_4OCH_3$-p
d: $C_6H_4OCH_3$-p	
e: $C_6H_4NO_2$-p	
f: 2-thienyl	
g: 2-furyl	

When **12** was allowed to react with **1** in the presence of anhydrous zinc chloride, the corresponding 4-(4-arylmethylene-5-oxo-2-phenyl-2-imidazolin-1-ylamino)-2-phenylquinazolines (**14a—d**) were produced directly.

On the other hand, **14a** was also obtained by heating of **13a** above its melting point for 5 min in the presence of anhydrous zinc chloride. The IR spectra of **14a—d** showed absorption bands at 1690—1680 cm^{-1} and 3280—3240 cm^{-1} which were assigned to C=O and NH stretching frequencies respectively. The mass spectrum of **14c** showed a molecular ion peak at *m/e* 497, which agreed with the molecular weight expected.

Experimental

Melting points reported are uncorrected. Elemental analysis was carried out on a CHN analyzer. IR spectra were recorded on a Perkin-Elmer infracord. NMR spectra were recorded by a Varian EM 60 MHz spectrometer. Mass spectra were taken on MAT 112 Massenspectrometer.

*2-Phenyl-4-quinazolinylhydrazine (**1**)* was prepared as previously described by Postovskii, mp 215 °C, lit,[9] mp 214—215 °C.

*2-Benzoyl-1-(2-phenyl-4-quinazolinyl)hydrazine (**2a**).* To a solution of **1** (5.9 g, 0.025 mol) in dry chloroform (100 ml) containing anhydrous potassium carbonate (0.5 g), benzoyl chloride (0.02 mol) was added slowly. After the addition was complete, the reaction mixture was stirred at room temperature for 30 min and then heated on a steam bath for 1 h. The mixture was filtered, evaporated and the crude product was collected and crystallized from ethanol as yellow needles, 7 g (82%); mp 248 °C. IR(KBr), 3360 (NH), 3220(NH), 1650 cm^{-1} (C=O); UV(ethanol), λ_{max} 248 (log ε 4.59), 320(4.0), 356 nm (3.84); MS (70 eV), M^+, *m/e* (rel intensity) 340 (100%), 221(53%), 205(38%), 103 (59%), 102(65%). Found: C, 74.11; H, 4.70; N, 16.45%. Calcd for $C_{21}H_{16}N_4O$: C, 74.10; H, 4.47; N, 16.46%.

The isomer **2b** was obtained by heating a mixture of **1** (0.02 mol) and benzoyl chloride (0.04 mol) at 170—175 °C for 1 h. The product was isolated by pouring the reaction mixture into cooled 5% sodium carbonate solution. The crude product crystallized from ethanol as colorless needles, 4.5 g (66%), mp 260 °C. IR (KBr); 3320(NH), 3210 (NH), 1660 cm^{-1} (C=O); UV (ethanol) λ_{max} 232(log ε 4.56), 256 (4.5), 308 nm(4.06); MS (70 eV), M^+, *m/e* (rel intensity) 340 (80%), 322 (100%), 219(35%). Found: C, 73.92; H, 4.83; N, 16.32%. Calcd for $C_{21}H_{16}N_4O$: C, 74.10; H, 4.74; N, 16.46%.

The same product was also obtained by heating of **2a** (1 g) in benzoyl chloride (5 ml) at 170—175 °C for 1 h.

*2,5-Diphenyl[1,2,4]triazolo[1,5-c]quinazoline (**3a**).* A mixture of **1** (4.69 g, 0.02 mol) and benzoyl chloride (0.04 mol) was refluxed for 2 h. The product was isolated as before and crystallized from ethanol as colorless needles, 4.6 g (72%).

The same compound was also obtained by heating of **2a** or **2b** (1 g) over their melting points for 15 min, and crystallized from ethanol, 0.5 g (63%). There is no melting point depression and the infrared spectrum was superimposable on that of **3a** prepared according to the above procedure (Table 2).

*1,2-Diacetyl-1-(2-phenyl-4-quinazolinyl)hydrazine (**4**).* **1** (2.3 g, 0.01 mol) was refluxed with acetic anhydride or acetyl chloride (10 ml) for 2 h. The colorless product separated was collected and crystallized from ethanol as colorless needles, 2.8 g (87%), mp 255 °C; IR (KBr); 3300 (NH), 1700, 1670 cm^{-1} (C=O), NMR, δ=2.5 (s, 3H, CH_3CO), 2.7 (s, 3H, CH_3CO), 3.0 (s, 1H, NH), 7.7—9.0 (m, 9H, aromatic protons).

Found: C, 67.36; H, 5.11; N, 17.28%. Calcd for $C_{18}H_{16}N_4O_2$: C, 67.49; H, 5.03; N, 17.49%.

*2-Acetyl-1-(2-phenyl-4-quinazolinyl)hydrazine (**5**).* It was prepared from **1** (9.2 g, 0.04 mol) and acetyl chloride (0.02 mol) in chloroform solution containing anhydrous potassium carbonate as previously described in **2a**. The crude product was crystallized from ethanol as pale orange prisms, 2 g (74%), mp 252—253 °C. IR (KBr) 3300, 3220 (NH), 1650 cm^{-1} (C=O).

Found: C, 69.18; H, 5.15; N, 20.46%. Calcd for $C_{16}H_{14}N_4O$: C, 69.05; H, 5.07; N, 20.13%.

Refluxing of **5** (1 g) with acetic anhydride or acetyl chloride (5 ml) for 1 h yielded **4** (0.9 g; 75%).

*2-Methyl-5-phenyl[1,2,4]triazolo[1,5-c]quinazoline (**6**).* **5** (1 g) was heated above its melting point for 10 min and after cooling to room temperature, crystallized from methanol as colorless needles, 0.6 g (60%), mp 128 °C; IR (KBr) 3050 (C–H), 1615 cm^{-1} (C=N); UV (ethanol) λ_{max} 259 (log ε 4.38), 264(4.25), 294 (4.3) nm; MS (70 eV), M^+, *m/e* (rel intensity) 260 (100%), 219 (36%); NMR, δ=2.7 (s, 3H, CH_3), 7.5—7.8 (m, 5H, C_6H_5), 8.5—8.8 (m, 4H, quinazoline ring).

Found: C, 73.71; H, 4.56, N, 21.31%. Calcd for $C_{16}H_{12}N_4$: C, 73.83; H, 4.65; N, 21.52%.

*2-Formyl-1-(2-phenyl-4-quinazolinyl)hydrazine (**7**).* A mixture of **1** (1 g) and formic acid (6 ml) was heated at 50—80 °C for 45 min and the excess acid was evaporated under reduced pressure. The crude product was collected and crystallized from ethanol as yellow crystals, 0.8 g (71%), mp 240 °C; IR (KBr) 3270, 3220(NH), 1650 cm^{-1}, (C=O); UV (ethanol), λ_{max} 275 (log ε 3.99), 288 sh (3.86), 350 nm (3.25).

*5-Phenyl-1,2,4-triazolo[4,3-c]quinazoline (**8b**).* **7** (1 g) was heated above its melting point for 5 min and after cooling, crystallized from ethanol as colorless needles, 0.7 g (77%); mp 207 °C, lit[5] mp 205—206 °C.

Found: C, 73.00; H, 4.38; N, 22.41%. Calcd for $C_{15}H_{10}N_4$: C, 73.16; H, 4.09; N, 22.75%.

*5-Phenyl[1,2,4]triazolo[1,5-c]quinazoline (**8a**).* Heating of **7** (1 g) as described above for 15 min gave colorless needles from ethanol 0.8 g (88%), mp 185 °C, lit[5] mp 184—185 °C.

The same product was also obtained by refluxing a mixture of **1** (0.5 g) and formic acid (3 ml) for 1 h as described[5] before, or by heating of **8b** (0.5 g) above its melting point for 10 min. UV (ethanol), λ_{max} 249 (log ε 4.22), 292 nm, (3.89).

Found: C, 73.18; H, 4.16; N, 22.54%. Calcd for $C_{15}H_{10}N_4$: C, 73.16; H, 4.09; N, 22.75%.

*Reaction of 2-Phenyl-4-quinazolinylhydrazine (**1**) with Aldehydes (**9a—i**).* A mixture of **1** (0.01 mol), aromatic aldehyde (0.012 mol), ethanol (30 ml), and piperidine (0.2 ml) was refluxed for 4 h. The separated hydrazone was filtered and crystallized from the same solvent as yellow to red needles. The same products were also obtained by fusion of **1** with the appropriate aldehyde for 2 min (Table 1).

*2-Aryl-5-phenyl[1,2,4]triazolo[1,5-c]quinazolines (**3a—i**).* A mixture of appropriate aldehyde (0.015 mol) and **1** (0.01 mol) was heated at 210—220 °C for 2 h. The pyrolysis residue in the reaction flask was cooled and triturated with cold ethanol. The crude product was dissolved in ethanol–benzene (charcoal), forming colorless needles of **3a—i** (Table 2).

3a was also obtained by heating **9a** for 1 h at 210—215

Table 1. Aromatic aldehyde (2-phenyl-4-quinazolinyl)hydrazones (**9a—i**)

Compd No.	Mp θ_m/°C	Yield %	Mol formula	Calcd (Found)(%) C	H	N	λ_{max}/nm (log ε)
9a	134	81	$C_{21}H_{16}N_4$	77.76 (77.66	4.97 4.89	17.27 17.21)	362 (3.96)
b	170	87	$C_{21}H_{15}N_4Br$	62.54 (62.58	3.75 3.64	13.89 13.68)	370 (4.10)
c	172	86	$C_{21}H_{15}N_4Cl$	70.29 (70.36	4.21 4.11	15.61 15.46)	368 (4.06)
d	162	74	$C_{21}H_{15}N_4Cl$	70.29 (70.03	4.21 4.36	15.61 15.36)	364 (4.14)
e	145	72	$C_{22}H_{18}N_4O$	74.56 (74.31	5.12 5.26	15.81 15.88)	382 (4.00)
f	138	76	$C_{22}H_{18}N_4$	78.08 (78.22	5.36 5.28	16.56 16.41)	364 (3.96)
g	238	89	$C_{21}H_{15}N_5O_2$	68.28 (68.11	4.09 4.01	18.96 18.80)	376 (3.98)
h	213	86	$C_{21}H_{15}N_5O_2$	68.28 (68.06	4.09 4.12	18.96 18.76)	370 (3.74)
i	201	70	$C_{19}H_{14}N_4S$	69.07 (69.16	4.27 4.41	16.96 16.74)	381 (3.77)

Table 2. 2-aryl-5-phenyl[1,2,4]triazolo[1,5-*c*]quinazolines (**3a—i**)

Compd No.	Mp θ_m/°C	Yield %	Mol formula	Calcd (Found)(%) C	H	N	λ_{max}/nm (log ε)
3a[a]	170	65	$C_{21}H_{14}N_4$	78.24 (78.13	4.38 4.29	17.38 17.36)	300 (3.80)
b	235	50	$C_{21}H_{13}N_4Br$	62.86 (62.60	3.27 3.21	13.96 13.74)	300 (3.51)
c	182	70	$C_{21}H_{13}N_4Cl$	70.69 (70.71	3.67 3.56	15.70 15.76)	300 (3.78)
d	165	62	$C_{21}H_{13}N_4Cl$	70.69 (70.81	3.67 3.46	15.70 15.61)	305 (3.35)
e[b]	178	72	$C_{22}H_{16}N_4O$	74.98 (74.81	4.58 4.38	15.90 15.81)	310 (3.74)
f[c]	185	84	$C_{22}H_{16}N_4$	78.47 (78.21	4.75 4.36	16.64 16.51)	303 (3.34)
g	234	45	$C_{21}H_{13}N_5O_2$	68.66 (68.86	3.57 3.46	19.06 19.15)	299 (4.17)
h	175	56	$C_{21}H_{13}N_5O_2$	68.66 (68.71	3.57 3.48	19.06 18.78)	301 (3.90)
i[d]	210	35	$C_{19}H_{12}N_4S$	69.49 (69.18	3.68 3.56	17.06 17.15)	310 (3.69)

a) MS, M^+, *m/e* (rel intensity), 322(100%), 219(30%). b) MS, M^+, *m/e* (rel intensity), 352(100%), 219(39%).
c) MS, M^+, *m/e* (rel intensity), 336(100%), 219(25%). d) MS, M^+, *m/e* (rel intensity), 328(100%), 219(24%).

Table 3. 3-Alkylthio-5-phenyl-1,2,4-triazolo[4,3-*c*]quinazolines (**11a—e**)

Compd No.	Mp θ_m/°C	Yield %	Mol formula	Calcd (Found)(%) C	H	N	S
11a	140	64	$C_{16}H_{12}N_4S$	65.73 (65.63	4.14 4.36	19.16 19.28	10.97 10.47)
b	185	68	$C_{17}H_{14}N_4S$	66.64 (66.48	4.61 4.56	18.29 18.42	10.47 10.31)
c	163	72	$C_{19}H_{18}N_4S$	68.24 (68.02	5.43 5.48	16.75 16.61	9.59 9.26)
d[a]	101	52	$C_{22}H_{16}N_4S$	71.72 (71.94	4.38 4.16	15.21 15.46	8.70 8.66)
e	186	77	$C_{19}H_{16}N_4O_2S$	62.62 (62.43	4.43 4.26	15.37 15.18	8.80 9.12)

a) Recrystallized from benzene-pet. ether (60—80 °C).

Table 4. 1-(2-Phenyl-4-quinazolinyl)-2-(2-benzamido-3-arylacryloyl)hydrazines (**13a—g**)

Compd No.	Mp θ_m/°C	Yield %	Mol formula	Calcd(Found)(%) C	H	N
13a	220	82	$C_{30}H_{23}N_5O_2$	74.21 (74.00	4.77 4.55	14.42 14.90)
b	185	86	$C_{30}H_{22}ClN_5O_2$	69.30 (69.20	4.26 4.03	13.47 13.25)
c	190	91	$C_{31}H_{25}N_5O_3$	72.22 (72.11	4.89 4.81	13.58 13.47)
d	228	76	$C_{31}H_{25}N_5O_3$	72.22 (72.20	4.89 4.79	13.58 13.50)
e	229	88	$C_{30}H_{22}N_6O_4$	67.92 (67.70	4.18 4.07	15.84 15.88)
f	227	85	$C_{28}H_{21}N_5O_2S$	68.42 (68.40	4.31 4.26	14.25 14.16)
g	199	86	$C_{28}H_{21}N_5O_3$	70.73 (70.71	4.45 4.52	14.73 14.83)

Table 5. 4-(4-Arylmethylene-5-oxo-2-phenyl-2-imidazolin-1-ylamino)-2-phenylquinazolines (**14a—d**)

Compd No.	Mp θ_m/°C	Yield %	Mol formula	Calcd (Found)(%) C	H	N
14a	248	45	$C_{30}H_{21}N_5O$	77.07 (77.18	4.53 4.50	14.98 14.88)
b	239	56	$C_{30}H_{20}ClN_5O$	71.78 (71.76	4.02 4.13	13.95 13.90)
c	264	42	$C_{31}H_{23}N_5O_2$	74.83 (74.70	4.66 4.56	14.08 14.18)
d	265	58	$C_{28}H_{19}N_5OS$	71.02 (71.31	4.04 4.22	14.79 14.81)

°C. The pyrolysis residue, treated as described above yielded **3a**, no depression of melting point occurred on admixture with the sample prepared by the reaction of **1** with benzoyl chloride.

*5-Phenyl-1,2,4-triazolo[4,3-c]quinazoline-3-thiol (**10**).* **1** (2 g), methanol (100 ml), potassium hydroxide (0.7 g), and carbon disulfide (6 ml) were refluxed for 4 h. After removal of the excess solvent, dilute potassium hydroxide was added and the alkaline solution was filtered and acidified. The crude product was collected and crystallized from ethanol–benzene mixture as colorless needles, 1.89 g (75%), mp 265—268 °C; UV (ethanol) λ_{max} 290 nm (log ε 3.97).

Found: C, 64.45; H 3.58; N 20.36; S 11.50%. Calcd for $C_{15}H_{10}N_4S$: C, 64.73; H, 3.62; N, 20.13; S, 11.52%.

*Reaction of **10** with Alkyl Halides (**11a—e**).* A solution of **10** (0.59 g, 0.002 mol) and sodium hydroxide (0.1 g) in water (10 ml) was stirred while the appropriate alkyl halide (0.004 mol) was added dropwise. The reaction mixture was stirred for 2 h, then the excess alkyl halide was evaporated and the residue crystallized from ethanol, forming colorless needles of the products **11a—e** (Table 3).

*1-(2-Phenyl-4-quinazolinyl)-2-(2-benzamido-3-arylacryloyl)hydrazines (**13a—g**).* A mixture of **1** (1 g; 0.004 mol) and 4-arylmethylene-2-phenyl-2-oxazolin-5-one (**12**) (0.005 ml) in 30 ml dry benzene or ethanol was refluxed for 1 h. The reaction mixture was cooled and the product was filtered and recrystallized from ethanol to give **13a—g** nearly in a quantitative yield (Table 4).

*4-(4-Arylmethylene-5-oxo-2-phenyl-2-imidazolin-1-ylamino)-2-phenylquinazolines (**14a—d**).* A mixture of **1** (1 g; 0.004 mol), **12** (0.005 mol), and 0.1 g of anhydrous zinc chloride was fused together for 5 min. Benzene was added to the reaction mixture and filtered. The product was recrystallized from benzene–ethanol mixture to give **14a—d** in 40—60% yield (Table 5). **14c** was also obtained by heating **13d** above its melting point for 5 min in the presence of anhydrous zinc chloride.

References

1) D. Shiho and S. Tagami, *J. Am. Chem. Soc.*, **82**, 4044 (1960).

2) K. T. Potts and H. R. Burton, *J. Org. Chem.*, **31**, 251 (1966); D. Shiho and S. Tagami, *Yakugaku Zasshi*, **76**, 804 (1965).

3) G. W. Miller and F. L. Rose, *J. Chem. Soc.*, **1965**, 3357, 3369; K. T. Potts, H. R. Burton, and S. K. Roy, *J. Org. Chem.*, **31**, 265 (1966); K. T. Potts and S. W. Schneller, *J. Heterocycl. Chem.*, **5**, 485 (1968).

4) J. A. Bee and F. L. Rose, *J. Chem. Soc., C.*, **1966**, 2031; G. W. Miller and F. L. Rose, *ibid.*, **1966**, 5642.

5) K. T. Potts and E. G. Brugel, *J. Org. Chem.*, **35**, 3448 (1970).

6) D. S. Tarbell, C. W. Todd, M. C. Paulson, E. G. Linstrom, and V. P. Wystrack, *J. Am. Chem. Soc.*, **70**, 1381 (1948).

7) K. T. Potts and S. Husain, *J. Org. Chem.*, **36**, 10 (1971).

8) M. Crawford and W. T. Little, *J. Chem. Soc.*, **1959**, 729; E. Erlenmeyer, *Justus Liebigs Ann. Chem.*, **275**, 1 (1893).

9) I. Ya. Postovskii, N. N. Vereschagina, and S. L. Mertsalov, *Khim. Geterotsikl. Soedin., Akad. Nauk Latv. SSR*, **1966**, 130.