Reaction of 1,3,4-Thiadiazolo and 1,3,4-Triazolo[3,2-*c*]quinazoline Derivatives With Active Methylene Compounds

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Fused quinazoline derivatives **1** and **4** react with active methylene compounds and depending on the annelated five-membered ring, two types of transformations have been observed. The 1,3,4-thiadiazolo[3,2-*c*]quinazoline **1**, underwent the five-membered ring opening reaction to afford the 3,4-dihydroquinazolines **2** in good yields, whereas the 1,3,4-triazolo[3,2-*c*]quinazoline **4** underwent nucleophilic attack at 2-position of the quinazoline ring to yield the corresponding 1,2,4-triazoles **5**.

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Recently, it has been reported [1] that the high reactivity at position 2 of the pyrimidine ring in fused pyrido[2,3-*d*]pyrimidines or pyrido[2,3-*d*]pyrimidine 3-oxides, results in ring opening. These results prompted us to investigate in detail the reactivity of fused quinazolines such as 5-phenyl-2-methylthio-1,3,4-thiadiazolo[3,2-*c*]quinazolin-4-ium iodide **1** and 1-aryl-5-phenyl-2-methylthio-1,3,4-triazolo[3,2-*c*]quinazolin-4-ium iodide **4** toward various nucleophiles, in particular toward active methylene compounds.

Compound **1**, readily available from 3-amino-5-phenyl-4-thioxo-3,4-dihydroquinazoline, carbon disulfide and methyl iodide [2], reacts with a series of acetonitriles activated by another electron-delocalizing group such as an ester, amide or a second nitrile group and 1,3-dicarbonyl compounds, in the presence of base to yield the corresponding 3,4-dihydroquinazolines **2** in good yields (Table I).

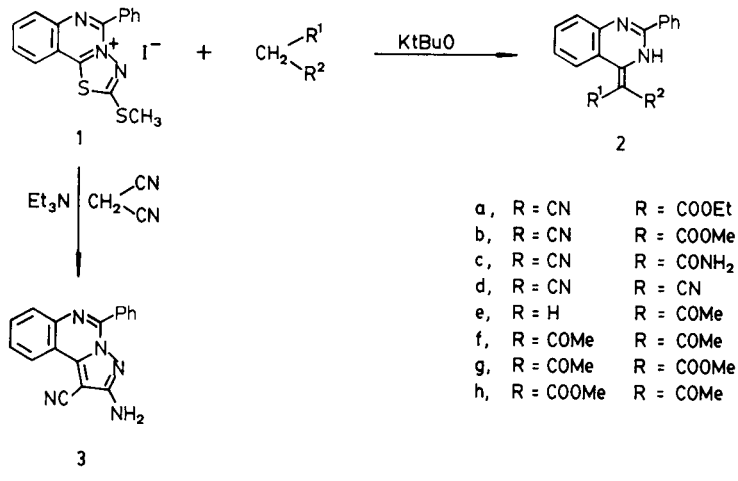
For ethyl cyanoacetate, methyl cyanoacetate and cyanoacetamide, the best results are obtained when the reaction is carried out in the presence of potassium *t*-butoxide with acetonitrile as the solvent. Attempts with weaker bases such as triethylamine and pyrrolidine were unsuccessful.

Compound **1** reacts with malononitrile in the presence of potassium *t*-butoxide to give a mixture of 3,4-dihydroquinazoline **2d** ($R^1 = R^2 = CN$) and 2-amino-3-cyanopyrazolo-[1,5-*c*]quinazoline **3** (44%). This latter compound can be obtained as the only reaction product in good yield (56%) from **1**, malononitrile and triethylamine as base. The structure of **3** follows from its proton nmr, ir and ms spectra as well as from analytical data.

Compound **1** reacts with 2,4-pentanedione and the nature of the reaction product is dependent upon the base. Thus, with potassium *t*-butoxide, it leads to **2e** while with triethylamine it leads to **2f**. On the other hand, compound **1** reacts with methyl acetoacetate to give the corresponding 3,4-dihydroquinazoline derivative. From the proton nmr spectral data it can be concluded that the reaction afforded a mixture of **2g** and **2h** isomers.

The ir spectra of compounds **2a-d** show absorption in the region 2190-2200 cm^{-1} due to the stretching vibration of the cyano group, in addition compounds **2a-c** show absorption around 1650 cm^{-1} due to the carbonyl group. Mass spectra of compounds **2** show the expected molecular ion peak and the fragmentation pattern is in accord with

Scheme 1



1 2 3

a, R = CN R = COOEt
b, R = CN R = COOMe
c, R = CN R = $CONH_2$
d, R = CN R = CN
e, R = H R = COMe
f, R = COMe R = COMe
g, R = COMe R = COOMe
h, R = COOMe R = COMe

Table I

3,4-Dihydroquinazoline Derivatives **2**

Compound No.	Yield (%)	Mp °C	Appearance Solvent	Reaction Time (hours)	Molecular Formula	Analyses (%) Calcd. C	H	N	Found C	H	N
2a	81	179-181	yellow prisms acetonitrile	3	$C_{19}H_{15}N_3O_2$ 317.35	71.91	4.76	13.24	72.10	4.81	13.11
2b	86	206-209	yellow prisms acetonitrile	4	$C_{18}H_{13}N_3O_2$ 303.32	71.27	4.32	13.85	71.10	4.34	14.05
2c	93	228-230	yellow prisms ethanol	15	$C_{17}H_{12}N_4O$ 288.31	70.82	4.19	19.43	71.02	4.13	19.28
2d	24	330-335 dec	yellow prisms ethanol	3	$C_{17}H_{10}N_4$ 270.30	75.54	3.73	20.73	75.42	3.84	20.52
2e	63	138-140	yellow prisms chloroform	6	$C_{17}H_{14}N_2O$ 262.31	77.84	5.38	10.68	77.62	5.36	10.50
2f	74	142-144	brown prisms chloroform	6	$C_{19}H_{16}N_2O_2$ 304.35	74.98	5.30	9.20	74.86	5.41	9.25

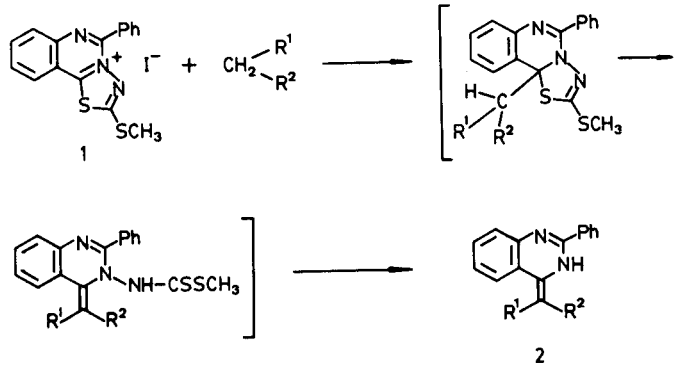
Table II

Spectral Data of Compounds **2**

Compound No.	IR (cm^{-1})	^{1}H-NMR (δ ppm) [a]	MS m/e (%)
2a	3170, 3080, 2208, 1650, 1596, 1569, 1471, 1280, 1253, 1146, 1022, 848, 826, 763, 692	14.9 (1H, s, broad), 9.35 (1H, d), 8.4-7.3 (9H, m), 4.4 (2H, q), 1.45 (3H, t)	318 (M^++1, 10), 317 (M^+, 91), 272 (27), 271 (67), 246 (26), 245 (61), 217 (11), 206 (16), 205 (100), 114 (50), 104 (35), 102 (42), 77 (75), 76 (41), 29 (69)
2b	3270, 3090, 2202, 1647, 1596, 1545, 1279, 1251, 1138, 934, 826, 764, 690	15.2 (1H, s), 9.57 (1H, d), 8.6-7.5 (9H, m), 4.05 (3H, s)	304 (M^++1, 10), 303 (M^+, 98), 272 (35), 271 (79), 245 (8), 244 (10), 243 (19), 206 (17), 205 (100), 102 (60), 77 (67)
2c	3477, 3437, 3369, 3216, 2188, 1609, 1582, 1469, 1397, 1379, 768, 706, 687, 646	9.5 (1H, d), 8.5-7.8 (9H, m), 7.35 (2H, s, broad)	289 (M^++1, 2), 288 (M^+, 56), 287 (8), 272 (17), 271 (71), 243 (15), 206 (16), 205 (100), 102 (53), 77 (61)
2d	3256, 3143, 2216, 2197, 1623, 1605, 1569, 1509, 1464, 1341, 1160, 751, 696		271 (M^++1, 9), 270 (M^+, 100), 269 (28), 244 (10), 243 (3), 206 (5), 205 (33), 77 (45), 76 (35), 51 (29), 50 (89)
2e	1611, 1583, 1484, 1357, 1272, 1135, 980, 853, 768, 688	8.4-7.3 (10H, m), 6.05 (1H, s), 2.25 (3H, s)	262 (M^+, 65), 247 (100), 220 (51), 205 (18), 204 (16), 165 (24), 117 (38), 103 (35), 77 (85), 43 (84)
2f	1615, 1562, 1540, 1487, 1377, 1029, 963, 822, 774, 708, 686	9.0-7.6 (10H, m), 1.9 (6H, s)	304 (M^+, 27), 303 (27), 289 (19), 261 (6), 247 (47), 219 (8), 205 (13), 102 (22), 89 (17), 77 (47), 43 (100)
2g + 2h	3300, 3150, 1654, 1617, 1569, 1540, 1439, 1338, 1245, 1067, 840, 783, 710, 690	15.0 (1H, s), 9.0-7.4 (9H, m), 3.9 (1.5H, s), 3.70 (1.5H, s), 2.45 (1.5H, s), 1.95 (1.5H, s)	320 (M^+, 9), 305 (9), 278 (8), 247 (10), 246 (21), 220 (10), 206 (6), 205 (29), 102 (19), 77 (35), 76 (25), 59 (7), 43 (100)

[a] Solvent, deuteriochloroform for **2a**, **2b**, **2e**, **2f** and **2g + 2h**; deuteriochloroform-trifluoroacetic acid for **2c**.

Scheme 2



1 2

Table III

1-Aryl-5-phenyl-2-methylthio-1,3,4-triazolo[3,2-c]quinazolin-4-ium Iodides **4**

Compound No.	Ar	Yield (%)	Mp °C	Molecular Formula	Analyses (%) Calcd. C	Calcd. H	Calcd. N	Found C	Found H	Found N
4a	4-$H_3CC_6H_4$	55	278-279	$C_{23}H_{19}N_4IS$ 510.40	54.12	3.75	10.98	54.37	3.71	11.22
4b	C_6H_5	76	286-287	$C_{22}H_{17}N_4IS$ 496.35	53.23	3.45	11.29	53.45	3.40	11.41
4c	4-BrC_6H_4	75	276-277	$C_{22}H_{16}BrN_4IS$ 575.27	45.93	2.80	9.74	46.09	2.87	9.91
4d	4-ClC_6H_4	45	280-281	$C_{22}H_{16}ClN_4IS$ 530.82	49.78	3.04	10.55	49.50	2.89	10.72

Table IV

Spectral Data of Compounds **4**

Compound No.	IR (cm^{-1})	^1H-NMR (δ ppm) [a]	MS m/e (%)
4a	1625, 1562, 1528, 1279, 1143, 832, 764, 718, 707, 645	8.8-7.4 (13H, m), 2.9 (3H, s), 2.65 (3H, s)	368 (M^+-ICH_3, 30), 367 (26), 336 (10), 335 (10), 310 (5), 205 (66) 142 (100), 127 (48), 102 (37), 91 (20), 77 (55)
4b	1625, 1562, 1534, 1279, 775, 707, 690, 622	7.8-6.8 (14H, m), 2.9 (3H, s)	354 (M^+-ICH_3, 33), 353 (30), 322 (12), 321 (16), 296 (12), 277 (5), 205 (80), 142 (100), 127 (61), 102 (46), 77 (95)
4c	1624, 1602, 1568, 1534, 1421, 1279, 1143, 1070, 1010, 826, 764, 701	8.8-7.2 (13H, m), 2.9 (3H, s)	434 (M^++2-ICH_3, 13), 433 (14), 432 (M^+-ICH_3, 13), 431 (12), 401 (3), 399 (4), 294 (13), 205 (49), 142 (100), 127 (49), 102 (19), 77 (22)
4d	1625, 1562, 1534, 1506, 1274, 1143, 1087, 949, 764, 707, 628	8.6-7.3 (13H, m), 2.9 (3H, s)	390 (M^++2-ICH_3, 9), 389 (10), 388 (M^+-ICH_3, 25), 387 (27), 356 (4), 294 (7), 277 (6), 205 (46), 142 (100), 127 (50), 102 (20), 77 (28)

[a] Solvent, deuteriochloroform-trifluoroacetic acid for **4a**, **4c** and **4d**; dimethylsulfoxide-d_6 for **4b**.

the proposed structure. Salient features of the proton nmr spectra are given in the Table II.

We believe that the formation of **2** involves nucleophilic attack of the active methylene compound on the 4-position of the quinazoline ring followed by ring-opening of the five-membered ring and cleavage of the N-N bond.

Compound **1**, reacts with aromatic amines to give the corresponding 1-aryl-5-phenyl-2-methylthio-1,3,4-triazolo-[3,2-*c*]quinazolin-4-ium iodides **4** in good yields. This reaction is similar to that reported for the conversion of monocyclic 2-methylthio-1,3,4-thiadiazolium into 2-methylthio-1,3,4-triazolium [3]. Compounds **4** can be also prepared from 3-amino-2-phenyl-4-thioxo-3,4-dihydroquinazoline, arylisothiocyanates and methyl iodide [4].

Compound **4** (Ar = 4-H_3C-C_6H_4), reacts with carbon nucleophiles such as malononitrile, ethyl cyanoacetate, methyl cyanoacetate and cyanoacetamide in the presence of base to give the corresponding 1,2,4-triazoles **5** as crystalline solids in good yields (Table V). It appears that the annelated 1,2,4-triazole ring influences the reactivity at posi-

Scheme 3

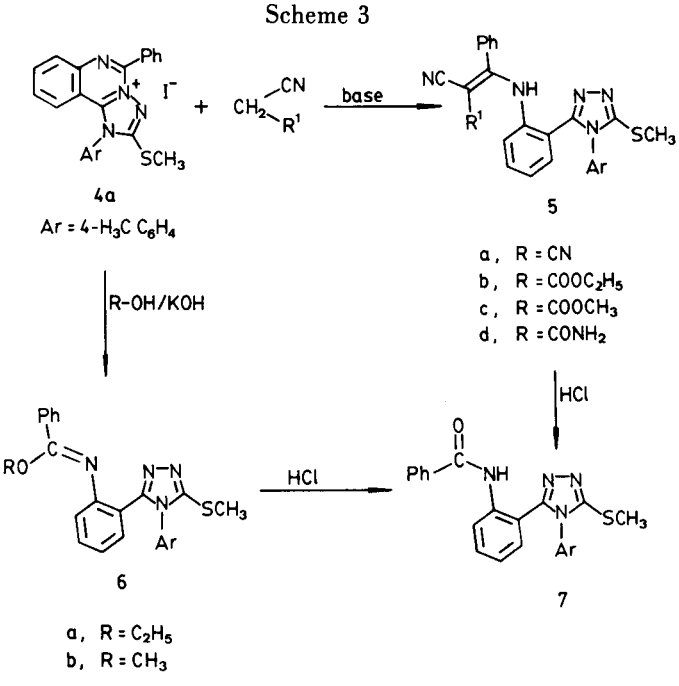


Table V

1,2,4-Triazole Derivatives **5** and **6**

Compound No.	Yield (%)	Mp °C	Appearance Solvent	Reaction Conditions Base	Time (hours)	Temperature
5a	67	242-244	yellow prisms chloroform-ethanol	Et_3N	24	reflux
5b	58	246-248	white needles ethanol	pyrrolidine	16	reflux
5c	91	235-237	white needles chloroform-ethanol	K *t*-BuO	70	rt
5d	55	239-240	white needles ethanol	K *t*-BuO	23	rt
6a	50	78-80	white needles ethanol	KOH	5	rt
6b	87	63	white needles ethanol	KOH	2	rt

Table V (continued)

Compound No.	Molecular Formula	Analyses (%) Calcd. C	H	N	Found C	H	N
5a	$C_{26}H_{20}N_6S$ 448.55	69.62	4.49	18.74	69.39	4.32	18.87
5b	$C_{28}H_{24}N_5O_2S$ 494.6	68.00	4.89	14.16	68.12	4.75	14.26
5c	$C_{27}H_{23}N_5O_2S$ 481.58	67.34	4.81	14.54	67.22	4.85	14.51
5d	$C_{26}H_{22}N_6OS$ 466.57	66.93	4.75	18.01	67.12	4.91	17.91
6a	$C_{25}H_{24}N_4OS$ 428.56	70.07	5.64	13.07	69.83	5.48	12.91
6b	$C_{24}H_{22}N_4OS$ 414.54	69.54	5.35	13.52	69.48	5.23	13.64

tion 2 of the quinazoline part. Similarly, compound **4** reacts with alcohols in the presence of base to give **6**. The above results show the so far unrevealed high reactivity at position 2 of the pyrimidine ring in fused 1,3,4-triazolo-[3,2-*c*]quinazoline derivatives.

Of interest is also the behaviour of the 1,2,4-triazoles **5** and **6**. When treated with hydrochloric acid they were transformed into the 1,2,4-triazole **7**.

EXPERIMENTAL

The melting points were determined with a Kofler hot stage microscope and are uncorrected. The ir spectra were recorded in mineral oil mulls with a Nicolet-FT 5DX instrument. The proton nmr spectra were recorded with a Varian EM-360 instrument with TMS as the internal

Table VI

Spectral Data of Compounds **5** and **6**

Compound No.	IR (cm^{-1})	^1H-NMR (δ ppm) [a]	MS m/e (%)
5a	2215, 1616, 1589, 1562, 1509, 1463, 1410, 1377, 1303, 977, 825, 779, 752, 719, 699	9.15 (1H, s), 8.0-7.1 (13H, m), 2.85 (3H, s), 2.45 (3H, s)	448 (M^+, 51), 421 (5), 383 (94), 293 (10), 205 (100), 194 (20), 192 (12), 91 (58), 77 (80)
5b	2214, 1685, 1587, 1560, 1496, 1462, 1377, 1263, 1132, 767, 723, 700	7.7-7.1 (13H, m), 4.45 (2H, q), 2.9 (3H, s), 2.45 (3H, s), 1.4 (3H, t)	495 (M^+, 32), 422 (27), 383 (100), 335 (42), 205 (58), 192 (4), 164 (11), 102 (16), 91 (14), 77 (32)
5c	2208, 1693, 1613, 1585, 1557, 1511, 1449, 1381, 1268, 1189, 1132, 1007, 973, 832, 786, 758, 735, 718	7.8-7.3 (13H, m), 3.95 (3H, s), 2.8 (3H, s), 2.4 (3H, s)	481 (M^+, 35), 422 (15), 383 (100), 335 (10), 205 (65), 192 (10), 164 (19), 102 (15), 77 (27)
5d	3471, 3318, 3188, 2202, 1653, 1619, 1579, 1511, 1455, 1438, 1359, 1296, 1268, 1223, 1109, 973, 871, 820, 758, 707, 662	12.9 (1H, s), 8.0-6.7 (13H, m), 2.65 (3H, s), 2.35 (3H, s)	466 (M^+, 10), 423 (25), 422 (21), 303 (100) 335 (11), 205 (20), 192 (8), 164 (7), 102 (5), 91 (8), 77 (11)
6a	1653, 1602, 1557, 1511, 1279, 1160, 1115, 1020, 820, 769, 701, 690	8.8-6.9 (12H, m), 6.5 (1H, m), 4.25 (2H, q), 2.63 (3H, s), 2.25 (3H, s), 1.25 (3H, t)	428 (M^+, 2), 399 (2), 385 (8), 384 (25), 383 (100), 337 (5), 310 (89), 222 (20), 205 (17), 91 (10), 77 (15)
6b	1653, 1602, 1574, 1517, 1285, 1177, 1115, 973, 894, 820, 764, 741, 724, 701	7.8-6.9 (12H, m), 6.5 (1H, m), 3.88 (3H, s), 2.75 (3H, s), 2.28 (3H, s)	414 (M^+, 5), 384 (26), 383 (100), 367 (5), 337 (6), 310 (5), 205 (7), 91 (5), 77 (8)

[a] Solvent, deuteriochloroform-trifluoroacetic acid for **5a** and **5b**; deuteriochloroform for **5c** and dimethylsulfoxide-d_6 for **5d**, **6a** and **6b**.

standard. Mass spectra were obtained with a Hewlett-Packard 5993C GC/MS system; compounds were introduced through the direct insertion probe. Microanalyses were performed with a Perkin-Elmer 240C instrument.

Preparation of Compounds **2a**, **2b** and **2c**.

To a stirred solution of the nitrile (3.2 mmoles) in dry acetonitrile (50 ml), potassium *t*-butoxide (0.36 g, 3.2 mmoles) was added under nitrogen. The resultant solution was stirred for 30 minutes at 30° and a solution of 5-phenyl-2-methylthio-1,3,4-thiadiazolo[3,2-*c*]quinazolin-4-ium iodide **1** (0.70 g, 1.6 mmoles) in dry acetonitrile (10 ml) was added. The deep red solution was refluxed for 3-15 hours (Table I) and allowed to warm to room temperature. The precipitated solid was collected by filtration and recrystallized from the appropriate solvent gave **2a-c** as crystalline solids.

Similar results can be obtained when dry dimethylformamide was used as solvent.

Reaction of **1** with Malononitrile. Procedure A.

To a stirred solution of malononitrile (0.181 g, 2.74 mmoles) in dry acetonitrile (10 ml), potassium *t*-butoxide (0.31 g, 2.74 mmoles) was added under nitrogen. The resultant solution was stirred for 15 minutes at room temperature and a solution of **1** (0.60 g, 1.37 mmoles) in dry acetonitrile (10 ml) was added. The reaction mixture was refluxed for 3 hours and allowed to warm to room temperature. The precipitated solid was collected by filtration, washed with cooled ethanol (10 ml), and treated with hot chloroform-benzene (1:1, w:w) (25 ml). The insoluble material was separated by filtration and recrystallized from ethanol gave **2d** (24%). The filtrate was kept at 0° overnight and the precipitated solid was found to be the 2-amino-3-cyanopyrazolo[1,5-*c*]quinazoline **3** (54%).

Procedure B.

To a well stirred solution of malononitrile (0.15 g, 2.28 mmoles) in dry acetonitrile (10 ml), triethylamine (0.23 g, 2.28 mmoles) and **1** (1.0 g, 2.28 mmoles) were added. The reaction mixture was refluxed for 24 hours. After cooling to room temperature the precipitated solid was collected by filtration and recrystallized from chloroform-benzene (1:1, w:w) gave **3** as yellow prisms in 56% yield; ir (Nujol): ν 3325, 3222, 2214, 1636, 1557, 1517, 1376, 758, 719, 696; ms: m/e (relative intensity), 285 (M^+, 17), 271 (20), 270 (100), 244 (11), 205 (27), 140 (12), 104 (10), 102 (18), 77 (26).

Anal. Calcd. for $C_{17}H_{11}N_5$: C, 71.57; H, 3.86; N, 24.56. Found: C, 71.66; H, 4.01; N, 24.36.

Reaction of **1** with 2,4-Pentanedione. Procedure A.

To a stirred solution of 2,4-pentanedione (0.74 g, 7.73 mmoles) in dry acetonitrile (25 ml), potassium *t*-butoxide (0.83 g, 7.73 mmoles) was added. The solution was stirred for 30 minutes at room temperature under nitrogen. Then a solution of **1** (0.65 g, 1.48 mmoles) in dry acetonitrile (10 ml) was added. The reaction mixture refluxed for 6 hours. After cooling to room temperature, the precipitated solid was separated by filtration and recrystallized from chloroform gave **2e**.

Procedure B.

To a stirred solution of 2,4-pentanedione (0.23 g, 2.28 mmoles) in dry acetonitrile (20 ml), triethylamine (0.23 g, 2.28 mmoles) and **1** (0.5 g, 1.14 mmoles) were added. The reaction mixture was refluxed for 6 hours. Work-up was similar to the above procedure to give **2f**.

Reaction of **1** with Methyl Acetoacetate.

To a well stirred solution of methyl acetoacetate (0.37 g, 3.2 mmoles) in dry acetonitrile (15 ml), potassium *t*-butoxide (0.36 g, 3.2 mmoles) was added. The deep red solution was stirred for 30 minutes at room temperature under nitrogen. Then a solution of **1** (0.7 g, 1.6 mmoles) in dry acetonitrile (10 ml) was added. The reaction mixture was refluxed for 15 hours. After cooling to room temperature, the solvent was removed off under reduced pressure and the crude product recrystallized from ethanol gave a mixture of the isomers **2g** and **2h** in 83% yield (Table II).

Reaction of **1** with Aromatic Amines. Preparation of 1-Aryl-5-phenyl-2-methylthio-1,3,4-triazolo[3,2-*c*]quinazolin-4-ium Iodides **4**.

To a solution of 5-phenyl-2-methylthio-1,3,4-thiadiazolo[3,2-*c*]quinazolin-4-ium iodide **1** (0.88 g, 2 mmoles) in toluene (20 ml), the appropriate arylamine was added. The reaction mixture was refluxed for 24 hours and the colour turned deep red. After cooling to room temperature the mixture was kept at 0° overnight. The precipitated solid was collected by filtration and recrystallized from ethanol-chloroform (1:1, w:w) gave **4** as crystalline solids (Table III).

Reaction of Compound **4a** with Nitriles. Preparation of Compounds **5**. Procedure A.

To a well stirred solution of compound **4a** (0.41 g, 1 mmole) in dry acetonitrile (10 ml), the appropriate nitrile (1 mmole) and base (2 mmoles) were added. The reaction mixture was refluxed for the time indicated in the Table V. After cooling to room temperature the precipitated solid was separated by filtration and recrystallized from adequate solvent gave **5a** or **5b**.

Procedure B.

To a solution of the nitrile (1 mmole) in dry dimethylformamide (10 ml), potassium *t*-butoxide (5 mmoles) was added. The reaction mixture was stirred at room temperature for 45 minutes under nitrogen. Then a solution of **4a** (1 mmole) in dry dimethylformamide (10 ml) was added. The mixture was stirred at room temperature for the time indicated in the Table V and then it was poured into cold water (20 ml) and the precipitated solid was separated by filtration and recrystallized from the adequate solvent gave **5c** or **5d**.

Reaction of Compound **4a** with Alcohols. Preparation of **6**.

To a 5% solution of potassium hydroxide in the appropriate alcohol, compound **4a** (0.82 g, 2 mmoles) was added. The reaction mixture which adquires a deep yellow-green colouration was stirred at room temperature for 2-5 hours. The solvent was removed under reduced pressure and the solid residue was washed with water and recrystallized from ethanol gave **6a** or **6b** (Table V).

Hydrolysis of Compounds **5** and **6**. Preparation of **7**.

To a solution of compound **5** or **6** (1 mmole) in methanol (20 ml) 2*N* hydrochloric acid (20 ml) was added. The resultant solution was refluxed for 7 hours. After cooling to room temperature the precipitated solid was collected by filtration and recrystallized from ethanol gave **7** (86%) as colourless prisms, mp 218-220°; ir (Nujol): ν max 1670, 1619, 1591, 1551, 1323, 1274, 1030, 973, 832, 769, 742, 724, 707 cm^{-1}; ^{1}H-nmr (deuteriochloroform): δ 12.15 (1H, s), 8.9 (1H, d), 8.5-6.8 (12H, m), 2.72 (3H, s), 2.42 (3H, s); ms: m/e (relative intensity) 400 (M^+, 100), 399 (10), 383 (17), 372 (28), 323 (83), 105 (22), 91 (3), 77 (13).

Anal. Calcd. for $C_{23}H_{20}N_4OS$: C, 71.84; H, 5.24; N, 14.57. Found: C, 72.02; H, 5.31; N, 14.31.

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