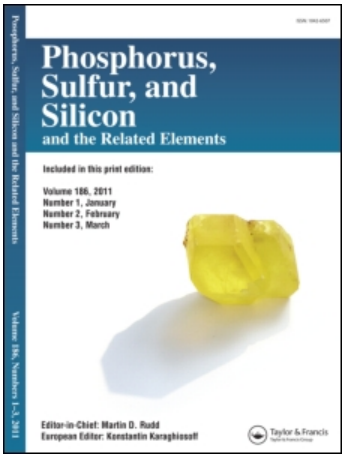

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REACTIONS WITH 2- (THIOCYANATOACETYL) AND 2-(SELENOCYANATOACETYL)-2'-BENZOFURAN: SYNTHESIS OF SOME NEW THIADIAZOLINE, SELENODIAZOLINE, THIA-DIAZOLO[2,3-b]QUINAZOLINE AND ARYLAZOTHIAZOLE DERIVATIVES

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REACTIONS WITH 2-(THIOCYANATOACETYL) AND 2-(SELENOCYANATOACETYL)-2′-BENZOFURAN: SYNTHESIS OF SOME NEW THIADIAZOLINE, SELENODIAZOLINE, THIADIAZOLO[2,3-*b*]QUINAZOLINE AND ARYLAZOTHIAZOLE DERIVATIVES.

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Several new thiadiazoline, selenodiazoline, thiadiazolo[2,3-b]quinazoline and arylazothiazole derivatives were synthesized via the reaction of 2-(thiocyanatoacetyl)-2′-benzofuran with arendiazonium chlorides, nitrous acid, acetic anhydride, benzoyl chloride, diazotised anthranilic acid and its methyl ester and thiourea. The structures of the newly synthesized derivatives were established on the basis of elemental analyses and spectral data studies.

Key words: Thiadiazoline; selenadiazoline; thiazole; benzofuran.

INTRODUCTION

A few 1,3,4-thiadiazole derivatives and seleno analoges have been shown to possess significant antibacterial activity.[1] Several furan derivatives have a broad spectrum of insecticidal, acaricidal and nematocidal activities.[2] In conjunction with our previous work,[3–8] we report here on the synthesis of several derivatives of the above mentioned systems required for biological screening as well as for further chemical transformations.

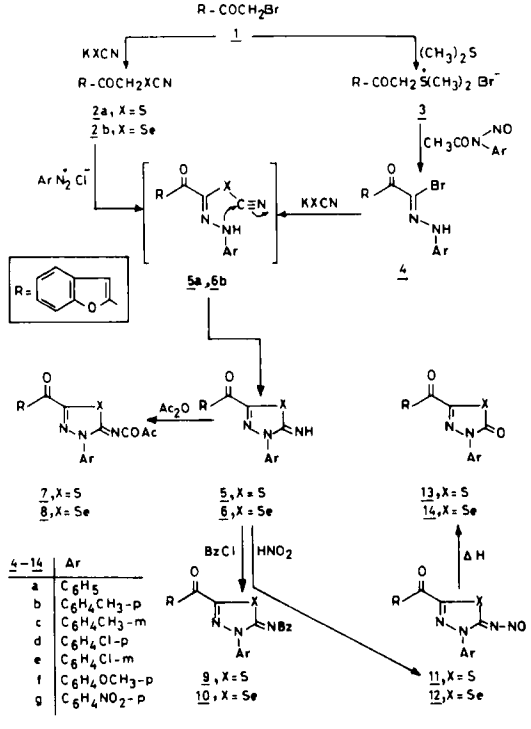
RESULTS AND DISCUSSION

2-(Thiocyanatoacetyl)benzofuran (**2a**) and 2-(selenocyanatoacetyl)-benzofuran (**2b**) coupled with aryldiazonium chlorides in a sodium acetate buffered solution of ethanol to give 2-imino-3-aryl-5-benzofuroylthiadiazolines **5** and 2-imino-3-aryl-5-benzofuroylselenodiazolines **6**, respectively. The structure of the latter were deduced from their spectra and their chemical reactions. IR spectra of **5** and **6** revealed no bands in the 2000–2200 cm^{-1} region due to a free SCN and SeCN groups. The spectra contained, however, bands at 3320 cm^{-1} (imino NH), 1650 cm^{-1} (CO) and at 1620 cm^{-1} (C=N). The absorption pattern of **5** and **6** in the UV region was, in each case, characterized by three maxima in the 380–360,

280–250 and 230–210 nm regions. As an example of the series, the prm spectrum of **5** (Ar = $C_6H_4CH_3$-p) in deuterated chloroform showed a multiplet at δ 7.0–8.5 (9H, ArH's and NH); a singlet at δ 6.3 (1H, furan H-3) and a singlet at δ 2.4 ($3H_3\underline{CH_3}$ Ar) ppm. Upon shaking with deuterium oxide a new singlet appeared at δ 4.50 ppm assignable to DOH proton and the multipelt at δ 7.0–8.5 ppm corresponds to eight protons only. The structure of **5** and **6** was further elucidated by independent synthesis. Thus, treatment of **4** with potassium thiocyanate and potassium selenocyanate in ethanol at room temperature produced products that proved to be identical in all respects with those of **5** and **6** prepared above. These results indicated that both the azo coupling of **2** and the reaction of **4** with potassium thiocyanate (potassium selenocyanate) proceed through one common intermediate, the hydrazones **5a** and **6a** which cyclised readily to give **5** and **6** respectively (Scheme 1).

Acylation of **5** and **6** with acetic anhydride (and with benzoyl chloride in pyridine) yielded the corresponding N-acetyl- **7**, **8** and N-benzoyl derivatives **9**, **10** respectively. Both elemental and spectral data confirmed the assigned structure of the products **7–10**. The pmr spectrum of **7** (Ar = $C_6H_4CH_3$-p) showed the presence of two singlets at δ 2.3 (3H, CH_3CON-) and δ 2.4 (3H, $\underline{CH_3}Ar$), δ 6.3 (s, 1H, furan H-3) and δ 7.0–8.5 (m, 8H, ArH's) ppm. IR spectra of **7** and **8** revealed bands at 1660 (CO) and 1630 ($CH_3CON=$) cm^{-1}.

Nitrosation of **5** and **6** gave the nitroso derivatives **11** and **12** respectively. The

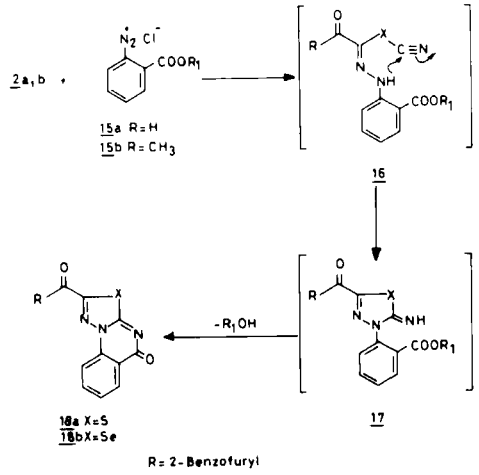


SCHEME 1

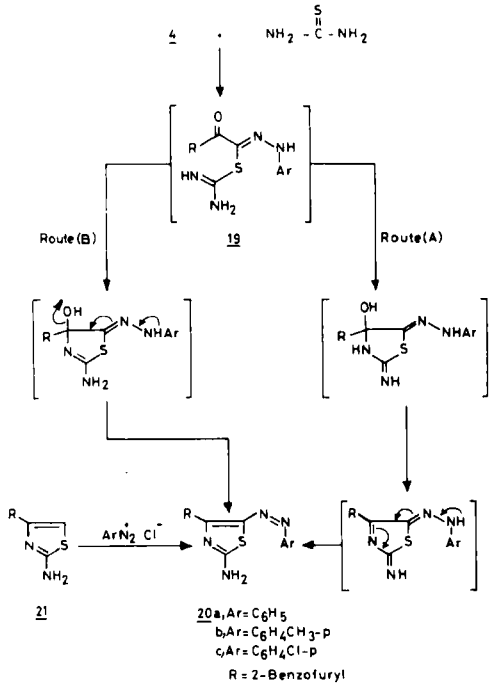
UV of the latter revealed two common maxima in the region of 510–470 nm ($\log \epsilon < 2$) and 365–340 nm ($\log \epsilon > 4$). These are assigned to the $n - \pi^*$ and $\pi - \pi^*$ transitions of the nitroso imino group. IR spectra of **11** and **12** showed no NH band.

All compounds **11** and **12** decomposed to the corresponding thiadiazolinones and selenodiazolinones **13** and **14**, respectively, upon refluxing in xylene. IR spectra of **13** and **14** showed two CO absorption bands near 1650 and 1705 cm^{-1}. The pmr spectra of **13** (Ar = $C_6H_4CH_3$-*p*) showed signals at δ 2.4 (s, 3H, CH_2Ar), δ 6.3 (s, 1H, furan H-3) and δ 7.0–8.5 (m, 8H, ArH's) ppm. Treatment of **2a, b** with diazotized anthranilic acid (**15a**) in ethanolic sodium acetate solution gave yellow coloured products. Mass spectra and analytical data indicated the molecular formulae as $C_{18}H_9N_3SO_2$ and $C_{18}H_9N_3SeO_2$, respectively. The IR spectra of these products were free of SCN (2165 cm^{-1}), NH (3340 cm^{-1}) and OH (3400–3100 cm^{-1}) bands but revealed two carbonyl bands near 1650 and 1705 cm^{-1}. Thus, it is clear that the hydrazone **16** was not the end product of the reaction. It was thought that **16** undergoes spontaneous cycloaddition[9,10] to give the iminothiadiazoline derivatives **17a** and iminoselenodiazoline derivatives **17b**, which completed the reaction by loss of water or methanol to yield the final products **18a** and **18b** respectively. The proposed structure of **18** was confirmed by our finding that **18** was also obtained by coupling of **2a, b** with diazotized methyl anthranilate (**15b**) (Scheme 2).

Treatment of **4** with an excess of thiourea in ethanol afforded products which were identified as 5-arylazo-4-benzofuroyl-2-aminothiazoles **20**. The structure of the latter was inferred from their spectral and elemental analyses. The pmr spectra of **20** showed, in each case, an NH_2 singlet at 5.9 ppm. The UV spectra of **20** exhibited two intense maxima ($\log \epsilon > 4$) in the 470–420 and 280–260 nm regions. The structure of **20** was further elucidated by coupling diazotized primary aromatic amines and 2-imino-4-benzofurylthiazole in ethanolic sodium acetate solution.



SCHEME 2



SCHEME 3

Scheme 3 shows two possible pathways that can account for the formation of **20** from **4** and thiourea. It is assumed that the first step involves formation of carbon-sulfur linkage by elimination of a molecule of hydrogen bromide to give **19**, by analogy to the reaction of thioamides with α-halocarbonyl compounds.[11] In the second step, ring closure occurs through the direct attack by either the imino-(pathway A) or the imino nitrogen atom (pathway B) on the carbonyl carbon, and a molecule of water is then eliminated.

EXPERIMENTAL

All melting points were determined on a Gallenkamp melting point apparatus and are uncorrected. IR spectra were recorded on a Pye-Unican SP 1000 spectrophotometer, PMR ($CDCl_3$ and $(CD_3)_2SO$) spectra on a Varian-T 60 A spectrometer, UV spectra (EtOH) on a Pye-Unicam SP 8000 and Cary 118 spectrophotometers, and mass spectra on a Perkin-Elmer RMU-6E spectrometer (Ionization energy 70 eV). Elemental analyses were performed by the Microanalytical Center at Cairo University, Giza, Egypt.

2-Bromoacetylbenzofuran[12] (**1**), 2-amino-4-benzofurylthiazole[13] (**23**), and sulfonium bromide[14] (**3**) were prepared according to previously described methods.

2-(Thiocyanatoacetyl)-benzofuran (**2a**) *and 2-(selenocyanatoacetyl)-benzofuran* (**2b**). To a suspension of 2-(bromoacetyl)benzofuran 1, (2.4 g, 0.005 mol) in ethanol (50 ml) a solution of the appropriate potassium thiocyante or potassium selenocyanate (0.006 mol) in water was added and the reaction mixture was refluxed for 30 min, cooled and diluted with water. The solid so formed was collected and recrystallised from ethanol to give **2a**, **b** respectively (cf. Table 1).

TABLE I

Characterization data of the newly synthesised products

Compound	M.P.°C	Method	Molecular formula	C% Calcd. (Found)	H% Calcd. (Found)	S% Calcd. (Found)
2a	152	—	$C_{11}H_7NO_2S$	60.82	3.24	14.75
				60.60	3.10	14.60
2b	126	—	$C_{11}H_7NO_2Se$	50.01	2.67	—
				49.80	2.50	—
4a	154–5	—	$C_{16}H_{11}BrN_2O_2$	55.99	3.23	—
				56.10	3.10	—
4b	158–60	—	$C_{17}H_{13}BrN_2O_2$	57.16	3.66	—
				57.00	3.50	—
4c	218–20	—	$C_{16}H_{10}BrClN_2O_2$	50.89	2.66	—
				51.00	2.50	—
5a	148–50	A, B	$C_{17}H_{11}N_3O_2S$	63.54	3.45	9.97
				63.60	3.50	10.10
5b	173–4	A, B	$C_{18}H_{13}N_3O_2S$	64.66	3.90	9.55
				64.50	3.70	9.70
5c	130–1	B	$C_{18}H_{13}N_3O_2S$	64.66	3.90	9.55
				64.80	3.80	9.80
5d	192	A, B	$C_{17}H_{10}N_3O_2SCl$	57.39	2.83	9.00
				57.50	2.90	9.20
5e	152–54	B	$C_{17}H_{10}N_3O_2SCl$	57.39	2.83	9.00
				57.20	2.70	9.10
5f	147–8	B	$C_{18}H_{13}N_3O_3S$	61.53	3.72	9.12
				61.40	3.90	9.30
5g	237–8	B	$C_{17}H_{10}N_4O_4S$	55.73	2.75	8.74
				55.90	2.90	8.90
6a	153–5	A, B	$C_{12}H_{11}N_3O_2Se$	55.44	3.01	—
				55.60	3.20	—
6b	185	A, B	$C_{18}H_{12}N_3O_2Se$	56.70	3.17	—
				56.60	3.30	—
6c	203–5	A, B	$C_{17}H_{10}ClN_3O_2Se$	50.70	2.50	—
				50.90	2.70	—
7a	227	—	$C_{19}H_{13}N_3O_3S$	62.80	3.60	8.81
				63.00	3.70	9.00
7b	217	—	$C_{20}H_{15}N_3O_3S$	63.65	4.00	8.49
				63.80	4.10	8.60
7c	183	—	$C_{20}H_{15}N_3O_3S$	63.65	4.00	8.49
				63.50	3.90	8.50
7d	206	—	$C_{19}H_{12}N_3O_3SCl$	57.36	3.04	8.05
				57.40	2.90	8.10
7e	210–12	—	$C_{19}H_{12}N_3O_3SCl$	57.36	3.04	8.05
				57.40	3.10	8.00
7f	190–1	—	$C_{20}H_{15}N_3O_4S$	61.06	3.84	8.14
				61.20	3.70	7.90
7g	243–4	—	$C_{19}H_{12}N_4O_5S$	55.88	2.96	7.84
				55.70	3.00	7.90
8a	230–1	—	$C_{19}H_{13}N_3O_3Se$	55.62	3.19	—
				55.70	3.00	—
8b	218–20	—	$C_{20}H_{15}N_3O_3Se$	56.61	3.56	—
				56.80	3.70	—
8c	211–13	—	$C_{19}H_{12}ClN_3O_3Se$	51.31	2.71	—
				51.40	2.80	—
9a	259–60	—	$C_{24}H_{15}N_3O_3S$	67.99	3.55	7.53
				68.00	3.70	7.40

TABLE I (Cont'd)

Compound	M.P.°C	Method	Molecular formula	C% Calcd. (Found)	H% Calcd. (Found)	S% Calcd. (Found)
9b	278–8	—	$C_{25}H_{17}N_3O_3S$	68.32 68.50	3.89 4.00	7.29 7.40
9c	243	—	$C_{25}H_{17}N_3O_3S$	68.32 68.20	3.89 3.70	7.29 7.40
9d	276–7	—	$C_{24}H_{14}ClN_3O_3S$	62.71 62.90	3.07 3.10	6.97 7.10
9e	217–18	—	$C_{24}H_{14}ClN_3O_3S$	62.71 62.90	3.07 2.90	6.97 7.10
9f	259–60	—	$C_{25}H_{17}N_3O_4S$	65.92 66.10	3.76 3.80	7.03 6.90
9g	253–5	—	$C_{24}H_{14}N_4O_5S$	61.30 61.40	3.00 2.90	6.81 6.70
10a	265–7	—	$C_{24}H_{15}N_3O_3Se$	61.02 60.90	3.20 3.30	— —
10b	243–5	—	$C_{25}H_{17}N_3O_3Se$	61.73 61.90	3.52 3.40	— —
10c	270–2	—	$C_{24}H_{14}ClN_3O_3Se$	56.87 57.00	2.78 2.90	— —
11a	121 dec.	—	$C_{17}H_{10}N_4O_3S$	58.28 58.40	2.87 2.70	9.14 9.00
11b	126 dec.	—	$C_{18}H_{12}N_4O_3S$	59.33 59.10	3.31 3.40	8.79 8.90
11c	116 dec.	—	$C_{18}H_{12}N_4O_3S$	59.33 59.30	3.31 3.10	8.79 8.60
11d	138 dec.	—	$C_{17}H_9ClN_4O_3S$	53.06 53.10	2.35 2.10	8.32 8.10
11e	125 dec.	—	$C_{17}H_9ClN_4O_3S$	53.06 53.20	2.35 2.40	8.32 8.50
11f	129–30 dec.	—	$C_{18}H_{12}N_4O_4S$	56.84 56.90	3.18 3.00	8.42 8.20
11g	135 dec.	—	$C_{17}H_9N_5O_5S$	51.64 51.80	2.29 2.10	8.10 8.00
12a	141–3 dec.	—	$C_{17}H_{10}N_4O_3Se$	51.39 51.20	2.53 2.30	— —
12b	139–40 dec.	—	$C_{18}H_{12}N_4O_3Se$	52.56 52.80	2.94 3.00	— —
12c	147–8 dec.	—	$C_{17}H_9ClN_4O_3Se$	47.29 47.40	2.10 2.20	— —
13a	174–6	—	$C_{17}H_{10}N_2O_3S$	63.34 63.50	3.12 3.00	9.94 10.10
13b	172–4	—	$C_{18}H_{12}N_2O_3S$	64.27 64.40	3.59 3.40	9.52 9.30
13c	183	—	$C_{18}H_{12}N_2O_3S$	64.27 64.30	3.59 3.70	9.52 9.60
13d	180–1	—	$C_{17}H_9ClN_2O_3S$	57.23 57.40	2.54 2.70	8.98 9.10
13f	166–2	—	$C_{18}H_{12}N_2O_4S$	61.35 61.50	3.43 3.20	9.09 8.90
13g	208–9	—	$C_{17}H_9N_3O_5S$	55.58 55.70	2.46 2.30	8.72 8.90
14a	190–2	—	$C_{17}H_{10}N_2O_3Se$	55.29 55.40	2.72 2.80	— —
14b	199–200	—	$C_{18}H_{12}N_2O_3Se$	56.40 56.50	3.15 3.00	— —
14c	158–60	—	$C_{17}H_9ClN_2O_3Se$	50.58 50.70	2.24 2.10	— —

TABLE I (Cont'd)

Compound	M.P.°C	Method	Molecular formula	C% Calcd. (Found)	H% Calcd. (Found)	S% Calcd. (Found)
18a	245–6	—	$C_{18}H_9N_3O_3S$	62.24	2.61	9.22
				62.40	2.50	9.40
18b	264–5	—	$C_{18}H_9N_3O_3Se$	54.83	2.30	—
				54.70	2.40	—
20a	260	A, B	$C_{17}H_{12}N_4OS$	63.74	3.77	10.00
				63.90	3.60	9.90
20b	295	A, B	$C_{18}H_{14}N_4OS$	64.65	4.22	9.58
				64.50	4.30	9.70
20c	286–8	A, B	$C_{17}H_{11}ClN_4OS$	57.54	3.12	9.03
				57.70	3.00	8.90

Hydrazidoyl bromides (**4a–c**). A suspension of the appropriate *N*-nitroso-substituted acetanilide (0.007 mol) in ethanol (100 ml) and sulfonium salt (15 g, 0.005 mol) was stirred for 1 h at room temperature and then left overnight. Upon dilution of the reaction mixture with water, a yellow solid precipitated which was collected and crystallized from ethanol or acetic acid to give **4a–c** (cf. Table I).

3-Aryl-5-(2'-benzofuryl)-2-imino-Δ^2-1,3,4-thiadiazolines **5a–c** *and 3-aryl-5-(2'-benzofuroyl)-2-imino-Δ^2-1,3,4-selenodiazolines* **6a–c**. Method (A): To a cold solution of the appropriate 2 (0.01 mol) and sodium acetate (1.3 g) in ethanol (50 ml) was added dropwise a solution of the appropriate diazotized primary aromatic amines (0.01 mol) with stirring. After the addition was compelted (30 min), the reaction mixture was left overnight in a refrigerator. The yellow solid that precipitated was collected and crystallized from ethanol (except **5g** from dioxane) to give **5a–c** and **6a–c** respectively (cf. Table I).

Method (B): To a suspension of the appropriate **4a–c** (0.005 mol) in ethanol (30 ml) a solution of potassium thiocyanate (1.06 g, 0.01 mol) or potassium selenocyanate (1.4 g, 0.01 mol) in water (10 ml) was added. The reaction mixture was stirred for 4 h at room temperature. The crude product was collected, washed with water and crystallized from ethanol. The product obtained was found to be identical in all respects (m.p., mixed m.p. and spectra) with that obtained above by coupling of **2** with diazotized primary aromatic amines.

Acylation of **5** *and* **6**. Compound **5** or **6** (0.5 g) was stirred in acetic anhydride (10 ml) for 5 min. The reaction mixture was left for 3 h at room temperature. The solid so formed was collected and crystallized from acetic acid. The *N*-acetyl derivatives **7** and **8** were obtained in almost quantitative yields (cf. Table I).

A mixture of equimolecular (0.01 mol) amount of **5** or **6** and benzoyl chloride in pyridine (10 ml) was allowed to react at room temperature for 15 min. The reaction mixture was poured onto ice cold water and acidified (HCl) and the solid product was filtered off and fractionally crystallized from hot H_2O. The insoluble portion was then filtered off and crystallised from DMF to afford **9** and **10** respectively in almost quantitative yields (cf. Table I).

Nitrosation of **5** *and* **6**. A solution of **5** or **6** (1.0 g) in acetic acid (30 ml) was treated with a saturated solution of sodium nitrite with stirring. The reddish-orange products which precipitated were collected and crystallized from ethanol to give **11** and **12** respectively (cf. Table I).

2-(2'-Benzofuroyl)-4-aryl-Δ^2-1,3,4-thiadiazolin-5-ones (**13**) *and 2-(2'-benzofuroyl)-4-aryl-Δ^2-1,3,4-selenodiazolin-5-ones* (**14**). A solution of the appropriate nitrosoimino derivatives **11** and **12** (0.5 g) were refluxed in xylene (20 ml) for 30 minutes. The solvent was then removed under reduced pressure. Trituration with petroleum ether (40/60) caused precipitation of **13** or **14** respectively which were collected and crystallized from ethanol (cf. Table I).

Thiadiazolo[2,3-b]quinozoline **18a** *and selenodiazolo[2,3-b]quinozoline* **18b**. To a cold solution of the appropriate **2** (0.01 mol) and sodium acetate (1.3 g) in ethanol (50 ml) was added dropwise a solution of diazotized anthranilic acid or methyl anthranilate (0.01 mol) with stirring. After the addition was completed (30 min.) the reaction mixture was stirred for additional 3 h at 0–5°C. The

pale yellow solids which precipitated were collected and crystallized from acetic acid to give **18a, b** (cf. Table I).

2-Amino-5-arylazo-4-(2'-benzofuroyl)thiazoles **20a–c.**
Method (A): A mixture of the appropriate hydrazidoyl bromide **4** (0.005 mol) and thiourea (0.76 g, 0.01 mol) in ethanol (30 ml) was refluxed for 3 h, then poured onto ice, and two drops of ammonium hydroxide were added. The solid, so formed, was collected, washed with water and crystallized from dimethyl formamide to give **20a–c** respectively (Table I).
Method (B): A diazonium salt solution, prepared from 0.01 mol of the appropriate primary aromatic amines, was added at 0–5°C to 2-amino-4-benzofuroyl thiazole[13] (**21**, 0.01 mol) in ethanol (50 ml) containing sodium acetate (1.3 g). The obtained solid products were proved to be identical in all aspects (m.p., mixed m.p. and spectra) with **22a–c** prepared by method A above.

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