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SYNTHESIS OF 2,5-DISUBSTITUTED 1,3,4-OXADIAZOLES CONTAINING BENZOTHIAZOLYLTHIOL GROUPING

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A series of 2,5-substituted 1,3,4-oxadiazoles containing 2-benzothiazolylthiomethyl grouping has been synthesized by condensing derivatives of (2-benzothiazolylthio)acetic acid with imino ester hydrochlorides and hydrazides of carboxylic acids, by the cyclodehydration of N-acyl-N'-(2-benzothiazolylthioacetyl)hydrazines under the action of $POCl_3$, and also by the reaction of 2-mercaptobenzothiazole with 2-chloromethyl-1,3,4-oxadiazoles in the presence of sodium methylate.

Keywords: benzothiazole, carboxylic acid hydrazides, carboxylic acid imino ester hydrochlorides, 2-mercaptobenzothiazole, 1,3,4-oxadiazole, condensation.

N-Substituted 2-amino- and 2-alkylthio-1,3,4-oxadiazoles containing benzothiazole fragments in position 5 possess a wide spectrum of biological activity, including anti-inflammatory [1,2], antimicrobial [3], antibacterial [2], and hypotensive [4] activity. There is extremely limited information in the literature [5,6] regarding 2-alkyl(aryl)-5-substituted 1,3,4-oxadiazoles containing benzothiazole fragments.

In continuation of our investigations on the synthesis of heteryl-substituted 1,3,4-oxadiazoles, we give data in the present study on the preparation of 2,5-disubstituted 1,3,4-oxadiazoles containing 2-benzothiazolylthiomethyl grouping. Compounds of this type may be of interest as potentially biologically active substances and also as stabilizers and additives for polymeric material, hydrocarbon fuel, and lubricating oil [10].

Hydrochlorides of carboxylic acid imino esters may serve as convenient synthons in the synthesis of 1,3,4-oxadiazoles [7,10,11]. In the present work the methyl imino ester hydrochlorides of the following acids were used as starting materials: butyric (**1a**), substituted acetic (**1b-f**), benzoic (**1g**), 4-nitro- (**1h**), and 4-hydroxy-3,5-di(*tert*-butyl)benzoic (**1i**), β-[4-hydroxy-3,5-di(*tert*-butyl)phenyl]propionic (**1j**), 5-nitro-2-furancarboxylic (**1k**), and 3-indolecarboxylic (**1l**) acids. 2-alkyl(aryl,heteryl)-5-(2-benzothiazolylthiomethyl)-1,3,4-oxadiazoles (**3a-m**) were formed as a result of the condensation of the imino ester hydrochlorides **1a-l** with (2-benzothiazolylthiomethyl)acetic acid hydrazide (**2a**) (method A).

The best yields of compounds **3a-l** (Table 1) were achieved on boiling the reactants in ethanol or dioxane at a molar ratio of **1** : **2a** of 1.2 : 1. The duration of the process depends on the reactivity of the initial imino ester hydrochloride **1a-l**. For example, the formation of compounds **3a-h,j,k** is complete after boiling the reactants for 4-5 h in ethanol. When obtaining 1,3,4-oxadiazoles **3i,l** from imino ester hydrochlorides **1i,l**, which have reduced reactivity due to the effect of bonding of the electron-donating hydroxyaryl or indole substituent with the imino-ester group [11], it was necessary to boil in dioxane for 10-12 h.

We also used the condensation of (2-benzothiazolylthio)acetic acid imino ester dihydrochloride (**1f**) with hydrazides of various carboxylic acids (**2b-f**) in the synthesis of 1,3,4-oxadiazoles **3g,h,j-l** (method B). The reaction was carried out by boiling the reactants (molar ratio of **1f** : **2** was 1.25 : 1) in ethanol for several hours. The corresponding products **3g,h,j-l** were formed in 65-76% yields. In addition, compounds **3g,h** and 2-methyl-

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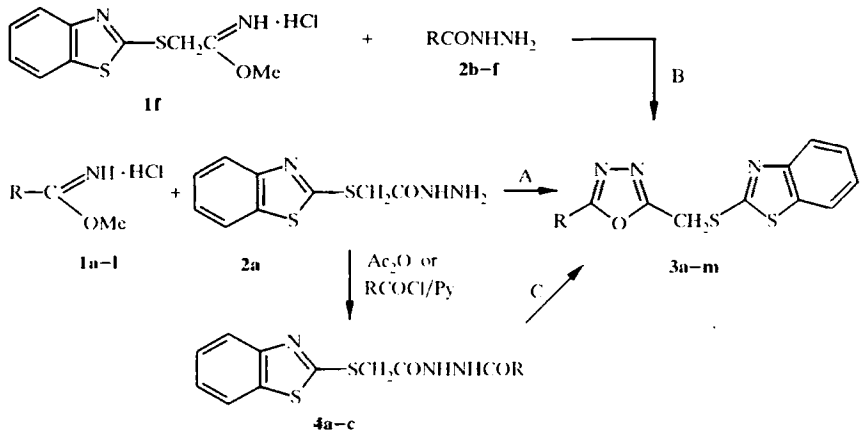
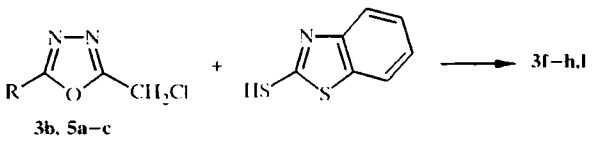


1a, **3a** R = Pr; **1b**, **3b** R = $ClCH_2$; **1c**, **3c** R = $EtOOCCH_2$; **1d**, **3d** R = $PhCH_2$; **1e**, **3e** R = 4-$NO_2C_6H_4CH_2$; **1f**, **3f** R = 2-benzothiazolylthiomethyl; **1g**, **2b**, **3g**, **4b** R = Ph; **1h**, **2b**, **3h**, **4c** R = 4-$NO_2C_6H_4$; **1i**, **3i** R = 4-HO-3,5-(*t*-Bu)$_2C_6H_2$; **1j**, **2d**, **3j** R = 4-HO-3,5-(*t*-Bu)$_2C_6H_2CH_2CH_2$; **1k**, **2e**, **3k** R = 5-nitrofur-2-yl; **1l**, **2f**, **3l** R = 3-indolyl; **3m**, **4a** R = Me

1,3,4-oxadiazole **3i** were obtained by cyclodehydration of the corresponding N-acyl-N'-(2-benzothiazolylthioacetyl)hydrazines (**4a-c**) under the action of phosphorus oxychloride (method C) [7,9,12]. The latter were obtained by acylation of hydrazide **2a** with acetic anhydride in an inert solvent at room temperature or with the acid chlorides in pyridine in 78-85% yield. However even brief heating with phosphorus oxychloride leads to strong resinification of the reaction mixture from which the desired products **3g,h,i** were isolated in 38-47% yield. On boiling N,N'-diacylhydrazines **4a-c** in an excess of acetic anhydride for several hours no cyclodehydration to the corresponding 1,3,4-oxadiazoles occurred.

2-Arylthiomethyl-5-R-1,3,4-oxadiazoles may be obtained by the interaction of 2-chloromethyl-5-R-1,3,4-oxadiazoles with arylthiols [13]. We have used this method in the present work for the synthesis of compounds **3f-h,l**. As a result of the condensation of 2-chloromethyl-5-R-1,3,4-oxadiazoles **3b**, **5a-c** with 2-mercapto-benzothiazole in the presence of equimolar quantity of sodium methylate at 0-10°C (0.5 h), the corresponding 1,3,4-oxadiazoles **3f-h,l** were formed in 78-87% yield (method D).



5a R = Ph, **5b** R = 4-$NO_2C_6H_4$, **5c** R = 3-indolyl

The characteristics of the disubstituted 1,3,4-oxadiazoles **3a-m** synthesized are given in Table 1. The composition and structure of these compounds were confirmed by data of elemental analysis, IR and ^{1}H NMR spectroscopy. In the IR spectra intense absorption maxima were observed in the ranges of 1600-1615, 1570-1585, and 1460-1490 cm^{-1} characteristic of the stretching vibrations of the oxadiazole ring [14,15]. The presence of the latter was confirmed by occurrence of absorption bands at 1225-1250 and 1020-1050 cm^{-1} assigned to the stretching vibrations of the =C–O–C= fragment in 1,3,4-oxadiazoles [16], and of an absorption maximum near 950 cm^{-1} related to the breathing vibrations of the oxadiazole ring [14,15]. Absorption due to the benzothiazole fragment [14] was also observed for all the compounds considered at 1520-1530, 1430-1445 (stretching vibrations of the ring), 1060-1085, and 735-740 cm^{-1}.

In the ^{1}H NMR spectra of the synthesized compounds the signals of the thiomethyl group protons were displayed as singlets in the range of 3.94-4.27 ppm. The multiplet signals at 7.14-8.14 ppm correspond to the protons of the benzothiazole fragments.

TABLE 1. Characteristics of the Compounds Synthesized

Compound	Empirical formula	Found, % / Calculated, %				mp, °C*	R_f (solvent system)	^{1}H NMR spectrum, δ, ppm**	Yield, % (method)
		C	H	N	S				
1	2	3	4	5	6	7	8	9	10
3a	$C_{13}H_{14}N_4OS_2$	53.47 / 53.61	4.56 / 4.47	14.87 / 14.73	21.81 / 21.99	134–136	0.57 (a)	1.10 (3H, s, CH_3); 1.33–1.44 (2H, m, CH_2); 2.24 (2H, t, $CH_2CH_2CH_3$); 3.94 (2H, s, CH_2S); 7.38–7.97 (4H, m, H_{Ar})	78 (A)
3b	$C_{11}H_9ClN_4OS_2$	44.51 / 44.37	2.77 / 2.69	14.01 / 14.12	21.35 / 21.51	110–113	0.74 (b)	2.87 (2H, s, CH_2Cl); 4.12 (2H, s, CH_2S); 7.52–8.04 (4H, m, H_{Ar})	72 (A)
3c	$C_{14}H_{14}N_4O_3S_2$	50.03 / 50.15	3.97 / 3.88	12.66 / 12.54	18.91 / 19.10	Oil	0.43 (a)	1.37 (3H, t, J = 6 Hz, CH_3); 3.18 (2H, s, CH_2); 3.84 (2H, q, J = 6 Hz, CH_2O); 4.22 (2H, s, CH_2S); 7.14–7.55 (4H, m, H_{Ar})	67 (A)
3d	$C_{17}H_{14}N_4OS_2$	60.06 / 60.18	3.94 / 3.83	12.56 / 12.39	18.72 / 18.88	82–84	0.45 (c)	3.34 (2H, s, $PhCH_2$); 4.07 (2H, s, CH_2S); 6.85–7.30 (9H, m, H_{Ar})	84 (A)
3e	$C_{17}H_{12}N_4O_3S_2$	53.25 / 53.12	3.04 / 3.12	14.71 / 14.58	16.49 / 16.67	195–197	0.52 (b)	3.50 (2H, s, $ArCH_2$); 4.21 (2H, s, CH_2S); 7.04–7.59 (8H, m, H_{Ar})	77 (A)
3f	$C_{16}H_{12}N_4OS_3$	50.32 / 50.47	2.92 / 2.80	12.84 / 13.08	30.14 / 29.91	176–178	0.62 (c)	4.24 (4H, s, CH_2S); 7.59–8.08 (8H, m, H_{Ar})	81 (A) 78 (D)
3g	$C_{18}H_{14}N_4OS_2$	58.92 / 59.08	3.47 / 3.38	13.11 / 12.92	19.47 / 19.69	95–97	0.62 (b)	4.12 (2H, s, CH_2S); 6.92–7.37 (9H, m, H_{Ar})	87 (A), 76 (B), 42 (C), 83 (D)

TABLE 1 (continued)

1	2	3	4	5	6	7	8	9	10
3h	$C_{18}H_{10}N_4O_3S_2$	52.04 51.89	2.83 2.70	15.01 15.13	17.14 17.30	157-158.5	0.53 (a)	4.20 (2H, s, CH_2S); 7.05-7.48 (8H, m, H_{Ar})	83 (A), 72 (B), 47 (C), 87 (D)
3i	$C_{24}H_{22}N_4O_2S_2$	63.69 63.58	6.11 5.96	9.05 9.27	14.25 14.13	145-146	0.75 (a)	1.52 (18H, br. s, *t*-Bu); 3.94 (2H, s, CH_2S); 5.18 (1H, s, OH); 7.12 (2H, s, 2-, 6-H_{Ar}); 7.55-8.14 (4H, m, H_{Ar})	64 (A)
3j	$C_{26}H_{31}N_3O_2S_2$	65.07 64.86	6.30 6.44	8.95 8.73	13.16 13.30	Oil (n_D^{20} 1.5818)	0.66 (c)	1.60 (18H, br. s, *t*-Bu); 3.72-4.04 (4H, m, CH_2CH_2); 4.27 (2H, s, CH_2S); 5.26 (1H, s, OH); 6.84 (2H, s, 2-,6-H_{Ar}); 7.28-7.62 (4H, m, H_{Ar})	73 (A), 70 (B)
3k	$C_{14}H_8N_4O_4S_2$	46.53 46.67	2.10 2.22	15.77 15.55	17.61 17.78	204-206	0.43 (a)	4.02 (H, s, CH_2S); 6.58 (1H, d, J = 3.7 Hz, 3-H furan); 6.92 (1H, d, 4-H furan); 7.38-7.84 (4H, m, H_{Ar})	82 (A), 74 (B)
3l	$C_{18}H_{12}N_4OS_2$	59.21 59.34	3.38 3.30	15.61 15.38	17.33 17.58	195-196	0.53 (b)	4.15 (2H, s, CH_2S); 7.22-7.78 (9H, m, H_{Ar}); 8.14 (1H, br. s, NH)	76(A), 65 (B), 80 (D)
3m	$C_{11}H_8N_4OS_2$	49.96 50.19	3.50 3.42	16.15 15.97	24.47 24.33	79-81	0.58 (c)	2.64 (3H, s, CH_3); 4.04 (2H, s, CH_2S); 7.48-7.96 (4H, m, H_{Ar})	38 (C)

* Compounds were recrystallized as follows: **3a,m** from toluene–petroleum ether, 1 : 3; **3b-f,h,l** from dioxane–water, 1 : 1.5; **3g** from acetone–water, 1 : 2.5; **3i** from heptane–dioxane, 3 : 1; **3k** from aqueous DMF.

*[2] The spectra of compounds **3a,d-i,k,l** were recorded in DMSO-d_6 and those of compounds **3b,c,j** in $CDCl_3$.

TABLE 2. The IR Spectral Characteristics of the Synthesized Compounds **3a,b** and **4**, ν, cm^{-1}

Compound	Solvent	C=O	C=C	$O=\overset{+}{C}-N$ (C=N)	C=O ester	NH
3a	Nujol	1770	1655	1580, 1550	1740	3270-2600
	CH_2Cl_2	1795	1665	1590	1747	3290-2650
3b	Nujol	1788	1660	1590, 1551	1730	3290-2610
	CH_2Cl_2	1780	1657	1575	1742	3300-2670
4	Nujol	1740	1650	(1575)	1740	3320-2610
	$CHCl_3$	1745	1640	(1590)	1740	3260-2860

EXPERIMENTAL

The IR spectra were taken on a Bruker IFS 48 instrument in KBr disks, nujol suspensions, or in thin films. The ^1H NMR spectra were recorded on a Bruker WP 250 spectrometer, internal standard was TMS. A check on the progress of reactions and the purity of the compounds obtained was made with the aid of TLC on Al_2O_3 of Brockmann activity grade III in the solvent systems a) benzene–ethanol, 20 : 1; b) benzene–ethanol, 10 : 1; and c) CCl_4–acetone, 20 : 1. Visualization was with iodine vapor.

The initial methyl imino ester hydrochlorides of butyric (**1a**) [18], phenylacetic (**1d**) [19], 4-nitrophenylacetic (**1e**) [19], benzoic (**1g**) [17], 4-nitrobenzoic (**1h**) [19], 4-hydroxy-3,5-di(*tert*-butyl)benzoic (**1i**) [20], β-[4-hydroxy-3,5-di(*tert*-butyl)phenyl]propionic (**1j**) [20], 5-nitro-2-furancarboxylic (**1k**) [21], and 3-indolecarboxylic (**1l**) [22] acids, hydrazides of (2-benzothiazolylthio)acetic (**2a**) [2], β-[4-hydroxy-3,5-di(*tert*-butyl)phenyl]propionic (**2d**) [23], and 3-indolecarboxylic (**2f**) [7] acids, and also 2-chloromethyl-1,3,4-oxadiazoles **5a** [24], **5b** [24], and **5c** [25] were obtained by known methods (citations are given above for each compound).

(2-Benzothiazolylthio)acetic Acid Methyl Imino Ester Dihydrochloride (1f). A stream of dry gaseous HCl was passed for 2.5 h into stirred solution of (2-benzothiazolylthio)acetonitrile (8.24 g, 0.04 mol) and absolute methanol (3.2 g, 0.1 mol) in anhydrous 1,2-dimethoxyethane (150 ml) at 0-5°C. The reaction mixture was maintained at 20°C for 24 h, anhydrous ether (150 ml) was poured in, and the mixture cooled to -5°C. The precipitated solid was filtered off, washed on the filter with anhydrous ether to neutral reaction, and dried in vacuum over KOH. Imino ester dihydrochloride **1f** (9.44 g, 86%) was obtained; mp 173-175°C (decomp.). IR spectrum, cm^{-1}: 740, 3120, 1675, 1525, 1420, 1030. Found, %: Cl 23.04; N 3.91. $C_{10}H_{10}N_2OS_2 \cdot 2HCl$. Calculated, %: Cl 22.82; N 9.00.

2,5-Disubstituted 1,3,4-Oxadiazoles (3a-l). A. Mixture of imino ester hydrochloride **1a-l** (12.0 mmol) and hydrazide **2a** (2.39 g, 10.0 mmol) in absolute ethanol (45 ml) was boiled with stirring for 5 h (when obtaining compounds **3i,l** the mixture was boiled for 12 h in 45 ml of anhydrous dioxane). The reaction mixture was evaporated to dryness at reduced pressure. In the case of compounds **1a,e-i,k,l** the residue was crystallized from a suitable solvent (see Table 2), and on making compounds **3b-d,j** it was chromatographed on column with Al_2O_3 (h = 80 cm, d = 4.5 cm) eluting with mixture of benzene–methanol, 15 : 1. After removing the solvent, 1,3,4-oxadiazoles **3c,j** were obtained as viscous dark yellow uncrystallizable oils.

5-(2-Benzothiazolylthiomethyl)-2-phenyl-1,3,4-oxadiazole (3g). B. Mixture of imino ester dihydrochloride **1f** (3.87 g, 12.5 mmol) and hydrazide **2b** (1.48 g, 10.0 mmol) in absolute ethanol (40 ml) was boiled with stirring for 4-5 h until the initial hydrazide **2b** had disappeared from the reaction mixture (check by TLC). The solvent was removed at reduced pressure, the residue crystallized from mixture of acetone–water, 1 : 1.25, and 1,3,4-oxadiazole **3g** was obtained.

1,3,4-Oxadiazoles **3h,j-l** were synthesized similarly from imino ester **1f** and hydrazides **2c-f** respectively.

N-Acetyl-N'-(2-benzothiazolylthioacetyl)hydrazine (4a). Acetic anhydride (3.6 g, 35 mmol) was added during 0.5 h to stirred mixture of hydrazide **2a** (7.17 g, 30.0 mmol) and 2-propanol (70 ml) at 20°C. The reaction mixture was stirred at 20°C for 2 h then ice water (50 ml) was added dropwise. The precipitated solid was filtered off, dried, and crystallized from ethanol. Hydrazine **4a** (7.16 g, 85%) was obtained; mp 168-170°C, R_f 0.67

(system b). IR spectrum: 3200-3320, 3050, 2910, 1665, 1640, 1530, 1440, 1260, 1240, 1150, 890, 825, 735 cm^{-1}. ^1H NMR spectrum (DMSO-d_6): 2.74 (3H, s, Me); 4.18 (2H, s, CH_2S); 7.37-7.84 (4H, m, H_{Ar}); 8.18 ppm (2H, br. s, NH). Found, %: C 47.14; H 3.82; N 15.19; S 22.54. $C_{11}H_{11}N_3O_2S_2$. Calculated, %: C 46.97; H 3.91; N 14.95; S 22.78.

N-Benzoyl-N'-(2-benzothiazolylthioacetyl)hydrazine (4b). Benzoyl chloride (2.81 g, 20.0 mmol) was added in portions to stirred solution of hydrazide **2a** (4.78 g, 20.0 mmol) in anhydrous pyridine (50 ml). The reaction mixture was boiled with stirring for 2 h, cooled to 20°C, and poured into ice water (200 ml). The precipitated solid was filtered off, washed on the filter with water, dried, and crystallized from 1-butanol. Hydrazine **4b** (5.35 g, 78%) was obtained; mp 154-156°C, R_f 0.43 (system b). ^1H NMR spectrum (DMSO-d_6): 4.04 (2H, s, CH_2); 6.74-7.22 (9H, m, H_{Ar}); 8.02 ppm (2H, br. s, NH). Found, %: C 56.16; H 3.63; N 12.02; S 18.85. $C_{16}H_{13}N_3O_2S_2$. Calculated, %: C 55.98; H 3.79; N 12.24; S 18.66.

N-(4-Nitrobenzoyl)-N'-(2-benzothiazolylthioacetyl)-hydrazine (4b) was obtained analogously from 4-nitrobenzoyl chloride. Yield 81%; mp 166-168°C (dioxane–water, 1 : 1); R_f 0.62 (system b). Found, %: C 49.35; H 2.90; N 14.61; S 16.62. $C_{16}H_{12}N_4O_4S_2$. Calculated, %: C 49.48; H 3.09; N 14.43; S 16.49.

5-(2-Benzothiazolylthiomethyl)-2-methyl-1,3,4-oxadiazole (3m). C. Mixture of hydrazine **4a** (4.20 g, 15.0 mmol) and $POCl_3$ (30 ml) was stirred at 80-85°C for 0.5 h. The reaction mixture was cooled to 0°C, poured onto ice (200 g), and the mixture neutralized to pH 7.5 with aqueous ammonia solution. The dark oil which precipitated was extracted with dichloromethane (3 × 20 ml), the extract washed with water, dried over Na_2SO_4, and evaporated to dryness under reduced pressure. The residue was crystallized from toluene–petroleum ether, 1 : 3, and 1,3,4-oxadiazole **3m** was obtained.

1,3,4-Oxadiazoles **3g,h** were synthesized analogously from N,N'-diacylhydrazines **4b,c** respectively.

5-(2-Benzothiazolylthiomethyl)-2-(4-nitrophenyl)-1,3,4-oxadiazole (3h). D. 2-Mercaptobenzothiazole (2.0 g, 12.0 mmol) was added in portions to stirred solution of sodium methylate obtained from sodium (0.2 g, 14.0 g-atom) in absolute methanol (65 ml). The reaction mixture was stirred at 20°C for 0.5 h, cooled to 0°C, and 2-chloromethyl-1,3,4-oxadiazole **5b** (2.87 g, 12.0 mmol) was added in portions. The reaction mixture was then stirred at 0-10°C for 0.5 h, left for 1 h at 20°C, and evaporated to dryness at reduced pressure. The residue was crystallized from dioxane–water, 1 : 1.5, and 1,3,4-oxadiazole **3h** was obtained.

1,3,4-Oxadiazoles **3f,g,l** were synthesized analogously from 2-chloromethyl-1,3,4-oxadiazoles **3b,5a,c** respectively.

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