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Dedicated to Full Member of the Russian Academy of Sciences B.A. Trofimov on his 70th anniversary

1,3-Dipolar Cycloaddition of 3-Phenylamino-5-phenylimino-1,2,4-dithiazole to 1-Acyl-2-phenylacetylenes—A New Route to Functionalized 1,3-Thiazole Derivatives

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Abstract—3-Phenylamino-5-phenylimino-1,2,4-dithiazole reacted with 1-acyl-2-phenylacetylenes in ethanol or toluene on heating (78–80°C, 1 h) in chemo- and regioselective fashion to give previously unknown *N*-[5-acyl-3,4-diphenyl-1,3-thiazol-2(3*H*)-ylidene]-*N*′-phenylthioureas (yield 57–60%). The structure of *N*-[5-benzoyl-3,4-diphenyl-1,3-thiazol-2(3*H*)-ylidene]-*N*′-phenylthiourea was proved by X-ray analysis.

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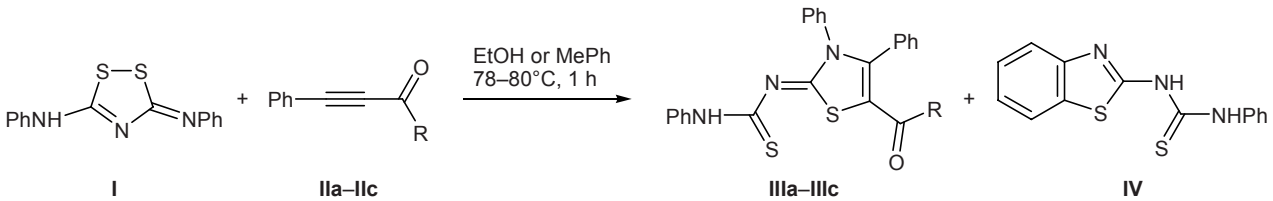
Thiazoles constitute one of the most important classes of heterocyclic compounds due to their role in many biological processes. Thiazole ring is a structural fragment of natural compounds (where it is formed from cysteine fragments of peptides) such as thiamine (vitamin B_1), carboxylase, and penicillin and some medical agents [1]. Many synthetic thiazole derivatives exhibit diverse biological activity, in particular antibacterial, fungicidal, spasmolytic, and antiinflammatory [2–10]. 2-Amino-substituted thiazoles (e.g., thiazolylthioureas) were found to be active against tumor cells [11] and human immunodeficiency virus [12–14]. Therefore, search for new functionalized thiazole derivatives and development of convenient methods for their preparation remain important problems.

The most commonly used procedure for building up 1,3-thiazole ring is based on the Hantzsch reaction of α-halo ketones with thioamides and related compounds [2, 4, 15–19]; thiazole derivatives were also obtained by reactions of thioureas with acylbromoacetylenes [20] and of thioamides with acetylenedicarboxylic acid esters [21, 22].

The goal of the present study was to develop a new synthetic approach to functionally substituted 1,3-thiazoles having an acyl group in the 5-position and a thiourea fragment in the 2-position via reaction of 1-acyl-

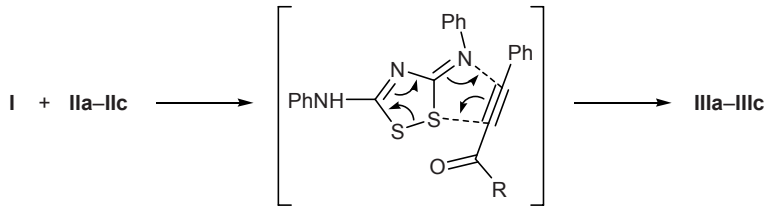
Scheme 1.



I **IIa–IIc** **IIIa–IIIc** **IV**

R = Me (**a**), Ph (**b**), 2-thienyl (**c**).

Scheme 2.

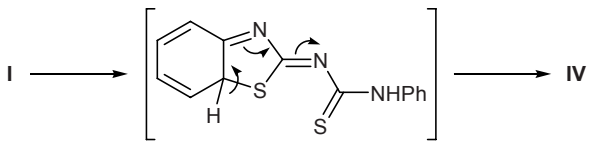


A

2-phenylacetylenes with 3-phenylamino-5-phenylimino-1,2,4-dithiazole. The latter is a representative of amino-substituted 1,2,4-dithiazoles that are obtained by oxidation of 2,4-dithiobiuret derivatives which are readily synthesized from accessible thiourea of substituted thioureas and isothiocyanates [23–26]. It is known that diamino-1,2,4-dithiazoles readily react with dipolarophiles (such as dimethyl acetylenedicarboxylate, nitriles, isothiocyanates, carbon disulfide) to give the corresponding *N*-(1,3-thiazol-2-ylidene-, 1,3,5-thiadiazol-2-ylidene, or 1,2,4-thiadiazol-5-ylidene)thioureas [27–29]. Analogous reactions with dipolarophilic 1-acyl-2-phenylacetylenes were not reported.

We found that 3-phenylamino-5-phenylimino-1,2,4-dithiazole (**I**) reacts with 1-acetyl-, 1-benzoyl-, and 1-(2-thenoyl)-2-phenylacetylenes **IIa**–**IIc** in toluene or ethanol at 78–80°C (reaction time 1 h) to give 57–60% of the corresponding *N*-[5-acyl-3,4-diphenyl-1,3-thiazol-2(3*H*)-ylidene]-*N*′-phenylthioureas **IIIa**–**IIIc** and 18–21% of *N*-(1,3-benzothiazol-2-yl)-*N*′-phenylthiourea (**IV**) (Scheme 1). Presumably, compounds **IIIa**–**IIIc** are formed as a result of concerted process through transition state **A** with circular delocalization of electrons; electron density transfer along the closed path is accompanied by closure of 1,3-thiazole ring, while cleavage of the S–S bond gives rise to thiourea fragment in position *2* of the newly formed thiazole ring (Scheme 2). The concerted mechanism of the process is supported by the fact that the yield of the target product remains almost the same upon variation of the solvent polarity (ε = 24.3 and 2.4 for ethanol and toluene, respectively), other conditions (reaction time, temperature, reactant concentration) being equal.

Scheme 3.



B

N-(1,3-Benzothiazol-2-yl)-*N*′-phenylthiourea (**IV**) is likely to be formed via intramolecular rearrangement of initial 1,2,4-dithiazole **I** through intermediate **B** (Scheme 3); this rearrangement is favored by thermal instability of compound **I**. Analogous thermal or acid-catalyzed rearrangements were described in [30, 31].

The molecular and crystalline structures of compound **IIIb** were determined by X-ray analysis of a single crystal which was obtained by isothermal evaporation of a solution of 0.1 g of **IIIb** in 40 ml of DMSO at 20°C; the IR and ^{1}H and ^{13}C NMR spectra confirmed the structure of thiazoles **IIIa**–**IIIc**.

The (*i*) fragment in the molecule of *N*-[5-benzoyl-3,4-diphenyl-1,3-thiazol-2(3*H*)-ylidene]-*N*′-phenyl-

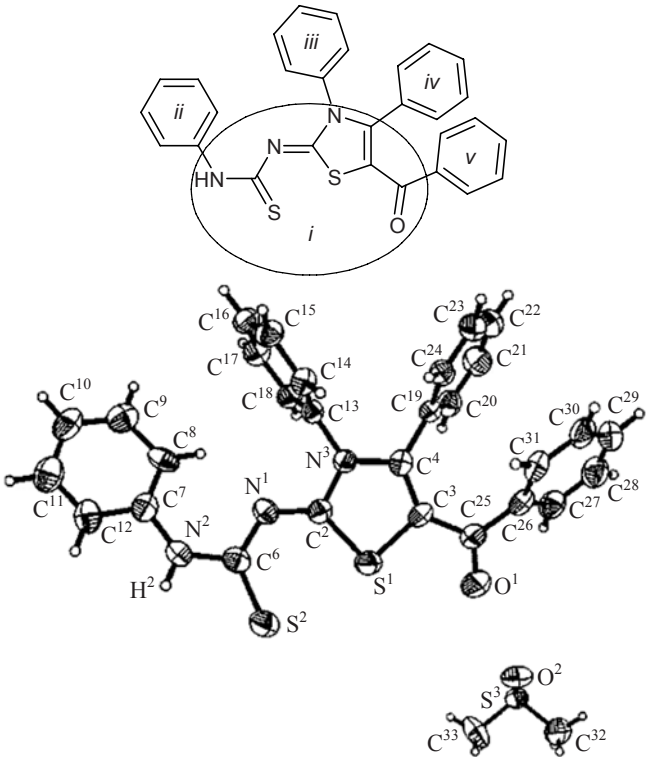


Fig. 1. Structure of the molecule of *N*-[5-benzoyl-3,4-diphenyl-1,3-thiazol-2(3*H*)-ylidene]-*N*′-phenylthiourea (**IIIb**) and solvate DMSO molecule according to the X-ray diffraction data.

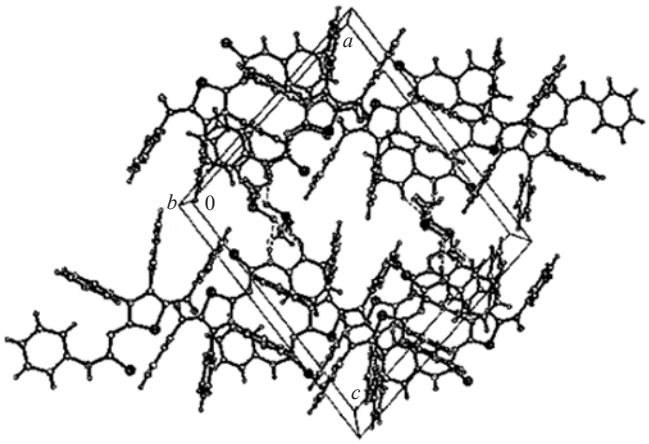


Fig. 2. Crystalline structure of *N*-[5-benzoyl-3,4-diphenyl-1,3-thiazol-2(3*H*)-ylidene]-*N*′-phenylthiourea (**IIIb**) according to the X-ray diffraction data.

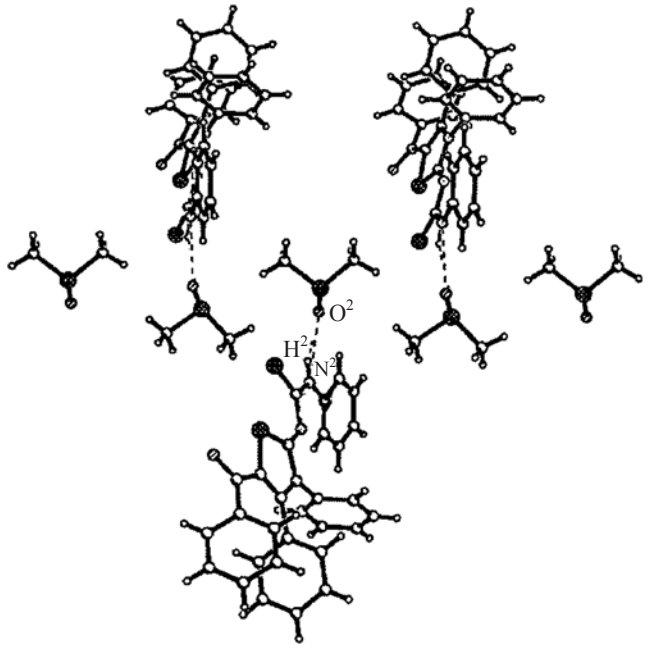


Fig. 3. Shortened contacts $O^2\cdots N^2$ and $O^2\cdots H^2–N^2$ between *N*-[5-benzoyl-3,4-diphenyl-1,3-thiazol-2(3*H*)-ylidene]-*N*′-phenylthiourea (**IIIb**) and DMSO molecules in crystal.

thiourea (**IIIb**) (Fig. 1) is approximately planar: the maximal deviation of non-hydrogen atoms from the mean-square plane is 0.23 Å (O^1). The dihedral angles formed by the (*i*) plane, on the one hand, and benzene ring planes (*ii*), (*iii*), (*iv*), and (*v*), on the other, are 159.4, 91.7, 113.5, and 124.3°, respectively; the dihedral angles between the (*ii*)–(*iii*), (*ii*)–(*iv*), (*ii*)–(*v*), (*iii*)–(*iv*), (*iii*)–(*v*), and (*iv*)–(*v*) planes are, respectively, 103.3, 104.5, 108.2, 108.1, 94.0, and 155.5°.

The crystalline structure of compound **IIIb** includes channels accommodating DMSO molecules which form with molecules **IIIb** shortened intermolecular contacts $O^2\cdots N^2$ 2.832(6) and $O^2\cdots H^2$ 2.03(6) Å (the corresponding sums of the van der Waals radii are 3.1 and 2.7 Å [32]), the angle $O^2H^2N^2$ being 161(6)° (Figs. 2, 3).

Thus 1,3-dipolar cycloaddition of 3-phenylamino-5-phenylimino-1,2,4-dithiazole to 1-acyl-2-phenylacetylenes ensures one-step formation of a thiazole ring and side-chain thiourea fragment. This reaction can be regarded as a general synthetic route to new functionalized 1,3-thiazolylthioureas that are promising as precursors of novel medicines.

EXPERIMENTAL

The IR spectra were recorded in KBr on a Bruker IFS25 spectrometer. The ^1H and ^{13}C NMR spectra were measured on a Bruker-400 spectrometer (400.13 and 100.61 MHz for ^1H and ^{13}C, respectively) using DMSO-d_6 as solvent and hexamethyldisiloxane as internal reference. X-Ray analysis was performed at room temperature on a KM-4 Kuma Diffraction instrument (MoK_α irradiation, graphite monochromator, ω/2θ scanning).

***N*-Phenyl-3-phenylimino-3*H*-1,2,4-dithiazol-5-amine (I).** A solution of 5 mmol of molecular iodine in 10 ml of ethanol was slowly added to a solution of 1.43 g (5 mmol) of 1,5-diphenyl-2,4-dithiobiuret (prepared from *N*-phenylthiourea and phenyl isothiocyanate according to the procedure reported in [26]) in 100 ml of diethyl ether under vigorous stirring at 20°C. The precipitate was filtered off, washed with diethyl ether, and dried under reduced pressure. We thus obtained 1.69 g (82%) of *N*-phenyl-3-phenylimino-3*H*-1,2,4-dithiazol-5-amine hydroiodide as yellow crystals with mp 177°C (from ethanol). IR spectrum: ν 2870–2980 cm^{-1}, br (NH_2^+). ^1H NMR spectrum, δ, ppm: 7.31–7.60 m (10H, H_{arom}), 12.13 br.s (NH). ^{13}C NMR spectrum, δ_C, ppm: 121.69, 127.14, 129.63, 137.83 (C_{arom}); 177.10 (C^3, C^5). Found, %: C 40.92; H 2.73; I 31.08; N 10.06; S 15.80. $C_{14}H_{11}N_3S_2\cdot HI$. Calculated, %: C 40.68; H 2.93; I 30.71; N 10.17; S 15.52.

N-Phenyl-3-phenylimino-3*H*-1,2,4-dithiazol-5-amine hydroiodide, 0.83 g (2 mmol), was dispersed in 30 ml of acetonitrile, 15 ml of 12% aqueous ammonia was added, the mixture was stirred for 15 min, 60 ml of water was added, and the mixture was stirred for an additional 1 h. The precipitate was filtered off, washed with water, and dried under reduced pressure over calcium chloride. Yield 0.53 g (93%), yellow

amorphous substance, mp 170–172°C. IR spectrum: ν 3250 cm^{-1} (NH). ^1H NMR spectrum, δ, ppm: 7.15–7.66 m (10H, H_{arom}), 11.19 br.s (1H, NH). ^{13}C NMR spectrum, δ_C, ppm: 120.64, 124.86, 129.69, 139.63 (C_{arom}); 150.63, 168.99 (C^3, C^5). Found, %: C 58.90; H 3.84; N 14.86; S 22.73. $C_{14}H_{11}N_3S_2$. Calculated, %: C 58.92; H 3.88; N 14.72; S 22.47.

1-Acyl-2-phenylacetylenes IIa–IIc (*general procedure*). The corresponding acetylenic alcohols were prepared by the Iotsitch reaction. A solution of 30.6 g (0.3 mol) of phenylacetylene in 20 ml of anhydrous diethyl ether was added at 0–5°C to the Grignard compound prepared from 7.2 g (0.3 mol) of magnesium and 32.7 g (0.3 mol) of ethyl bromide in 250 ml of anhydrous diethyl ether. The mixture was heated for 4 h on a water bath and cooled to 0–5°C, a solution of 0.3 mol of the corresponding aldehyde in 20 ml of anhydrous diethyl ether was added, and the mixture was stirred for 1 h and was left overnight. The mixture was treated with a saturated solution of ammonium chloride and extracted with diethyl ether (500 ml). The extract was dried over $MgSO_4$ and filtered, and the filtrate was treated with 130.5 g (1.5 mol) of active manganese dioxide at 20°C over a period of 5–6 h. The precipitate of MnO_2 was filtered off and washed with diethyl ether (500 ml), and the solvent was distilled off under reduced pressure to isolate compounds **IIa**–**IIc**.

4-Phenylbut-3-yn-2-one (IIa). Yield 36.3 g (84%), bp 122–124°C (12 mm); published data [33]: bp 122–128°C (12 mm).

1,3-Diphenylprop-2-yn-1-one (IIb). Yield 50.7 g (82%), yellow crystals, mp 49–50°C; published data [33]: mp 49–50°C.

3-Phenyl-1-(2-thienyl)prop-2-yn-1-one (IIc). Yield 47.7 g (75%), yellow crystals, mp 63–64°C (from EtOH); published data [34]: mp 54°C.

Reaction of *N*-phenyl-3-phenylimino-3*H*-1,2,4-dithiazol-5-amine (I) with 1-acyl-2-phenylacetylenes IIa–IIc (*general procedure*). Acylacetylene **IIa**–**IIc**, 2 mmol, was dissolved in 20 ml of (*a*) toluene or (*b*) ethanol, 0.57 g (2 mmol) of compound **I** was added, and the mixture was stirred for 1 h at 78–80°C. The mixture was cooled, and the precipitate was filtered off, washed with diethyl ether, and dried under reduced pressure. We thus isolated compounds **IIIa**–**IIIc**; the products were highly pure, and no recrystallization was necessary to obtain analytically pure samples. The filtrate was concentrated to a volume of 5–6 ml and kept for 24 h at 0–5°C, and the precipitate was filtered off. Yield of compound **IV** 0.1–0.12 g (18–21%).

***N*-[5-Acetyl-3,4-diphenyl-1,3-thiazol-2(3*H*)-ylidene]-*N′*-phenylthiourea (IIIa).** Yield 0.50 g (58%) (*a*), 0.49 g (57%) (*b*); yellow finely crystalline powder, mp 219–220°C. IR spectrum, ν, cm^{-1}: 3220 (NH); 1650 (C=O); 1470, 1490, 1510, 1600 (C=C, C=N, δNH). ^1H NMR spectrum, δ, ppm: 1.91 (3H, Me), 6.81–7.47 m (15H, H_{arom}), 10.29 br.s (1H, NH). ^{13}C NMR spectrum, δ_C, ppm: 28.52 (Me); 120.05, 122.56, 127.50, 128.16, 128.82, 129.10, 129.23, 129.37, 130.27, 130.99, 137.45, 137.78, 139.78 (C^5, C_{arom}); 144.76 (C^4); 169.37 (C^2); 183.58 (C=S); 190.72 (C=O). Found, %: C 67.36; H 4.42; N 10.02; S 14.85. $C_{24}H_{19}N_3OS_2$. Calculated, %: C 67.11; H 4.46; N 9.78; S 14.93.

***N*-[5-Benzoyl-3,4-diphenyl-1,3-thiazol-2(3*H*)-ylidene]-*N′*-phenylthiourea (IIIb).** Yield 0.59 g (60%) (*a*), 0.58 g (59%) (*b*); yellow finely crystalline powder, mp 219–220°C. IR spectrum, ν, cm^{-1}: 3200 (NH); 1610 (C=O); 1460, 1480, 1510, 1590 (C=C, C=N, δNH). ^1H NMR spectrum, δ, ppm: 6.82–7.54 m (20H, H_{arom}), 10.44 br.s (1H, NH). ^{13}C NMR spectrum, δ_C, ppm: 120.22, 122.56, 127.50, 127.97, 128.16, 128.82, 129.10, 129.23, 129.37, 130.99, 132.22, 137.45, 137.78, 139.78 (C^5, C_{arom}); 144.76 (C^4); 169.82 (C^2); 183.24 (C=S); 189.34 (C=O). Found, %: C 70.69; H 4.23; N 8.83; S 13.32. $C_{29}H_{21}N_3OS_2$. Calculated, %: C 70.85; H 4.31; N 8.55; S 13.04.

X-Ray diffraction data for compound (IIIb). $C_{31}H_{27}N_3O_2S_3$, M 569.74, monoclinic crystals, space group $P2_1/n$; unit cell parameters: a = 16.458(3), b = 9.932(2), c = 18.468(4) Å; β = 101.53(3)°; V = 2957.9(1) $Å^3$; Z = 4; d_{calc} = 1.28 g/cm^3; μ = 0.283 mm^{-1}. Total of 5467 reflections were measured, 4376 of which were independent; 367 refined parameters; R = 0.061 for 2090 reflections with $F_0 > 4\sigma(F_0)$. The structure was solved by the direct method with subsequent Fourier syntheses using SHELXS-97 software [35]. The structure was refined by the least-squares procedure in full-matrix anisotropic approximation for all non-hydrogen atoms using SHELXL-97 program [36]. The positions of hydrogen atoms were calculated on the basis of geometry considerations; the H^2 atom was localized experimentally. The sulfur atom in the DMSO molecule was disordered by two positions with populations of 0.86 and 0.14. The complete set of crystallographic data (coordinates of atoms and tables of bond lengths and bond angles) was deposited to the Cambridge Crystallographic Data Center (entry no. CCDC 684937).

***N*-[3,4-Diphenyl-5-(2-thienyl)-1,3-thiazol-2(3*H*)-ylidene]-*N′*-phenylthiourea (IIIc).** Yield 0.57 g

(57%) (*a*), 0.59 g (59%) (*b*); yellow finely crystalline powder, mp 209–210°C. IR spectrum, ν, cm^{-1}: 3200 (NH); 1605 (C=O); 1480, 1500, 1510, 1575 (C=C, C=N, δNH). ^1H NMR spectrum, δ, ppm: 6.79–7.88 m (18H, H_{arom}), 10.35 br.s (1H, NH). ^{13}C NMR spectrum, δ_C, ppm: 119.13, 120.64, 122.94, 128.22, 128.57, 129.53, 129.78, 131.19, 136.01, 136.08, 138.25, 140.23, 143.09 (C^5, C_{arom}); 143.65 (C^4); 169.82 (C^2); 180.33 (C=O); 183.44 (C=S). Found, %: C 65.24; H 4.01; N 8.75; S 19.07. $C_{27}H_{19}N_3OS_3$. Calculated, %: C 65.16; H 3.85; N 8.44; S 19.33.

***N*-(1,3-Benzothiazol-2-yl)-*N′*-phenylthiourea (IV).** Light red crystals, mp 198–200°C; published data [37]: mp 201°C. IR spectrum, ν, cm^{-1}: 3220, 3150 (NH); 1490, 1505, 1560, 1580 (C=C, C=N, δNH). ^1H NMR spectrum, δ, ppm: 7.09–7.82 m (9H, H_{arom}), 10.82 s (1H, NH), 12.78 s (1H, PhN**H**). ^{13}C NMR spectrum, δ_C, ppm: 115.31, 118.92, 122.44, 123.01, 123.58, 124.43, 126.76, 128.57, 129.06, 139.59 (C_{arom}); 166.34 (C^2); 182.61 (C=S). Found, %: C 58.67; H 3.90; N 14.53; S 22.67. $C_{14}H_{11}N_3S_2$. Calculated, %: C 58.92; H 3.88; N 14.72; S 22.47.

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