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## LETTERS TO THE EDITOR

# Reaction of *tert*-Butylimine of Phenylpropiolic Aldehyde with Substituted Imidazole-2-thiones

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It has been shown earlier that the interaction of *N*-*tert*-butyl-1-aza-1,3-enes with substituted benzimidazole-2-thiols leads to the formation of polynuclear sulfur-containing heterocyclic compounds, viz., benzo[4,5]imidazole[2,1-*b*][1,3]thiazin-4-ols [1, 2].

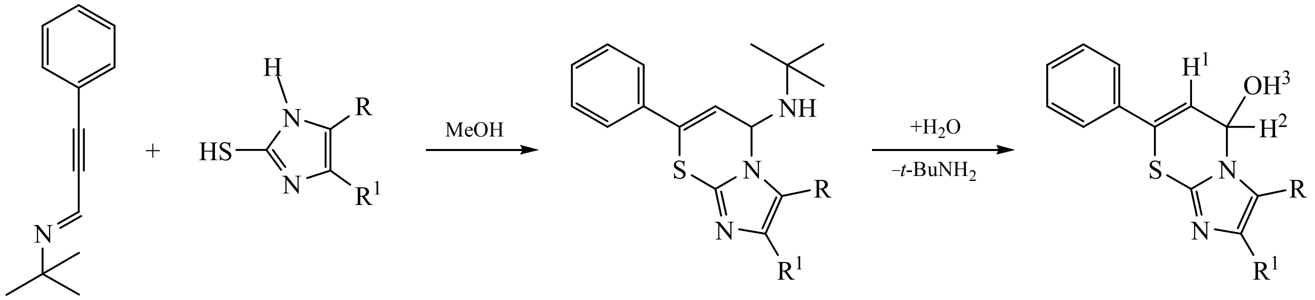
As a continuation of this research in the present work data are reported on the reaction of structural analogs of benzimidazole-2-thiols, viz., substituted imidazole-2-thiones, with phenylpropiolic aldehyde *tert*-butylimine (Scheme 1).

The reaction proceeds under stirring at 60°C for 2 h. Target compounds **IVa–IVc** were isolated with yields of 50–70% as crystal compounds melting with the decomposition. Their structure was confirmed by the data of $^1$H and $^{13}$C NMR spectroscopy. Thus, in the $^1$H NMR spectrum (**IVa**) the doublet at 6.26 ppm ($J$ = 4.9 Hz) corresponds to the proton H$^1$. The signal H$^2$ of the thiazine ring appears as a doublet at 6.66 ppm ($J$ = 8.8 Hz). A doublet of doublets at 6.16 ppm ($J$ = 8.8, 4.9 Hz) corresponds to the proton H$^3$ of the hydroxy group. Protons of two methyl groups and protons of aromatic rings appear in the typical regions.

As in case of substituted benzimidazole-2-thiols [1, 2] the initially formed imidazo[2,1-*b*][1,3]thiazine-5-*tert*-butylamines (**IIIa–IIIc**) undergo hydrolysis with the formation of imidazo[2,1-*b*][1,3]thiazin-5-ols (**IVa–IVc**). The hydrolytic cleavage of *tert*-bytylamine from imidazo[2,1-*b*]-[1,3]thiazine-5-*tert*-bytylamines (**IIIa–IIIc**) is due to the presence of minor quantities of water in the solvent [3].

As a result of the addition of 4-phenyl-2-imidazole-2-thione (**IIc**) to imine **I**, the formation of two

**Scheme 1.**



**I** **IIa–IIc** **IIIa–IIIc** **IVa–IVc**

R = R$^1$ = Me (**a**); R = R$^1$ = Ph (**b**); R = H, R$^1$ = Ph (**c**).

regioisomers is possible, which is due to the position of the phenyl group in the original imidazole-2-tione.

It has earlier been shown on the ground of DFT-calculations that in the reaction of 1-aza-1, 3-enynes with adenine-8-tiol a single regioisomer forms from two possible: the one whose formation in the course of the reaction meets minimal steric hindrances [4]. Because of that, the structures of 2,7-diphenylimidazo-[2,1-*b*][1,3]tiazine-5-*tert*-butylamine and 2,7-diphenyl-5*H*-imidazo[2,1-*b*][2,3]thiazin-5-ol, respectively, should be attributed to compounds **IIIc** and **IVc**.

**2,3-Dimethyl-7-phenyl-5*H*-imidazo[2,1-*b*][1,3]-thiazin-5-ol (IVa).** 4,5-Dimethylimidazole-2-thione (0.005 mol) was added to the solution of 5 mmol of *tert*-butylimine of phenylpropiolic aldehyde in 25 mL of methanol at stirring. In 30 min the reaction mixture was heated to 60°C and water (3 mL) was added. After cooling the residue was filtered off and recrystallized from aqueous methanol. The yield is 70%, colorless crystals, mp 215°C (dec.). $^1$H NMR spectrum (400 MHz, DMSO-$d_6$), δ, ppm: 7.57 d (2H, *J* 6.3 Hz), 7.51–7.32 m (3H), 6.66 d (1H, *J* 8.8 Hz), 6.26 d (1H, *J* 4.9 Hz), 6.16 d.d (1H, *J* 8.8, 4.9 Hz), 2.27 s (3H), 2.09 (3H). $^{13}$C NMR spectrum (101 MHz, DMSO-$d_6$), $\delta_C$, ppm: 136.84, 133.97, 132.44, 131.35, 129.94, 129.53, 126.53, 122.80, 116.17, 73.31, 12.82, 8.73. Found, %: C 64.8; H 5.9; N 10.3. $C_{14}H_{14}N_2OS$. Calculated, %: C 65.1; H 5.5; N 10.8; O 6.2; S 12.4.

**2,3,7-Triphenyl-5*H*-imidazolo[2,1-*b*][1,3]thiazin-5-ol (IVb)** was prepared analogously. Yield 65%, mp 228°C. The $^1$H NMR spectrum (400 MHz, DMSO-$d_6$), δ, ppm: 7.68–7.59 m (2H), 7.58–7.44 m (8H), 7.39 d (2H, *J* 7.3 Hz), 7.20 d.t (3H, *J* 24.2, 7.1 Hz), 6.81 d (1H, *J* 9.0 Hz), 6.49 d (1H, *J* 5.4 Hz), 6.04 d.d (1H, *J* 8.9, 5.5 Hz). The $^{13}$C NMR spectrum (101 MHz, DMSO-$d_6$), $\delta_C$, ppm: 138.36, 136.46, 131.85, 131.48, 130.15, 129.62, 129.37, 128.64, 128.26, 127.12, 126.75, 116.87, 73.20.Found, %: C 74.9; H 5.0; N 7.5. $C_{24}H_{18}N_2OS$. Calculated, %: C 75.4; H 4.7; N 7.3; O 4.2; S 8.4.

**2,7-Diphenyl-5*H*-imidazolo[2,1-*b*][1,3]thiazin-5-ol (IVc)** was prepared analogously. The yield 50%, mp 195°C (dec.). The $^1$H NMR spectrum (400 MHz, DMSO-$d_6$), δ, ppm: 7.94 s (1H), 7.80 d (2H, *J* 7.2 Hz), 7.70–7.59 m (2H), 7.57–7.45 m (3H), 7.39 t (2H, *J* 7.7 Hz), 7.25 d.d (2H, *J* 11.6, 5.5 Hz), 6.44 d.t (2H, *J* 13.1, 4.3 Hz). The $^{13}$C NMR spectrum (101 MHz, DMSO-$d_6$), $\delta_C$, ppm: 130.14, 129.64, 129.08, 127.29, 126.63, 124.81, 116.80, 115.63, 75.83. Found, %: C 70.0; H 4.9; N 9.3. $C_{18}H_{14}N_2OS$. Calculated, %: C 70.6; H 4.6; N 9.1; O 5.2; S 10.5.

$^1$H NMR spectra were registered on a spectrometer Bruker 400 [400 ($^1$H), 100 MHz ($^{13}$C)], solvent DMSO-$d_6$. The elemental analysis was carried out using the CHN-analyzer Perkin-Elmer 2400. Melting points were determined on the Koeffler heating block.

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