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SHORT COMMUNICATIONS

Synthesis of *N*,*N*-Dimethyl(diethyl)-7-aryl-5-methyl-4,7-dihydrotetrazolo[1,5-*a*]pyrimidine-6-carboxamides

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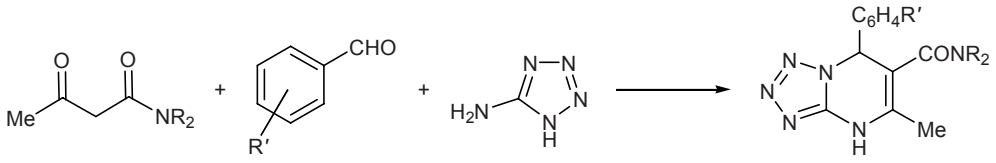
It is known that *N*,*N*-dimethyl-3-oxobutanamide reacts with aromatic aldehydes in the presence of piperidine to give *N*,*N*,*N*′,*N*′,6-pentamethyl-2-aryl-6-hydroxy-4-oxocyclohexane-1,3-dicarboxamides [1]. Zhidovinova et al. [2] described three-component condensation of ethyl 3-oxobutanoate with aromatic aldehydes and 5-aminotetrazole, which led to the formation of dihydrotetrazolopyrimidines [2].

While continuing studies in this line, we examined for the first time three-component condensation of aromatic aldehydes with 5-aminotetrazole and such substrates as *N*,*N*-dimethyl- and *N*,*N*-diethyl-3-oxobutanamides with a view to obtain new fused heterocyclic systems. The reactions occurred at 120–150°C in 20–60 min, and the products were the corresponding *N*,*N*-dimethyl- and *N*,*N*-diethyl-7-aryl-5-methyl-4,7-dihydrotetrazolo[1,5-*a*]pyrimidine-6-carboxamides **Ia**, **Ib**, and **IIa**–**IId**. These compounds were isolated as colorless crystalline substances which are soluble in chloroform, DMF, and DMSO and insoluble in water.

The IR spectra of **Ia**, **Ib**, and **IIa**–**IId** contained absorption bands due to stretching vibrations of the amide carbonyl group (1660–1680 cm^{-1}), NH group (3150–3200 cm^{-1}), and C=C bonds (1600–1620 cm^{-1}). In the ^1H NMR spectra of **Ia**, **Ib**, and **IIa**–**IId**, protons in the 5-methyl group resonated as a singlet at δ 1.80–1.90 ppm, the 7-H proton gave a singlet at δ 6.30–6.65 ppm, and signal from the NH proton appeared as δ 10.30–10.50 ppm; also signals from aromatic protons were present. Signals from protons in the *N*-alkyl group were located at δ 2.70 ppm (s, CH_3; **Ia**, **Ib**) or at δ 0.75-0.90 (m, CH_3) and 3.05–3.10 ppm (m, CH_2; **IIa**–**IId**). Compound **Ib** displayed in the mass spectrum $[M - CO]^+$ ion peak with *m*/*z* 301 and peaks from fragment ions $[M - CONMe_2]^+$ (*m*/*z* 260), $[M - NMe_2]^+$ (*m*/*z* 285), and $[Ph]^+$ (*m*/*z* 77) in support of the assumed structure.

***N*,*N*,5-Trimethyl-7-phenyl-4,7-dihydrotetrazolo[1,5-*a*]pyrimidine-6-carboxamide (Ia).** A mixture of 0.01 mol of *N*,*N*-dimethyl-3-oxobutanamide, 0.01 mol of benzaldehyde, and 0.01 mol of 1*H*-tetrazol-5-amine was heated at 120–150°C for 20–60 min (until gaseous products no longer evolved and the mixture solidified). The melt was cooled and treated with ethanol, and the precipitate was filtered off and recrystallized from ethanol. Yield 0.59 g (21%), mp 230–232°C. IR spectrum, ν, cm^{-1}: 1600 (C=C), 1675 (C=O), 3200 (NH). ^1H NMR spectrum, δ, ppm: 1.90 s (3H, 5-CH_3), 2.70 s (6H, NCH_3), 6.80 s (1H, 7-H), 8.14 m (5H, C_6H_5), 10.50 s (1H, NH). Found, %: C 59.08, 59.21; H 5.62, 5.73; N 29.48, 29.62. $C_{14}H_{16}N_6O$. Calculated, %: C 59.14; H 5.67; N 29.56.



Ia, **Ib**, **IIa**–**IId**

I, R = Me, R′ = H (**a**), 3-O_2N (**b**); **II**, R = Et, R′ = H (**a**), 4-Cl (**b**), 3-F (**c**), 4-*i*-Pr (**d**).

Compounds **Ib** and **IIa**–**IId** were synthesized in a similar way.

***N*,*N*,5-Trimethyl-7-(3-nitrophenyl)-4,7-dihydrotetrazolo[1,5-*a*]pyrimidine-6-carboxamide (Ib).** Yield 0.99 g (30%), mp 237–238°C. IR spectrum, ν, cm^{-1}: 1612 (C=C), 1680 (C=O), 3200 (NH). ^{1}H NMR spectrum, δ, ppm: 1.85 s (3H, 5-CH_3), 2.70 s (6H, NCH_3), 6.64 s (1H, 7-H), 8.14 m (4H, C_6H_4), 10.57 s (1H, NH). Found, %: C 50.98, 51.19; H 4.52, 4.67; N 29.65, 29.85. $C_{14}H_{15}N_7O_3$. Calculated, %: C 51.06; H 4.59; N 29.77.

***N*,*N*-Diethyl-5-methyl-7-phenyl-4,7-dihydrotetrazolo[1,5-*a*]pyrimidine-6-carboxamide (IIa).** Yield 1.44 g (46%), mp 267–268°C. IR spectrum, ν, cm^{-1}: 1600 (C=C), 1675 (C=O), 3150 (NH). ^{1}H NMR spectrum, δ, ppm: 0.90 m (6H, C**H**$_3$CH$_2$), 1.86 s (3H, 5-CH_3), 3.05 m (4H, NCH_2), 6.43 s (1H, 7-H), 7.35 m (5H, C_6H_5), 10.50 s (1H, NH). Found, %: C 61.42, 61.56; H 6.38, 6.48; N 26.85, 26.92. $C_{16}H_{20}N_6O$. Calculated, %: C 61.52; H 6.45; N 26.90.

7-(4-Chlorophenyl)-*N*,*N*-diethyl-5-methyl-4,7-dihydrotetrazolo[1,5-*a*]pyrimidine-6-carboxamide (IIb). Yield 1.25 g (36%), mp 244–246°C. IR spectrum, ν, cm^{-1}: 1600 (C=C), 1660 (C=O), 3150 (NH). ^{1}H NMR spectrum, δ, ppm: 0.75 m (6H, C**H**$_3$CH$_2$), 1.80 s (3H, 5-CH_3), 3.04 m (4H, NCH_2), 6.30 s (1H, 7-H), 7.00 m (4H, C_6H_4), 10.30 s (1H, NH). Found, %: C 55.35, 55.54; H 5.43, 5.56; N 24.15, 24.28. $C_{16}H_{19}ClN_6O$. Calculated, %: C 55.41; H 5.52; N 24.23.

***N*,*N*-Diethyl-7-(3-fluorophenyl)-5-methyl-4,7-dihydrotetrazolo[1,5-*a*]pyrimidine-6-carboxamide (IIc).** Yield 1.39 g (42%), mp 224–226°C. IR spectrum, ν, cm^{-1}: 1600 (C=C), 1675 (C=O), 3200 (NH). ^{1}H NMR spectrum, δ, ppm: 0.80 m (6H, C**H**$_3$CH$_2$), 1.80 s (3H, 5-CH_3), 3.03 m (4H, NCH_2), 6.37 s (1H, 7-H), 7.38 m (4H, C_6H_4), 10.35 s (1H, NH). Found, %: C 58.07, 58.29; H 5.75, 5.91; N 25.33, 25.56. $C_{16}H_{19}FN_6O$. Calculated, %: C 58.17; H 5.80; N 25.44.

***N*,*N*-Diethyl-7-(4-isopropylphenyl)-5-methyl-4,7-dihydrotetrazolo[1,5-*a*]pyrimidine-6-carboxamide (IId).** Yield 1.21 g (34%), mp 217–219°C. IR spectrum, ν, cm^{-1}: 1600 (C=C), 1680 (C=O), 3200 (NH). ^{1}H NMR spectrum, δ, ppm: 0.90 m (6H, C**H**$_3$CH$_2$), 1.17 d [6H, (C**H**$_3$)$_2$CH], 1.85 s (3H, 5-CH_3), 3.04 m (4H, NCH_2), 3.04 s [1H, (CH$_3$)$_2$C**H**], 6.45 s (1H, 7-H), 6.97 d and 7.08 d (2H each, C_6H_4, J = 7 Hz), 10.31 s (1H, NH). Found, %: C 64.32, 64.45; H 7.28, 7.48; N 23.63, 23.75. $C_{19}H_{26}N_6O$. Calculated, %: C 64.38; H 7.39; N 23.71.

The IR spectra were recorded on a Specord M-80 spectrometer from samples dispersed in mineral oil. The ^{1}H NMR spectra were measured from solutions in DMSO-d_6 on a Bruker 500 instrument (500.13 MHz) using tetramethylsilane as internal reference. The mass spectrum (electron impact, 70 eV) was obtained on a Finnigan MAT INCOS-50 mass spectrometer.

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