ISSN 1070-4280, Russian Journal of Organic Chemistry, 2010, Vol. 46, No. 12, p. 1891. © Pleiades Publishing, Ltd., 2010.
Original Russian Text © N.V. Bubnov, E.S. Denislamova, Z.G. Aliev, A.N. Maslivets, 2010, published in Zhurnal Organicheskoi Khimii, 2010, Vol. 46, No. 12, p. 1876.

SHORT COMMUNICATIONS

Spiro Heterocyclization of Methyl 3-Aroyl-1-aryl-4,5-dioxo-4,5-dihydro-1*H*-pyrrole-2-carboxylates by the Action of Diphenylguanidine

N. V. Bubnov[a], E. S. Denislamova[a], Z. G. Aliev[b], and A. N. Maslivets[a]

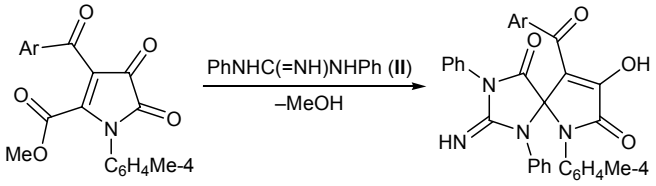
[a] *Perm State University, ul. Bukireva 15, Perm, 614990 Russia*
e-mail: koh2@psu.ru

[b] *Institute of Chemical Physics Problems, Russian Academy of Sciences, Chernogolovka, Moscow oblast, Russia*

Received January 4, 2010

DOI: 10.1134/S1070428010120201

Reactions of monocyclic 1*H*-pyrrole-2,3-diones with guanidines were not reported. We examined reactions of methyl 1-aryl-3-aroyl-4,5-dioxo-4,5-dihydro-1*H*-pyrrole-2-carboxylates **Ia** and **Ib** with 1,3-diphenylguanidine (**II**) at a molar ratio of 1:1 in boiling anhydrous 1,2-dichloroethane (reaction time 1–2 h; TLC monitoring) and isolated 6-aryl-9-aroyl-8-hydroxy-2-imino-1,3-diphenyl-1,3,6-triazaspiro[4.4]non-8-ene-4,7-diones **IIIa** and **IIIb** whose structure was confirmed by X-ray analysis. Presumably, compounds **IIIa** and **IIIb** are formed via addition of one secondary amino group in diphenylguanidine **II** at the C^2 atom of pyrroledione **Ia** or **Ib**, followed by closure of imidazole ring as a result of intramolecular attack by the second secondary amino group in **II** on the ester carbonyl group and elimination of methanol molecule.



Ia, **Ib** **IIIa**, **IIIb**

Ar = Ph (**a**), 4-$EtOC_6H_4$ (**b**).

The described reaction may be regarded as a novel method for building up polyfunctionalized spiro-fused heterocyclic 1,3,6-triazaspiro[4.4]nonane system.

9-Benzoyl-8-hydroxy-2-imino-6-(4-methylphenyl)-1,3-diphenyl-1,3,6-triazaspiro[4.4]non-8-ene-4,7-dione (IIIa). A solution of 1 mmol of compound **Ia** and 1 mmol of diphenylguanidine **II** in 10 ml of anhydrous 1,2-di-chloroethane was heated for 1 h under reflux. The mixture was cooled, and the precipitate was filtered off. Yield 79%, mp 243–244°C (from 1,2-dichloroethane). IR spectrum, ν, cm^{-1}: 3070 br (OH, NH), 1790 (C^4=O), 1730, 1700 (C^7=O), 1615 br (9-C=O). ^{1}H NMR spectrum, δ, ppm: 2.41 s (3H, Me), 7.10–7.69 m (19H, H_{arom}), 9.44 br.s (2H, NH, OH). Found, %: C 72.73; H 4.50; N 10.67. $C_{32}H_{24}N_4O_4$. Calculated, %: C 72.72; H 4.58; N 10.60.

9-(4-Ethoxybenzoyl)-8-hydroxy-2-imino-6-(4-methylphenyl)-1,3-diphenyl-1,3,6-triazaspiro-[4.4]non-8-ene-4,7-dione (IIIb) was synthesized in a similar way. Yield 75%, mp 251–252°C (from 1,2-dichloroethane). IR spectrum, ν, cm^{-1}: 3050 br (OH, NH), 1790 (C^4=O), 1730, 1700 (C^7=O), 1610 br (9-C=O). ^{1}H NMR spectrum, δ, ppm: 1.35 t (3H, CH_3CH_2, J = 7.0 Hz), 2.40 s (3H, Me), 4.08 q (2H, CH_2O, J = 7.0 Hz), 6.85–7.81 m (18H, H_{arom}), 9.28 br.s (2H, NH, OH). Found, %: C 71.38; H 4.88; N 9.70. $C_{34}H_{28}N_4O_5$. Calculated, %: C 71.32; H 4.93; N 9.78.

The IR spectra were recorded on an FMS-1201 spectrometer from samples dispersed in mineral oil. The ^{1}H NMR spectra were measured on a Bruker WP-400 instrument from solutions in DMSO-d_6 using tetramethylsilane as internal reference. The purity of the products was checked by TLC on Silufol plates using ethyl acetate as eluent; spots were visualized by treatment with iodine vapor.

This study was performed under financial support by the Russian Foundation for Basic Research (project no. 07-03-96036).