= SHORT COMMUNICATIONS

Spiro Heterocyclization of 4,5-Diaroyl-1*H*-pyrrole-2,3-diones with Heterocyclic Enamine

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Received February 16, 2012

DOI: 10.1134/S1070428013060298

We previously described recyclization of 4,5-diaroyl-1*H*-pyrrole-2,3-diones by the action of a difunctional 1,3-N,N-binucleophile (3-amino-4,6-dimethyl-2*H*-pyrazolo[3,4-*b*]pyridine) with formation of substituted pyrido[2',3':3,4]pyrazolo[1,5-*a*]pyrimidine [1]. There are no published data on reactions of 4,5-diaroyl-1*H*-pyrrole-2,3-diones with heterocyclic amines capable of acting as 1,3-C,N-binucleophiles.

In continuation of our studies on nucleophilic recyclizations [1–4] and spiro heterocyclizations [5] of 4,5-diaroyl-1*H*-pyrrole-2,3-diones [6], we now report on their reaction with 6-amino-1,3-dimethylpyrimidine-2,4(1H,3H)-dione (I). By heating a solution of equimolar amounts of 1-aryl-4,5-bis(4-methylbenzoyl)-1*H*-pyrrole-2,3-dione **IIa** or **IIb** and enamine **I** in boiling anhydrous 1,2-dichloroethane for 3-4 h (TLC) we obtained 1-aryl-4-hydroxy-3-(4-methylbenzoyl)-6'-(4-methylphenyl)-1',3'-dimethylspiro[pyrrole-2,5'-pyrrolo[2,3-d]pyrimidine]-2',4',5(1H,1'H,3'H)-triones IIIa and IIIb. Compounds IIIa and IIIb are likely to be formed via initial addition of the enamine β -CH group in molecule I to C⁵ of 1*H*-pyrrole-2,3dione II, followed by intramolecular closure of new 3*H*-pyrrole ring as a result of nucleophilic attack by the primary amino group on the carbonyl carbon atom

in the aroyl substituent on C^5 in the dioxopyrrole fragment.

This reaction may be regarded as the first example of direct spiro heterocyclization of substituted 4,5-diaroyl-1*H*-pyrrole-2,3-diones with heterocyclic enamine, leading to difficultly accessible spiro[pyrrole-2,5'-pyrrolo[2,3-*d*]pyrimidine] system.

4-Hydroxy-1',3'-dimethyl-3-(4-methylbenzoyl)-1,6'-bis(4-methylphenyl)spiro[pyrrole-2,5'-pyrrolo-[2,3-d]pyrimidine]-2',4',5(1H,1'H,3'H)-trione (IIIa). A solution of 1.0 mmol of compound I and 1.0 mmol of pyrroledione IIa in 80 ml of anhydrous 1,2-dichloroethane was heated for 3 h under reflux. The solvent was distilled off, 20 ml of ethyl acetate was added to the residue, the mixture was heated to the boiling point, and the undissolved material was filtered off. The filtrate was evaporated, and the residue was recrystallized from toluene. Yield 55%, mp 263-264°C. IR spectrum, v, cm⁻¹: 3180 (OH), 1721 (C²'=O), 1707 (C⁴'=O), 1647 (C⁵=O, 3-C=O). ¹H NMR spectrum, δ, ppm: 2.20 s (3H, Me), 2.34 s (3H, Me), 2.40 s (3H, Me), 3.13 s (3H, Me), 3.50 s (3H, Me), 7.09-7.98 m (12H, H_{arom}), 12.75 s (1H, OH). Found, %: C 70.72; H 5.07; N 9.96. C₃₃H₂₈N₄O₅. Calculated, %: C 70.70; H 5.03; N 9.99.



4-Hydroxy-1',3'-dimethyl-6'-(4-methoxyphenyl)-3-(4-methylbenzoyl)-1-(4-methylphenyl)spiro-[pvrrole-2,5'-pvrrolo[2,3-d]pvrimidine]-2',4',5(1H,1'H,3'H)-trione (IIIb) was synthesized in a similar way. Yield 58%, mp 250-251°C (from toluene). IR spectrum, v, cm⁻¹: 3195 (OH), 1725 (C²=O), 1719 (C^{4'=}O), 1651 (C⁵=O), 1635 (3-C=O). ¹H NMR spectrum, δ, ppm: 2.34 s (3H, Me), 2.40 s (3H, Me), 3.14 s (3H, Me), 3.50 s (3H, Me), 3.67 s (3H, OMe), 7.75–8.00 m (12H, H_{arom}), 12.73 s (1H, OH). ¹³C NMR spectrum, δ_C, ppm: 21.16 (Me), 21.40 (Me), 27.39 (Me), 30.73 (Me), 55.20 (OMe), 79.11 (C²), 96.28 (C^{4'a}), 114.61 (C³), 115.79–145.06 (C_{arom}), 151.70 $(C^{7'a})$, 156.92 $(C^{2'})$, 158.98 (C^{5}) , 161.19 $(C^{4'})$, 165.78 (C^{6'}), 184.69 (C⁴), 188.21 (5-C=O). Found, %: C 68.69; H 4.87; N 9.74. C₃₃H₂₈N₄O₆. Calculated, %: C 68.72; H 4.89; N 9.72.

The IR spectra were recorded on a Perkin Elmer Spectrum Two spectrometer from samples dispersed in mineral oil. The ¹H and ¹³C NMR spectra were measured on a Bruker AM-400 spectrometer at 400 and 100 MHz, respectively, using DMSO-*d*₆ as solvent and tetramethylsilane as internal reference. This study was performed under financial support by the Ministry of Education and Science of the Russian Federation, by the Ministry of Education of Perm Krai (International Research Teams Competition), and by the Russian Foundation for Basic Research (project nos. 12-03-31157, 12-03-00696).

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