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CYCLOCONDENSATION OF 2-(1-ARYL-5-PHENYLPYRAZOL-4-YL)-1H-BENZIMIDAZOLES WITH DMF DIMETHYLACETAL

I. B. Dzvinchuk and M. O. Lozinskii

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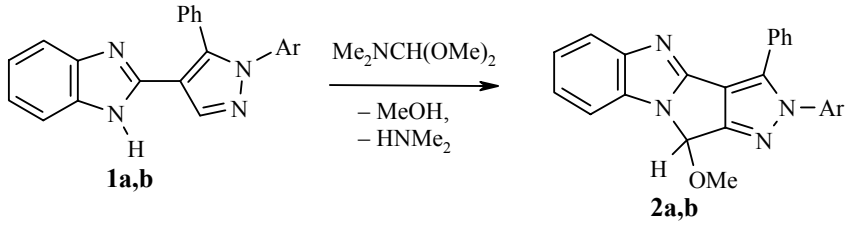
A number of compounds in the pyrazolo[4',3':3,4]pyrrolo[1,2-*a*]benzimidazole series have been recently synthesized by cyclocondensation of 2-cyanomethyl-1H-benzimidazole with hydrazonoyl chlorides of oxalic acid monoethyl ester [1]. We have found a route to novel derivatives of the same polyheterocyclic system, starting from the previously described (see [2]) 2-(1-aryl-5-phenylpyrazol-4-yl)-1H-benzimidazoles **1a,b**.



1a,b **2a,b**

1, **2 a** Ar = Ph, **b** Ar = 4-$O_2NC_6H_4$

Compounds **1a,b**, as we discovered, can react as 1,4-dinucleophiles containing electron-rich reaction centers on the nitrogen atom of the benzimidazole system and on the unsubstituted carbon atom of the pyrazole ring. Their reaction with DMF dimethylacetal occurs as a cyclocondensation with cleavage of one equivalent of methanol and also dimethylamine, and leads to tetracyclic methoxy-substituted compounds **2a,b**. We note that the known analogous reaction of DMF dimethylacetal with 1,2-diols occurs with retention of the dimethylamino group [3].

Compounds of type **2** contain a latent aldehyde group in their structure, and can be used for further chemical conversions.

10-Methoxy-2,3-diphenyl-2,10-dihydropyrazolo[4',3':3,4]pyrrolo[1,2-*a*]benzimidazole (2a). A mixture of compound **1a** (0.168 g, 0.5 mmol) and DMF dimethylacetal (0.6 g, 5 mmol) was heated for 7 h at 100-105°C. 2-Propanol (1 ml) was added and the mixture was heated with stirring until it started to boil, and then another water (1 ml) was added. After cooling, the precipitate was filtered out and washed with 2-propanol. Yield 0.155 g (82%). Colorless crystals, mp 190-191.5°C (1:2 pyridine–water). ^{1}H NMR spectrum (300 MHz, DMSO-d_6, TMS), δ, ppm: 3.43 (3H, s, OCH_3); 7.18-7.42 (12H, m, H-6,7 + CC_6H_5 + NC_6H_5); 7.47-7.50 (1H, m, H-8); 7.60-7.63 (1H, m, H-5); 8.20 (1H, s, H-10). Found, %: C 76.08; H 4.67; N 14.72. $C_{24}H_{18}N_4O$. Calculated, %: C 76.17; H 4.79; N 14.80.

Institute of Organic Chemistry, National Academy of Sciences of Ukraine, Kiev 02094; e-mail: iochkiev@ukrpack.net. Translated from Khimiya Geterotsiklicheskikh Soedinenii, No. 9, pp. 1404-1405, September, 2005. Original article submitted March 5, 2005.

10-Methoxy-2-(4-nitrophenyl)-3-phenyl-2,10-dihydropyrazolo[4',3':3,4]pyrrolo[1,2-*a*]benzimidazole (2b) was obtained similarly from compound **1b** (0.190 g, 0.5 mmol). Yield 0.171 g (81%). Yellowish crystals, mp 206-207.5°C (1:2 pyridine–water). ^{1}H NMR spectrum (300 MHz, DMSO-d_6, TMS), δ, ppm (*J*, Hz): 3.46 (3H, s, OCH_3); 7.19-7.43 (7H, m, H-6,7 + C_6H_5); 7.49-7.52 (1H, m, H-8); 7.57 and 7.28 (2 + 2H, two d, *J* = 9, $C_6H_4NO_2$); 7.61-7.64 (1H, m, H-5); 8.34 (1H, s, H-10). Found, %: C 68.13; H 4.07; N 16.46. $C_{24}H_{17}N_5O_3$. Calculated, %: C 68.08; H 4.05; N 16.54.

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