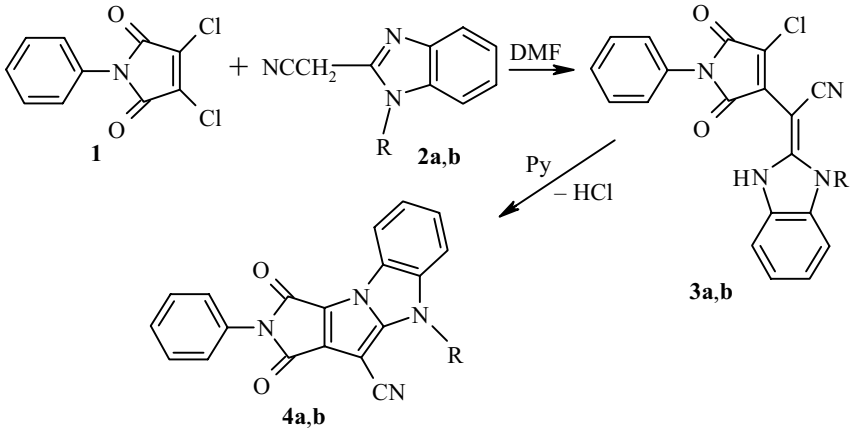
Chemistry of Heterocyclic Compounds, Vol. 37, No. 1, 2001

SYNTHESIS OF A NEW HETEROCYCLIC SYSTEM, 1,3-DIOXO-1,3-DIHYDROPYRROLO-[3',4':4,5]PYRROLO[1,2-*a*]BENZIMIDAZOLE

Yu. M. Volovenko and G. G. Dubinina

Keywords: 1,3-dioxo-1,3-dihydropyrrolo[3',4':4,5]pyrrolo[1,2-*a*]benzimidazole.

The reaction of dichloromaleimide **1** with active methylene compounds, in particular malononitrile and cyanoacetic ester, leads to arylation of the methylene group [1, 2]. We have established that when dichloromaleimide reacts with 2-benzimidazolylacetonitriles **2a,b** nucleophilic substitution of a chlorine atom occurs to give compounds **3a**,**b**, boiling of which in pyridine leads to intramolecular cyclization, the products of which are compounds **4a**,**b**, derivatives of a new heterocyclic system: 1,3-dioxo-1,3-dihydropyrrolo[3',4':4,5]pyrrolo[1,2-*a*]benzimidazole.



2a, 3a, 4a R = H; **2b, 3b, 4b** R = CH_3

2-(4-Chloro-2,5-dioxo-1-phenyl-2,5-dihydro-3-pyrrolyl)-2-(2,3-dihydrobenzimidazol-2-ylidene)-acetonitrile (3a); 2-(4-chloro-2,5-dioxo-1-phenyl-2,5-dihydro-3-pyrrolyl)-2-(1-methyl-2,3-dihydro-benzimidazol-2-ylidene)acetonitrile (3b). A solution of dichloromaleimide **1** (1.21 g, 5 mmol) and the corresponding 2-benzimidazolylacetonitrile **2a,b** (7.5 mmol) in DMF (15 ml) was stirred at room temperature for 2.5 h, the precipitated 2-benzylimidazolylacetonitrile hydrochloride was filtered off, water (150 ml) was added to the filtrate, and the resultant precipitate was filtered off. Compounds **3** were purified from starting imide **1** by boiling in chloroform. Yields calculated from maleimide **1** were 74% (**3a**) and 93% (**3b**).

Taras Shevchenko Kiev National University, Kiev 01033, Ukraine; e-mail: atver@mail.univ.kiev.ua.
Translated from Khimiya Geterotsiklicheskikh Soedinenii, No. 1, pp. 128-129, January, 2001. Original article submitted July 17, 2000.

 0009-3122/01/3701-0122$25.00

Compound 3a (R = H): mp 255-256°C (dec.). ^{1}H NMR spectrum (100 MHz, DMSO-d_6) δ, ppm, *J* (Hz): 7.2-7.6 (9H, m, Ph-H + Ar-H); 13.0 (2H, s, N-H). IR spectrum (KBr disks), ν, cm^{-1}: 2180 (CN), 1745, 1680 (C=O). Found, %: Cl 10.00; N 15.69. $C_{19}H_{11}ClN_4O_2$. Calculated, %: Cl 9.77; N 15.44.

Compound 3b (R = CH_3): mp 234-235°C (dec.). ^{1}H NMR spectrum (100 MHz, DMSO-d_6) δ, ppm, *J* (Hz): 3.84 (3H, s, CH_3); 7.35-7.50 (9H, m, Ph-H + Ar-H); 13.6 (1H, s, N-H). IR spectrum (KBr disks), ν, cm^{-1}: 2170 (CN), 1750, 1700 (C=O). Found, %: Cl 9.45; N 14.81. $C_{20}H_{13}ClN_4O_2$. Calculated, %: Cl 9.41; N 14.87.

1,3-Dioxo-2-phenyl-1,3-dihydropyrrolo[3',4':4,5]pyrrolo[1,2-*a*]benzimidazol-4-carbonitrile (4a); 5-methyl-1,3-dioxo-2-phenyl-1,3-dihydropyrrolo[3',4':4,5]pyrrolo[1,2-*a*]benzimidazol-4-carbonitrile (4b). The respective compounds **3a,b** (5 mmol) were boiled in dry pyridine (20 ml) for 2 h, the precipitate was filtered off, washed with water and recrystallized from pyridine. Yields 42% (**4a**) and 45% (**4b**).

Compound 4a (R = H): m.p. 276-277°C (dec., from pyridine). ^{1}H NMR spectrum (100 MHz, DMSO-d_6) δ, ppm, *J* (Hz): 7.3-7.7 (7H, m, Ph-H + 6,7-H); 8.1 (1H, t, 8-H); 8.75 (1H, d, *J* = 6.5, 9-H); 13.5 (1H, s, N-H). IR spectrum (KBr disks), ν, cm^{-1}: 2190 (CN), 1740, 1680 (C=O). Found, %: N 16.89. $C_{19}H_{10}N_4O_2$. Calculated, %: N 17.17.

Compound 4b (R = CH_3): mp 252-253°C (dec., pyridine). ^{1}H NMR spectrum (100 MHz, DMSO-d_6) δ, ppm, *J* (Hz): 3.88 (3H, s, CH_3); 7.35-7.5 (7H, m, Ph-H + 6,7-H); 8.0 (1H, t, 8-H); 8.5 (1H, d, *J* = 6.5, 9-H). IR spectrum (KBr disks), ν, cm^{-1}: 2180 (CN), 1740, 1685 (C=O). Found, %: N 16.26. $C_{20}H_{12}N_4O_2$. Calculated, %: N 16.46.

REFERENCES

1. K.-H. Etzbach and H. Eilingsfeld, *Synthesis*, **6**, 449 (1988).
2. M. Augustin, G. Fischer, B. Schneider, and M. Kohler, *J. Prakt. Chem.,* **321**, 787 (1979).