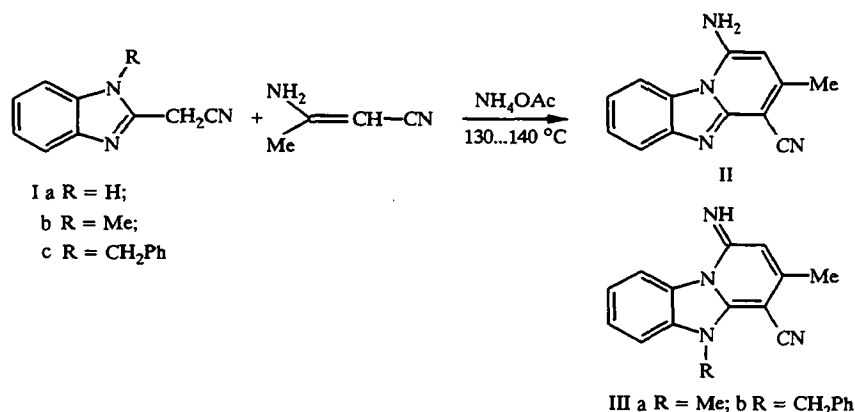


NEW, SIMPLE SYNTHESIS OF 1-AMINO(IMINO)-3-METHYL-PYRIDO[1,2-*a*]-BENZIMIDAZOLE-4-CARBONITRILES

Yu. M. Volovenko and V. V. Ivanov

1-Aminopyrido[1,2-*a*]benzimidazole-4-carbonitrile derivatives having fluorescent properties [1, 2] were investigated as potentially biologically active substances in [3]. Only one multiple stage synthesis of 1-amino-3-methylpyrido[1,2-*a*]benzimidazole-4-carbonitrile (II), with a total yield of approximately 58%, was described in [3].

We found that 2-cyanomethylbenzimidazole (Ia) and its N-substituted derivatives (Ib, c) react with β -aminocrotonic acid nitrile, forming 1-amino(imino)pyrido[1,2-*a*]benzimidazoles II, IIIa, b. The reaction takes place on heating (130-140°C) an equimolar mixture of the reagents for 0.5-1 h in the presence of ammonium acetate. The target compounds were isolated in pulverizing the reaction mixture with water.



The substances obtained had fluorescence which did not disappear on exposure to sunlight for many months.

1-Amino-3-methylpyrido[1,2-*a*]benzimidazole-4-carbonitrile (II). Yield of 60%, mp > 300°C. PMR spectrum (DMSO-*D*₆): 2.48 (3H, s, CH₃); 6.10 (1H, s, 2-CH); 7.2-7.6 (2H, m, 7-, 8-CH); 7.75 (1H, d, 6-CH); 7.77 (2H, br, NH₂); 8.42 ppm (1H, d, 9-CH). It should be noted that a different assignment (3.6 ppm) of the methyl group signal was made in [3]. IR spectrum (KBr pellets): 3470 m, 3300 w (NH₂), 2200 s (CN), 1648 cm⁻¹ s (C=N). Found, %: N 25.57. C₁₃H₁₀N₄. Calculated, %: N 25.21.

1-Imino-3,5-dimethylpyrido[1,2-*a*]benzimidazole-4-carbonitrile (IIIa). Yield of 64%. mp = 245-248°C. PMR spectrum (DMSO-*D*₆): 2.17 (3H, s, 3-CH₃); 3.99 (3H, s, 5-CH₃); 5.94 (1H, s, 2-CH); 7.3-7.7 (4H, m, NH + 6-, 7-, 8-CH); 8.93 ppm (1H, d, 9-CH). IR spectrum (KBr pellets): 3300 m (NH), 2175 s (CN), 1628 s, 1550 cm⁻¹ s. Found, %: N 23.93. C₁₄H₁₂N₄. Calculated, %: N 23.72.

1-Imino-5-benzyl-3-methylpyrido[1,2-*a*]benzimidazole-4-carbonitrile (IIIb). Yield of 47%. mp = 195-197°C. PMR spectrum (DMSO-*D*₆): 2.16 (3H, s, CH₃); 5.88 (2H, s, CH₂); 6.03 (1H, s, 2-CH); 7.3-7.7 (8H, m, C₆H₅ + 6-, 7-, 8-CH); 8.15 (1H, br, NH); 9.0 ppm (1H, d, 9-CH). IR spectrum (KBr pellets): 3317 w (NH), 2185 s (CN), 1630 s, 1543 cm⁻¹ s. Found, %: N 18.28. C₂₀N₁₆N₄. Calculated, %: N 17.93.

Taras Shevchenko Kiev National University, Kiev 252017. Translated from *Khimiya Geterotsiklicheskikh Soedinenii*, No. 9, pp. 1280-1281, September, 1997. Original article submitted August 28, 1997.

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